Thorium Research in the Manhattan Project Era

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Abstract

Research on thorium as an energy source began in 1940 under the direction of Glenn Seaborg at the University of California, Berkeley. Following the discovery of plutonium-239 and its fissile qualities, similar experiments demonstrated that uranium-233 bred from thorium was also fissile. Seaborg viewed uranium-233 as a potential backup to plutonium-239, whose production was one of the Manhattan Project's primary efforts. The central appeal of U-233 was that the chemistry of uranium was well understood, unlike plutonium, but plutonium-239 had the potential to be produced from natural uranium in a critical nuclear reactor. Natural thorium lacked fissile isotopes and so a critical nuclear reactor (to produce U-233) from thorium alone was not possible. Not until the X-10 graphite reactor was constructed at Oak Ridge in 1943 was sufficient U-233 created to conclusively assess its nuclear properties, which were found to be superior to Pu-239 in a thermal-spectrum reactor. Early production of plutonium at X-10 showed significant contamination by Pu-240, which made plutonium unsuitable for simple "gun-type" nuclear weapons. Researchers in the "Metallurgical Laboratory" at the University of Chicago, which included Seaborg's chemistry group, suggested that the plutonium produced be used as a fuel in a special reactor to convert thorium to uranium-233 for weapons. This effort encountered many severe difficulties in fuel fabrication and dissolution. Seaborg also recognized the severe issue that uranium-232 contamination would play in any effort to use uranium-233 for weapons. Through tremendous effort, weapons designers at Los Alamos were able to design workable weapons using the implosion principle, which accommodated for the impure plutonium produced. Interest in U-233 for weapons effectively disappeared by 1945, but the Metallurgical Laboratory continued to investigate the potential of a thorium-U-233 "breeder" reactor, based on a homogeneous mixture of uranium salts in heavy water. This effort also came to an end in early 1945. With the use of nuclear weapons on Japan in August 1945 and the end of World War II, many questions were asked about how nuclear reactors, nuclear power, and nuclear weapons would develop. Interest in the liquid-sodium-metal-cooled, fast-breeder reactor came to dominate, since it promised to increase the supply of rare fissile material rather than reduce it. Research on thorium and homogeneous reactors faded, but their involvement in early research led to later interest by Eugene Wigner, Glenn Seaborg, and Alvin Weinberg. The United States was fully focused on growing its nuclear weapons stockpile, and the success of the implosion weapon using plutonium, coupled with its natural advantage in production from unenriched uranium, led to limited interest in thorium as the Manhattan Project ended and the Atomic Energy Commission began in early 1947.

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Preface

Historians may one day lament that the discovery of nuclear fission in 1939 by Otto Hahn in Germany and the initiation of World War II by Hitler's Nazi regime took place in such close spatial and temporal proximity. Because of these proximities, the process of nuclear fission was immediately seen as a military threat against the United States by European refugee scientists, many of whom were Jewish or had Jewish relatives. They urged a national response that led to the development of nuclear weapons on an industrial scale without modern precedent. The discovery of fission, along with the advent of powerful particle accelerators in the United States, also made possible the discovery of new elements and new isotopes of existing elements. Their existence and potential was immediately viewed through the prism of war and destruction.

This paper is an attempt to understand the early history of thorium as a source of nuclear energy. In discussing thorium, I often refer to the isotope thorium-232 and its fissile daughter product uranium-233 as if they were one material. I realize that they are distinct and the manufacture of uranium-233 depends on neutron absorption in thorium from a source such as a nuclear reactor, but it is also true that thorium-232 is the only significant source of uranium-233, and that energy from uranium-233 is energy from thorium, abstracted by the step of neutron bombardment.

There have been scores of accounts written about the early history of the nuclear age, specifically the wartime program code-named the "Manhattan Project" which led to the development of nuclear weapons. These histories focus on the two strands of effort that led to successful nuclear weapons, namely the isotopic separation of U-235 from natural uranium and the creation of plutonium in dedicated production reactors fueled by natural uranium. One could easily wonder when the story of thorium as an energy source really began. The official history of the Atomic Energy Commission that covered this era, *The New World 1939-1946* [1], gives scant mention of thorium and its potential, and no details as to the discovery of uranium-233, its fissile nature, nor the potential of thorium as an energy source.

Other important works chronicling this time, including Richard Rhodes' Pulitzer-Prize-winning work *The Making of the Atomic Bomb*, also give little mention to thorium. This can easily be forgiven, for thorium and uranium-233 did not lead to the production of nuclear weapons, but the question of why they did not is a subject that has persistent historical relevance. Today we consider whether thorium is a natural energy source of sufficient magnitude and merit to power human civilization for many hundreds of thousands of years, and it is necessary to understand why decisions were made long ago, and what lessons those decisions could have for us today.

This thesis began originally as an attempt to tell the story of the thorium-fueled molten-salt reactor effort at Oak Ridge National Laboratory, strongly motivated by the discovery of a series of books [2] [3] [4] written by Glenn Seaborg and Benjamin Loeb covering Seaborg's years as the chairman of the US Atomic Energy Committee. In the last of these, *The Atomic Energy Commission Under Nixon* [4], Seaborg describes budget cuts early in the Nixon Administration that led the AEC to curtail research into breeder reactors. He describes how Alvin Weinberg, head of the Oak Ridge National Laboratory (ORNL) and ardent proponent of the thorium-fueled molten-salt breeder reactor (MSBR), argued for the continuation of that line of research. Seaborg goes on to describe how the MSBR program was cancelled in favor of the plutonium-fueled, liquid-metal

fast-breeder reactor (LMFBR) but concedes later that that may have been a mistake. [4, pg.179]

This significant admission led me to probe deeper into the earlier histories of both the fast-breeder and the molten-salt reactor. At the time of the molten-salt reactor's cancellation, the fast-breeder reactor effort was at a far greater stage of maturity. A commercial fast-breeder reactor called "Fermi-1" had been built near Detroit, Michigan, and had operated briefly in 1966 before suffering a partial core meltdown. [5] Fermi-1 had been built using the private funds of a large industrial consortium that included some of the most prominent utilities and nuclear suppliers of the time. In today's funds, it had cost hundreds of millions of dollars. The AEC had facilitated its construction and operation through fee waivers on nuclear fuel and a generous approach to siting and licensing. Several other smaller test reactors, such the Experimental Breeder Reactors -1 and -2 (EBR-1 and -2) and the Southwest Experimental Fast Oxide Reactor (SEFOR) had also been built and operated, with varying degrees of success. There was also a large international effort underway to develop the sodium-cooled fast breeder reactor.

In contrast, the effort to develop the molten-salt reactor was much smaller and almost entirely confined to ORNL. Why had there been so much more industrial and governmental interest in the fast breeder than the molten-salt reactor? That search took me further back in history, to the early 1950s when the Fermi project began as a commercial effort under the leadership of Walker Cisler of the Detroit Edison Company. Commercial nuclear reactors were being considered for the first time, and with the advent of Soviet nuclear testing and the Korean War, the Atomic Energy Commission was under great pressure to produce more and more fissile material for nuclear weapons. There were also stronger calls for the use of nuclear energy to produce electricity for civilian use. The technology decisions made for Fermi-1 were motivated in large part by the desire of the US Atomic Energy Commission to produce a high-quality form of plutonium, which was uniquely produced by fast breeder reactors. They were willing to incentivize industry to produce this material for them in civilian reactors. The AEC had a huge effort to produce highly-enriched uranium and high-isotopic-quality plutonium for nuclear weapons, and the Fermi project was seen as a contributor to that effort. The chairman of the AEC at that time, Gordon Dean, had no interest in thorium or uranium-233 as materials for a nuclear weapons effort.

Why was uranium/plutonium useful for weapons and thorium/U-233 wasn't? The search for that answer took me back to where this story begins, in the fervent atmosphere of the wartime program to develop nuclear weapons. It was in this era that discoveries were first made and the broad lines of reactor development were laid out.

In the search for referenceable descriptions of early research on thorium, I found to my disappointment that there was nearly no discussion in historical texts such as *The New World* of the early efforts on thorium and uranium-233. But to my delight, I discovered the wartime journals of Dr. Glenn Seaborg and the first-hand account they told of the discoveries surrounding thorium. Dr. Seaborg led a small research team that discovered uranium-233, and his curiosity about nuclear decay chains, along with his proximity to a powerful cyclotron at the University of California, Berkeley, led to this historic accomplishment. Seaborg's pre-war and wartime journals were primary sources that had never been "mined" along the direction of thorium and uranium-233, and the remarkable story of these materials merited a new perspective on the entire Manhattan Project.

Dr. Seaborg compiled his pre-war and wartime journals and records in the late 1970s in a series

of documents published by the University of California. The first, *Early History of Heavy Isotope Research at Berkeley* [6], covered a period from August 1940 to April 1942, when Seaborg left Berkeley to join the "Metallurgical Laboratory" at the University of Chicago. The Met Lab, as it was called, was a branch of the Manhattan Project whose divisions were charged with some of the greatest challenges of the Project, including the design of plutonium-producing chain-reacting "piles" and the chemical separation processes that would isolate the plutonium from the remaining fuel and fission products.

In the *Early History*, Seaborg describes the discovery of plutonium-239 and uranium-233 using the large 60-inch cyclotron at Berkeley. He also describes the processes by which their fissile natures were discovered through precision chemistry. Plutonium-239 was discovered about a year earlier than uranium-233, and this gave it an important head start in the effort to develop materials for the Manhattan Project.

Seaborg's later efforts at the Met Lab are chronicled in four volumes, titled *History of Met Lab Section C-I* [7] [8] [9] [10], each covering approximately a year of history. Events related to thorium and uranium-233 are mentioned tangentially in the first two volumes, but in volume 3 new data on the properties of U-233, which became available from the X-10 graphite reactor at Oak Ridge, led to extensive discussion about the potential of uranium-233 and the most concentrated efforts on its development. For a period it was even considered as a replacement for plutonium-239. But in this same period some of its persistent flaws as a weapons material were also uncovered.

The *Early History*, published in June 1976, is approximately 150 pages in length. The *History of the Met Lab* volumes, which cover shorter spaces of time, were published in February 1977, May 1978, 1979, and May 1980, respectively. The volumes range from 560 to 690 pages each in length, and represent a tremendous amount of source material from which to follow the thread of development of the thorium story.

Beginning in December 2012 and continuing until March 2013 I combed through the five volumes of Seaborg's history and extracted all of the relevant passages about the development of thorium and uranium-233. Volumes 1, 2, and 4 were in electronic form and this greatly facilitated scans on keywords like "thorium", "233", and "protactinium". Volume 3 was not available in electronic form, much to my dismay, but through the assistance of Dr. Jess Gehin at Oak Ridge National Laboratory I obtained access to a paper copy. Personally scanning through the pages and finding the relevant passages was tedious but volume 3 ended up having the majority of the source material that made its way into the final work. My goal was to find the precise moments in history when the understanding of the potential of thorium as an energy source began to distill upon the parties involved, and also the moments that made clear that thorium and uranium-233 would not have useful application in nuclear weapons. I believe that I have succeeded in this effort and can now tell the story, from primary sources, of how thorium as a weapons material *didn't happen*, and why that has persistent significance in the later development of nuclear energy.

The official history of the Manhattan Project, *The New World, 1939-1946*, served as an excellent "big-picture" view of the Manhattan Project and as a contrast to Seaborg's rather narrow focus on his chemistry group's effort to purify plutonium. But Seaborg's diaries remain the primary source material for this entire paper. A supplemental source was Alvin Weinberg's 1994 autobiography *The First Nuclear Era* [11]. Weinberg also served in the Met Lab, but in the reactor design

division rather than Seaborg's chemistry division. His insights into the period are limited to a single short chapter of his autobiography, but they offer a different perspective on some of the same events. Often, he is able to describe, from the perspective of many decades later, the ramifications of work that was pursued at the Met Lab and discarded as a failure.

But it is Glenn Seaborg that is the central character of this story for reasons that go beyond his authorship of the history volumes. In my opinion, Seaborg is one of history's truly great scientists. He had a vision that saw beyond the years, and it was his penetrating insights that gave clear guidance to the people that served under him. Some of them also showed great talent and insight; some were disappointingly short-sighted and dull. But his was the animating spark that led so many to accomplish greatness. Although he later went on to serve as the Chairman of the Atomic Energy Commission from 1961 to 1971, I do not think anything he ever did matched the significance of his work from 1939 to 1945 in the era of the Manhattan Project.

Although I cannot claim to have read every work that covers this subject and this period of time, I have studied this era extensively and have not been able to find anything that covers the topic of the discovery, development, and discarding of thorium during the Manhattan Project era to the degree these sources do. This thesis is a unique addition to the body of work on this era covering a subject that I believe will be of great importance to us as we seek to develop a lasting energy source for the twenty-first century and beyond.

Acknowledgements

I gratefully acknowledge the assistance and patience of Dr. Laurence Miller and Dr. Ondrej Chvala of the University of Tennessee. I also am very grateful for the help that Dr. Jess Gehin of the Oak Ridge National Laboratory provided in helping to procure a paper copy of the third volume of Seaborg's wartime history and several other memos of historical significance. Personnel at Lawrence Berkeley National Laboratory and Oak Ridge National Laboratory also made several historical photographs available for inclusion in this work, and their help is gratefully acknowledged.

1 Introduction

On August 8, 1967, Dr. Glenn Seaborg, chairman of the United States Atomic Energy Commission, sat at the controls of an experimental reactor in Oak Ridge, Tennessee. The reactor was about to become the first to operate on uranium-233, an isotope of uranium that no longer occurs naturally on the Earth. Uranium-233 had been created from thorium, a natural material that was three to four times more common in the Earth's crust than uranium.

Figure 1: Dr. Glenn Seaborg, head of the Atomic Energy Commission, at the controls of the Molten-Salt Reactor Experiment guiding the reactor to achieve criticality for the first time using uranium-233 as its fuel. Image courtesy of Oak Ridge National Laboratory, U.S. Department of Energy.

In the early 1940s, when Dr. Seaborg was a young professor at the University of California, Berkeley, he had suspected that thorium could be converted from its natural form into a nuclear fuel. The process of nuclear fission itself had only been discovered the previous year by scientists in Germany. Seaborg's suspicion was correct, and he was the first to realize that thorium could become one of the world's greatest energy resources.

But Seaborg's discoveries about thorium and its daughter product uranium-233 were overshadowed by his previous discovery of plutonium. Plutonium's relationship to natural isotopes of uranium positioned it to become the material of choice for nuclear explosives. Soon after the discovery of plutonium Seaborg and tens of thousands of other scientists, engineers, soldiers, and workers were part of a wartime project known as the Manhattan Project whose goal was to develop nuclear weapons to end the war against Germany and Japan.

But what of thorium? For a variety of reasons, it had not shown the promise for nuclear weapons that plutonium did, and research on thorium and uranium-233 received far less attention during the Manhattan Project than research on plutonium and the natural isotopes of uranium. To understand why, we must follow the thorium story from its discovery up through the Manhattan Project, its success, and its dissolution into the civilian US Atomic Energy Commission at the end of 1946.

2 Before the Manhattan Project

2.1 Radioactivity and the Stability of Matter

From its discovery in 1828 in Scandinavia until 1898, there was little to distinguish thorium from the other eighty or so elements that had been discovered. It was about as common as lead in the Earth's crust, accounting for roughly six parts per million on average. A cubic meter of average crustal material contained about two cubic centimeters of thorium, if refined to metallic form. A ceramic compound of thorium, thorium dioxide, attained a degree of commercial usefulness in the mantles of gas lanterns beginning in 1890, when Carl Auer von Welsbach showed how thorium dioxide gave off a glorious white light.

In 1896, Henri Becquerel discovered that uranium was emitting rays that caused a photographic plate to darken, even when it was kept from exposure in a drawer. His discovery intrigued 28-year old Marie Sklodowska Curie, a Polish physicist and chemist who worked with Becquerel along with her husband Pierre. Curie and her husband began a campaign to examine all materials to discover if they also displayed the "radioactivity" that Becquerel had seen in uranium. In 1898 she noted [12]:

> I examined a large number of metals, salts, oxides, and minerals.
>
> All the uranium compounds studied are active, and are, in general, more active to the extent that they contain more uranium.
>
> The compounds of thorium are very active. Thorium oxide surpasses even metallic uranium in activity.
>
> It is remarkable that the two most active elements, uranium and thorium, are the ones which possess the greatest atomic weight.

Her discovery that thorium was also radioactive, and the penetrating insight that thorium and uranium were the two elements with the greatest atomic masses, was one of the key intellectual steps that would lead to a revolution in our understanding of atomic structure.

Figure 2: A re-creation of Marie Curie experimenting with uranium and thorium.

Within a few decades, the notion of an atom as an impenetrable, unchangeable sphere of particular composition had given way to our current understanding of the atom—a positively charged nucleus composed of protons and neutrons, surrounding by tiny orbiting electrons in a variety of orbital configurations.

Furthermore, there were further forces acting at the scale of the nucleus that made the structure of matter possible. They had previously been indetectable due to their very short ranges, but without them there would be no reasonable explanation for the structure of matter or the order of the universe.

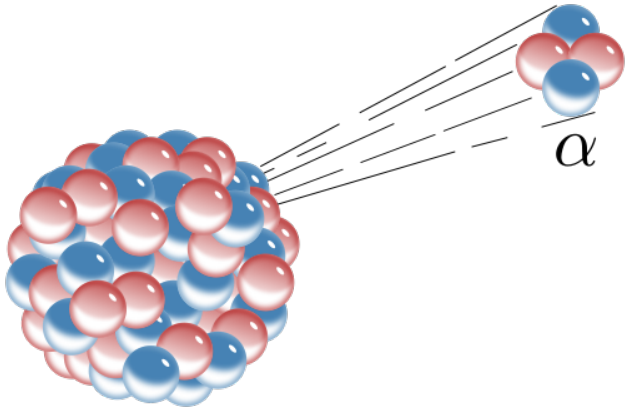
The most basic of these was the nuclear force, or the strong nuclear force as it is known today. The nucleus is composed of positively-charged protons and neutrons that have no electrical charge. Based on an understanding of electromagnetic forces (which was correct) these positively-charged protons should be continuously attempting to rip the nucleus apart, since electric charges of the same sign repel one another. This repulsion grows stronger and stronger the closer the particles are to one another, and by all indications, the protons were very very close together.

The neutrons should be no help at all in this situation, since they had no charge. But the nuclear force, as postulated, was a force that strongly bound nucleons (protons and neutrons) together. It had the ability, somehow, to overcome these forces of electric repulsion and to make nuclear structure possible. Now we understand these forces come from interactions of subatomic particles, quarks and gluons, which form the structure of nucleons.

The strong nuclear force also helped uncover the mystery of radiation, and why various nuclear configurations, particularly those associated with very heavy nuclear masses, were unstable and radioactive.

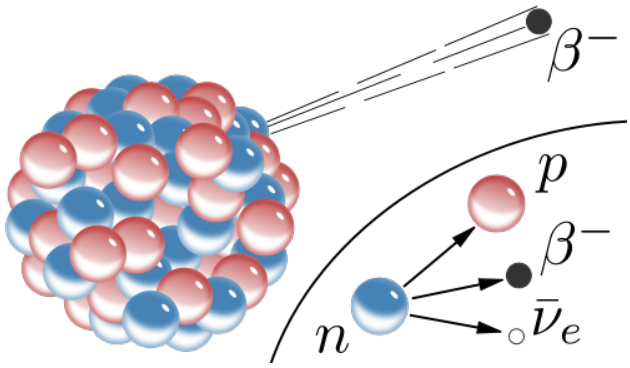
2.2 Three Decay Chains, One Missing

Long before the nature of radioactivity was understood, Marie Curie and those who extended her research studied what radioactivity did.

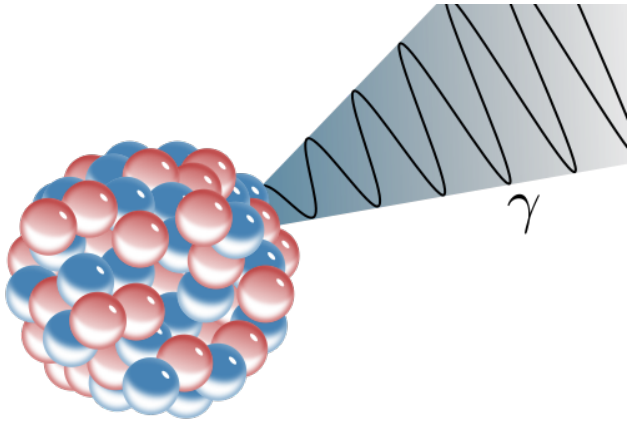


The first radioactive decay identified was the alpha decay, where an alpha-particle is emitted from the nucleus. The alpha particle was identical to a nucleus of helium-4, containing two protons

and two neutrons. It is the most stable nuclear arrangement. Through emission of an alpha particle, a nucleus loses four units of atomic mass and changes into another element, two locations lower on the periodic table. Thus uranium upon alpha emission immediately becomes thorium, radium becomes radon, radon becomes polonium, and so forth.



The second radioactive decay process was the beta decay, where a beta-particle (an electron) is emitted from the nucleus and a neutron transforms into a proton. Unlike alpha decay, the nucleus retains essentially the same atomic mass, since the proton has nearly the same mass as the neutron. However, the chemical nature of the nucleus changes. There are two types of beta decay: in beta-minus decay the nucleus moves up one step on the periodic table and in beta-plus decay it moves down one step. But atomic mass remains essentially constant.



The third type of radioactive decay process was the gamma decay, where a nucleus in an excited state emits a high-energy photon. These photons are distinguished from most others by their point of origin—rather than being emitted by changes in the electron configuration, they are emitted from the nucleus. The nucleus is typically left in an excited state after the emission of alpha or beta radiation, and gamma emission is a way for an atomic nucleus to radiate that excess energy out

as a photon. Like beta emissions, gamma emissions do not change the atomic mass of the nucleus by any significant amount. Unlike beta emissions, they do not change the chemical identity of the nucleus.

Thus the three common forms of radioactive emission have a variety of effects on the nucleus. Both alpha and beta emission change the chemical identity of the nucleus, but only alpha decay changes the atomic mass. Gamma emission changes neither atomic mass nor elemental identity. Since only one form of radioactive emission changed atomic mass, and since it always changed it by exactly four atomic mass units, an invariant relationship was soon discovered in the decay of heavy radioactive elements like uranium and thorium. When they lost atomic mass through alpha emission, it was always by four atomic mass units. Beta emission changed their elemental identity, but not their atomic mass. Thus if one knew what that atomic mass of a radionuclide was in an integer number of atomic mass units—for example, 238 atomic masses in a nucleus of uranium-238–then one could divide by four and examine the remainder.

It would always be the same, no matter how many times the nuclide decayed.

So it stood to reason that there would be four natural "chains" along which a radionuclide would decay, each characterized with a different remainder when their atomic mass was divided by four. The first one of these chains that was elucidated was the "uranium" chain, which began with common uranium-238 and ended on stable lead-206. Between U-238 and Pb-206 there were many intermediate steps, such as radium-226, radon-222, and polonium-210, but each of these radionuclides had a common basis. Their atomic masses, when divided by four, had a remainder of two.

Another decay chain began with common thorium-232 and terminated on stable lead-208. This decay chain was called the "thorium" decay chain and its atomic masses, when divided by four, had a remainder of zero.

The final natural decay chain began with rare uranium-235 and included notable isotopes of protactinium (Pa-231) and francium (Fr-223), and astatine (At-219), terminating on stable lead-207. This decay chain was called the "actinium" decay chain and its atomic masses, when divided by four, had a remainder of three.

Three decay chains, but four possible remainders when a number is divided by four. One appeared to be missing. The only hint left to its possible existence was bismuth, which consisted of a single isotope, bismuth-209. Bismuth-209, when its atomic mass was divided by four, had a remainder of one, the same as the missing decay chain. Where was it? Had it ever existed? Which radionuclides laid on that decay chain, and did they have any value to humanity?

Like an abandoned ruin, bismuth-209 was the only indication that another path of radioactive decay had once existed, perhaps billions of years ago when the solar system was young and its elements were fresh. And there was no way to recreate it, until insight and determination produced a powerful new tool to rearrange matter at the nuclear level.

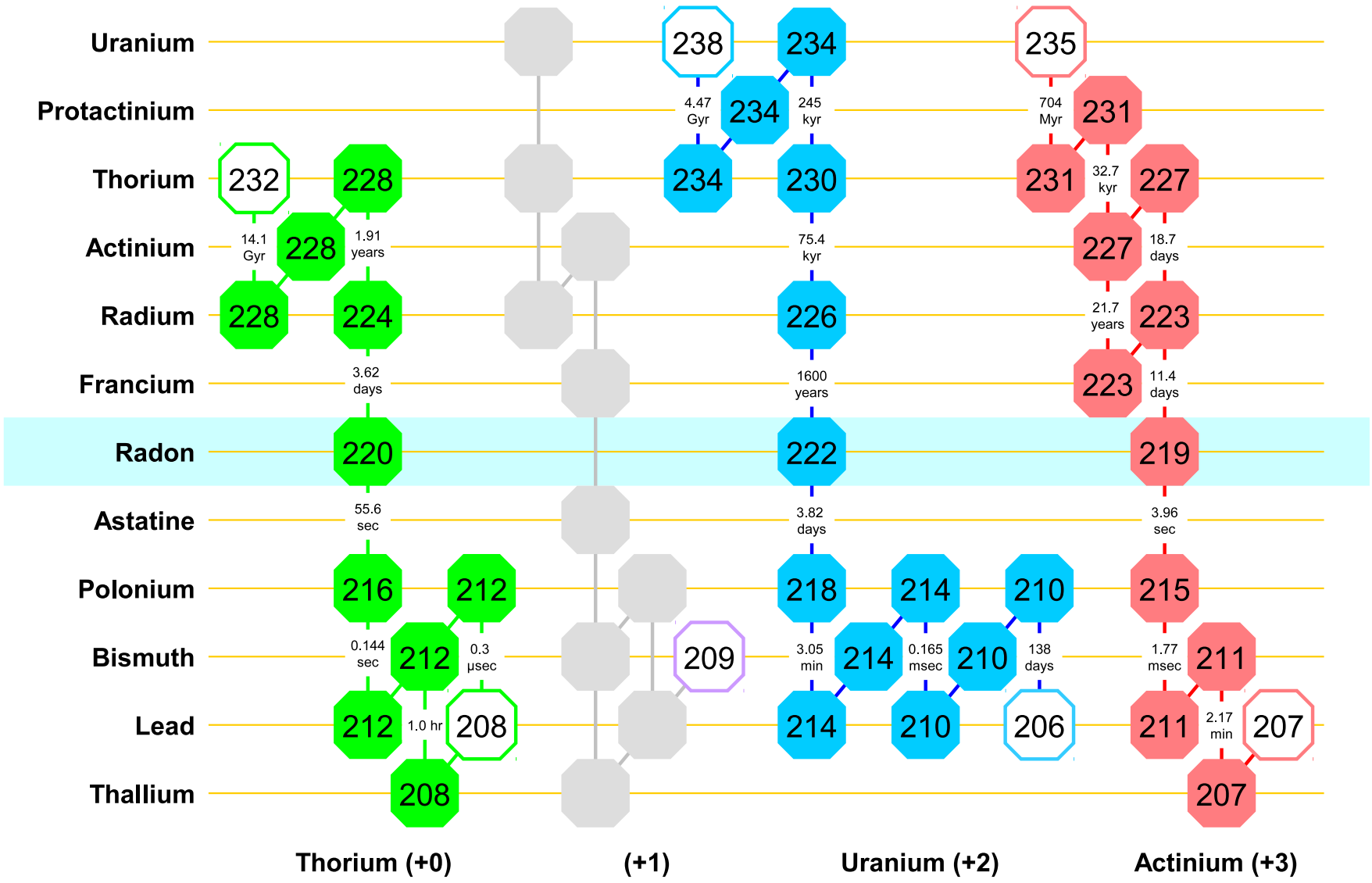


Figure 3: Three natural decay chains, but was one missing?

2.3 Ernest Lawrence and the Sixty-Inch Cyclotron

Ernest Orlando Lawrence was 27 years old when he arrived at Berkeley in the summer of 1928 to begin a campaign of physics research that would have the most profound consequences. Lawrence had been inspired by a paper from a Norwegian engineer to develop a unique circular particle accelerator, which he referred to as his "proton merry-go-round," but which became better known as the cyclotron. The earliest cyclotron was not much to look at, just a handful of glass and bronze, but the concept could be adapted to larger and larger units. Fundamentally, a cyclotron accelerated charged particle beams using a high frequency alternating voltage which is applied between two electrodes. Because it was based on accelerating charged particles, only particles like protons, deuterons, and helium-4 nuclei were suitable for use.

The first cyclotron of significant size was built in 1932 and was capable of boosting protons to energies of 80 keV. Soon a cyclotron 11 inches in diameter was attaining proton energies of over 1 MeV. It was followed by a 27-inch cyclotron that achieved 5 MeV, and by 1936 even that had been replaced by a 37-inch cyclotron which could accelerate deuterons to 8 MeV and alpha particles to 16 MeV.

But the machine that would make history was the seemingly colossal 60-inch cyclotron, which began operations in 1939. Its magnet weighed 220 tons, and it could generate a stream of 16 MeV deuterons. New innovations came when the chemists and physicists of Berkeley began to point that high energy stream of deuterons at various elements.

Initially physicists pointed the deuteron beam of the cyclotron directly at various materials to look for interactions at the nuclear scale. Despite the great power of the cyclotron, the high energy deuterons that it produced were still charged particles, and the positively charged nucleus of the atom repelled them, leading to relatively few events where a nuclear reaction took place.

High-energy deuteron bombardment was a difficult way to alter the configuration of a nucleus, yet it led to the discovery of both neptunium and plutonium, when abundant uranium-238 was exposed to the power of the cyclotron.

An uncharged particle, the recently discovered neutron, had the potential to bring about greater changes in the nucleus than the charged particles of the cyclotron. But how could neutrons be produced? There was no such thing as a neutron beam nor any real prospect of making one. But some materials tended to eject neutrons when bombarded by deuterons. One of these was phosphorus, which consists entirely of a single stable isotope, phosphorus-31. When phosphorus-31 is struck by a high-energy deuteron, it can absorb the deuteron and eject a neutron, in the process becoming sulfur-32. Thus deuteron bombardment of phosphorus was one of a very few ways known at the time to make a relatively large flux of neutrons, far more than had been produced ever before.

$$^{31}\mathrm{P}(d,n)^{32}\mathrm{S}$$

These neutrons were ejected from the phosphorus target at high energies, and at these high energies they had less potential to be absorbed in other atomic nuclei. By slowing neutrons down, it was found that the probability they would be absorbed increased. Neutrons could be slowed down through collisions with other atomic nuclei that had about the same mass they did. The best

Figure 4: Ernest Lawrence in 1939.

Figure 5: The 60-inch cyclotron where uranium-233 was first synthesized in 1941.

nucleus for the job was one that had a single proton—common hydrogen—and so materials that contained a great deal of hydrogen, like water or paraffin, were well suited to the task of slowing down the neutrons generated by the deuteron bombardment of phosphorus.

The recipe was coming together: a mighty cyclotron capable of accelerating deuterons to very high energies, impinging on a target of phosphorus, adjacent to a block of paraffin that would slow down the neutrons that were produced in the collision. The last ingredient was at the discretion of the researchers—what material should be placed in this flux of slow neutrons?

For young professor Glenn Seaborg, the ability to make neutrons, albeit indirectly, from the 60-inch cyclotron was an opportunity to recreate the missing decay chain and see what wonders it might hold.

Figure 6: Glenn Seaborg in 1938. Photo courtesy of Lawrence Berkeley National Laboratory.

2.4 Glenn Seaborg Begins to Use the Cyclotron

Glenn Theodore Seaborg was born in Ishpeming, Michigan on April 19, 1912 to Herman Theodore Seaborg and Selma Olivia Erickson Seaborg. While he was still a boy, the family moved from Michigan to an unincorporated area of Los Angeles County, California. Inspired by a high school chemistry teacher, Seaborg graduated high school at the top of his class and went on to study chemistry at the University of California, Los Angeles (UCLA), where he graduated in 1933. He went on to earn a Ph.D in chemistry from the University of California, Berkeley in 1937.

He remained at Berkeley after graduation and became an instructor in chemistry on July 1, 1939 and was fascinated by the recent discoveries by Otto Hahn and Lise Meitner in Germany of nuclear fission. Otto Hahn was a radiochemist as Seaborg aspired to be, and so it was natural that Seaborg would take an interest in using chemical techniques to ascertain whether nuclear processes such as fission or transmutation had occured in a sample. With the powerful 60-inch cyclotron at his disposal, Seaborg began to bombard a variety of material samples with neutrons, then to chemically "dissect" the resulting products in his lab looking for the results he expected, and occasionally, for some surprises.

Working with his friend and research partner Joseph Kennedy, Seaborg began to bombard samples of uranium with neutrons produced from the 60-inch cyclotron in late August 1940. He already knew that neptunium-239 would quickly form from that reaction, but since the Np-239 had a short half-life, he anticipated that it would decay into a new element—element 94—which at that time had no name. We now know element 94 as plutonium, and Seaborg, Kennedy, and several other colleagues are credited with its isolation and discovery on February 23, 1941. By March 28, 1941, they had ascertained that plutonium-239 undergoes fission by slow neutrons. This would have profound implications for the world, but the story of this paper follows another thread. Seaborg had shown that he could use the neutrons from the 60-inch cyclotron to turn uranium into a new element. His penetrating curiosity made him wonder what would happen if he repeated the attempt with thorium.

2.5 Gofman Begins Thorium Research Under Seaborg

Two new graduate students arrived at Berkeley to begin the fall semester of 1940, and both elected to study under Seaborg. One of them, John W. Gofman, had previously studied at Oberlin College, a private liberal arts college about 35 miles southwest of Cleveland, Ohio, where he had earned a bachelor of arts degree. Seaborg saw in Gofman an opportunity to investigate the missing 4n+1 decay chain of the elements, using the sixty-inch cyclotron to bombard the most suitable representative of the 4n decay chain, thorium-232. Seaborg anticipated that radionuclides from the 4n+1 decay chain would represent an entirely new family of radioactive isotopes, whose properties and half-lives would have unknown benefits. Seaborg was already aware of the existence of short-lived thorium-233 produced by neutron bombardment of natural thorium-232. Thorium-233 was the first representative of this 4n+1 decay chain, and it was already known to decay to protactinium-233. There were hints from the work of Hahn and Strassmann that a fissile nuclide might be produced. But what happened beyond the decay of Pa-233 was a new frontier of science. [6, pg.2]

Figure 7: Dr. Glenn Seaborg visiting Room 307 of Gilman Hall at the University of California, Berkeley. This was the room where on March 28, 1941 it was demonstrated that plutonium-239 was fissile.

Gofman arrived on September 23, 1940 and spent several weeks familiarizing himself with the chemistry of thorium, protactinium, and uranium. He gathered the chemical equipment that would be required as well as the radioactive decay event counting equipment that would be necessary to determine the rate of radioactive decay and whether it was based on alpha or beta emissions. His work began in earnest on November 4 [6, pg.3], and on Sunday, November 10 he undertook his first bombardment of thorium (in the form of thorium nitrate) by neutrons produced from the bombardment of phosphorus by deuterons accelerated by the sixty-inch cyclotron. The neutrons were slowed to thermal energies by a block of paraffin surrounding the thorium nitrate target, thus maximizing the likelihood that the neutrons would be absorbed by the thorium. The sample was exposed for only 75 minutes.

After the exposure of the thorium sample to neutrons Gofman took the sample to his chemistry lab. It was now far more radioactive than before, since some of the thorium-232 nuclei had absorbed neutrons to become thorium-233. The parent material, thorium-232, has a half-life of 14

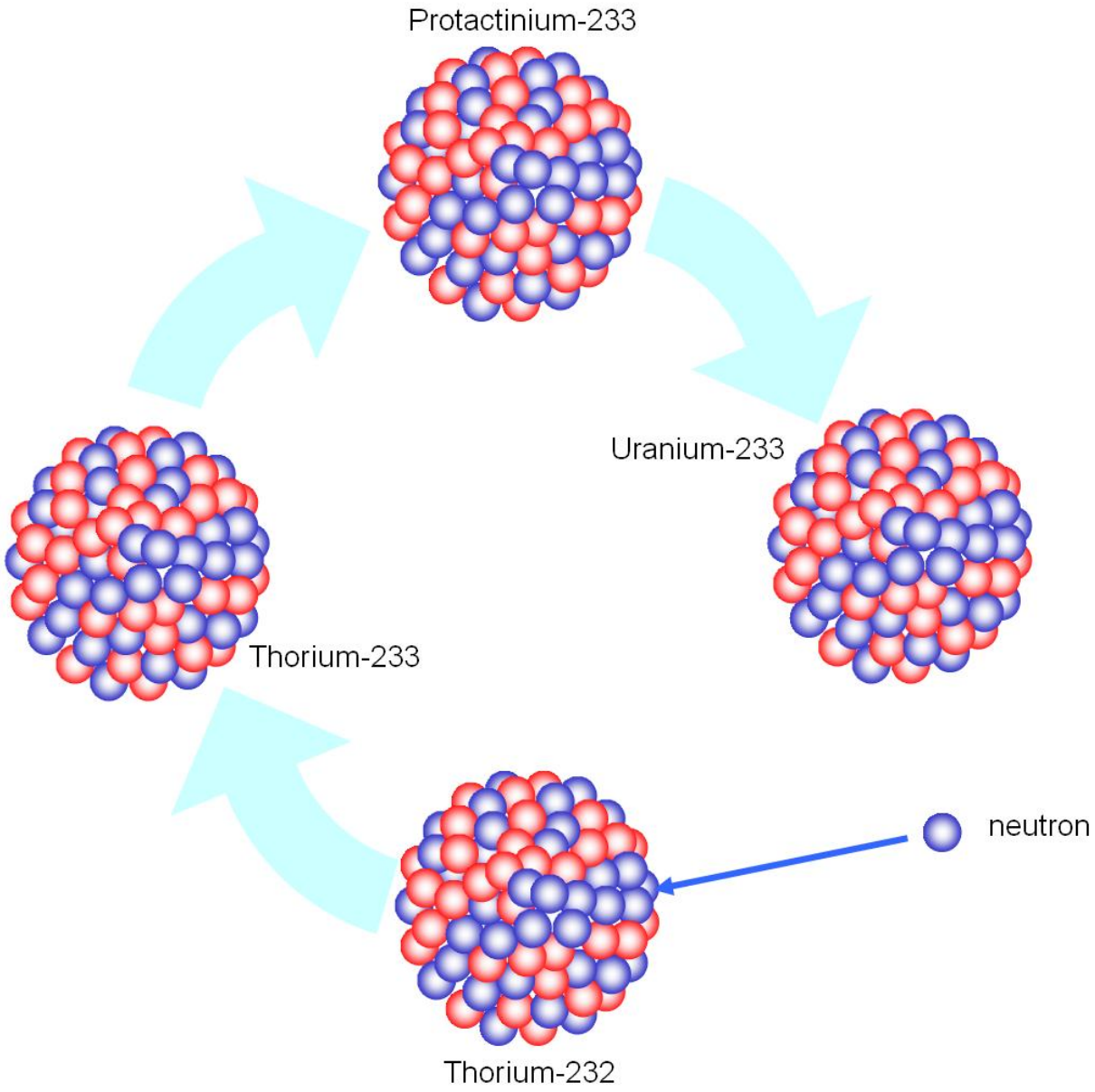


Figure 8: Synthesis of uranium-233 from thorium.

billion years, meaning that an individual decay event happened very rarely. But thorium-233 had a half-life of only 22 minutes, so despite there being very little thorium-233 in the sample, it was easily detectable.

Two weeks later, on November 24, Gofman repeated the experiment with 15 grams of thorium nitrate exposed for an hour [6, pg.4], and again on December 8 with 50 grams of thorium nitrate and an exposure time of several hours. [6, pg.5] A longer exposure was necessary to build up

Figure 9: Dr. John Gofman in 1962. Photo courtesy of Lawrence Berkeley National Laboratory.

more protactinium in the target, so that chemical separation techniques could be attempted. After a few days for the thorium-233 to decay away, Gofman judged the sample ready for an attempt to separate protactinium from thorium using chemical separation techniques that had previously been noted to concentrate protactinium.

The next step would take a bit longer. There was only one report of the half-life of protactinium-233, done the previous year by the pioneering discoverers of nuclear fission, Otto Hahn and Fritz Strassmann. They indicated that Pa-233 had a 25-day half-life, which meant that if Gofman was to find uranium-233 in his irradiated sample, he would have to wait several months for it to "grow in" and then wait even longer to determine what characteristics it might have. By January 14 of the new year of 1941, he had indeed confirmed the half-life of Pa-233 was 25 days, for he was seeing the radioactivity of his sample falling by half every 25 days. But the properties of uranium-233 were going to require further patience.

Seaborg sent a letter to *The Physical Review* on February 1 called "Radioactive Properties of Protoactinium" where they described their experiments and the chemical techniques they had used to isolate protactinium. [6, pg.21] As part of the wait for uranium-233, they began irradiating other samples of thorium, so that they would begin their decay process even as they waited for results from their first experiment. Fifty grams of thorium nitrate were exposed in the cyclotron previously on December 8, and then 1000 grams of thorium nitrate from February 21 through March 4. Gofman began to extract the protactinium from this much larger sample on March 28. [6, pg.37-38]

Seaborg left the Berkeley lab on April 6 to travel to New York to interview with a prospective employer, American Cyanamid Corporation. [13, pg.37] It was the first time Seaborg had flown in an airplane. While he was gone, Gofman completed the extraction of the protactinium inventory from the large 1000 gram sample on April 10. Seaborg returned to Berkeley on April 16, having been offered the job with American Cyanamid but having chosen to decline it. The magic he and Gofman had been hoping for was beginning to take shape in their isolated protactinium sample.

On April 23, Seaborg noted that there was "a small but discerable quantity of alpha activity" in their protactinium sample. [6, pg.38-39] The fact that they were discerning alpha radiation from the sample rather than beta radiation indicated that the source of the radiation was definitely not Pa-233. It was the new uranium-233 that was being generated as Pa-233 decayed away. The fact that its alpha radioactivity was growing even as the beta radioactivity was diminishing was a sure sign that the decay of the Pa-233 was the origin of the uranium-233, and more and more of it would be present as the isolated protactinium sample continued to decay.

No one knew what the half-life of uranium-233 was, but by counting the alpha particles from the sample they could make a rough estimate of 100,000 years. This was a radically different half-life than Pa-233. This meant that uranium-233 might decay slowly enough to be useful as a nuclear fuel, provided it had suitable nuclear characteristics. Seaborg made a prophetic statement in his journal that day:

> Of special importance is our demonstration through these results that U-233 is sufficiently long-lived to be a practical source of nuclear energy should it be found to be fissionable with slow neutrons and should methods for its large scale production be developed. [6, pg.38-39]

On March 28, the same day Gofman began his chemical extraction, Seaborg and another part of his research group had discovered that plutonium-239, which was formed from neutron absorption in abundant natural uranium-238, would fission with slowed-down (thermal) neutrons. This was a tremendous discovery since previously only naturally rare uranium-235 was known to fission with thermal neutrons. Seaborg's mind must have been fresh to the possibility that his newly-discovered uranium-233 might possess the same potential as well, and using the techniques that had led to the demonstration of fission in plutonium-239, he might be able to find out.

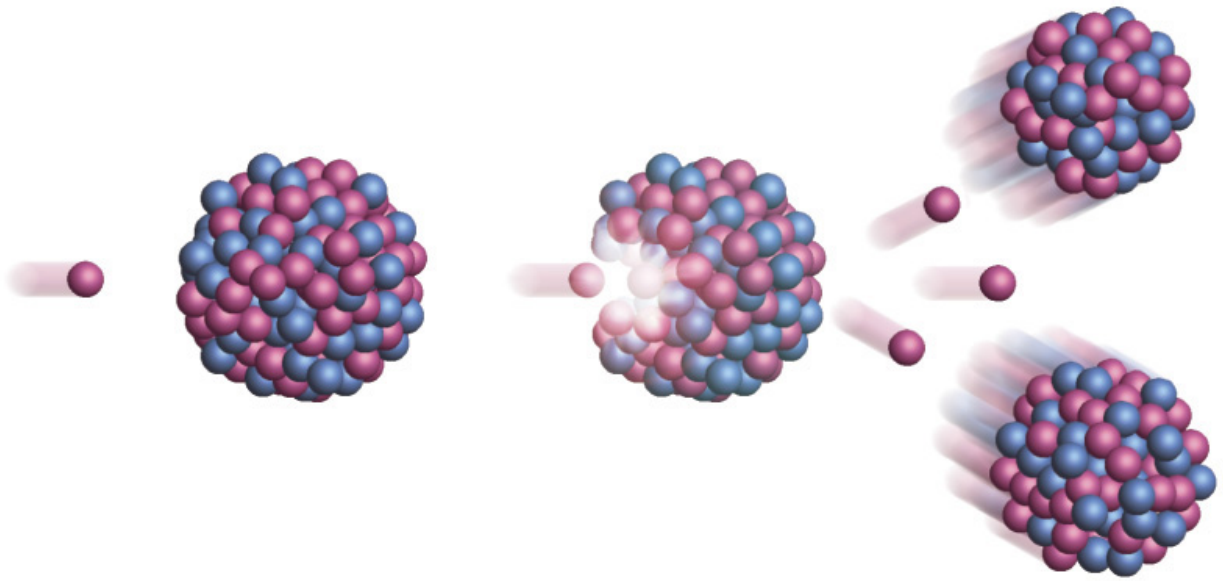


Figure 10: A fission reaction. A neutron strikes a heavy nucleus, such as uranium-233 or plutonium-239, causing it to split into two fission products and several neutrons. The two fission products emerge from the reaction with a great deal of kinetic energy, which as they slow down heats the medium in which they are located. The neutrons emitted go on to sustain the chain reaction and breed new fissile material from any fertile material present.

2.6 Elevating the Priority

Through the rest of the spring semester Gofman continued to refine his chemical techniques for isolating protactinium from irradiated thorium samples, but when the summer came Seaborg didn't have sufficient funding to continue his research and sent him home to his family in Cleveland until the fall semester started. [6, pg.47]

During the summer Seaborg reported the results of the discovery of uranium-233 to Dr. Lyman Briggs and Professor Lawrence. Briggs was the head of the National Bureau of Standards and was chairman of the committee whose purpose was to investigate the use of atomic energy in warfare. [13, pg.23]

Behind the scenes, Seaborg's discovery that plutonium was fissile had attracted a great deal of attention from the men who were secretly leading the American investigation of nuclear warfare. Seaborg had communicated his results to Ernest Lawrence on May 19th, who in turn passed the message on to Arthur Compton, the chairman of the National Academy of the Sciences Review Committee and to Vannevar Bush, the chairman of the National Defense Research Committee. Compton told Bush that the demonstration of the fissile nature of plutonium-239 (or "element 94" as it was then known) increases the importance of uranium by over 100 times, since now the previously worthless uranium-238 might be converted into a nuclear weapon material.

Up until this point, Seaborg's investigations of plutonium, thorium and uranium-233 had been reflections of his scientific curiosity. But that began to change in June 1941 when Seaborg was brought "into the fold" of the national effort to develop a nuclear weapon. On June 16, Seaborg received a letter from Vannevar Bush appointing him an official investigator in connection with a contract between the University of California and the National Defense Research Committee. Seaborg took a Pledge of Secrecy and an Oath of Allegiance before an authorized official. [13, pg.46] Plutonium work had become military work.

On June 28, 1941, President Roosevelt established by executive order the Office of Scientific Research and Development, located within the Office for Emergency Management. Vannevar Bush would be its first director and he would report directly to the President. OSRD was to mobilize the nation's scientists and apply the latest research to national defense. The NDRC would continue but would operate within the OSRD. [1, pg.41]

Rare uranium-235 had been the entire focus of the military effort to develop a nuclear weapon. It was an exceptionally difficult task to separate uranium-235, which constituted only 7 parts in a thousand of a natural uranium sample, from common uranium-238. Separating two isotopes of an element is always challenging because they have identical chemical properties. But plutonium was an entirely new element, and that meant that if it was created it must be possible to separate chemically—indeed, the act of discovery of plutonium was made possible by a chemical separation. If plutonium-239 was fissile, and if it was made from abundant and common uranium-238, and if natural uranium also contained rare but fissile uranium-235, then an entirely new option was now available. A nuclear reactor could be built where uranium-235 fission led to the production of neutrons that would be absorbed in the far-more-common uranium-238, producing plutonium-239. Then it was theoretically a straightforward step to extract the plutonium chemically.

This approach was extremely appealing and interesting to the infant nuclear weapons program in the United States. Rather than difficult isotopic separation techniques to pull U-235 out of uranium, they could use the U-235 to make something like it (Pu-239) but chemically extractable! Briggs, Lawrence, and Bush could see the challenges that must lie ahead but were intrigued at the possibilities.

It is important to consider that previous to the discovery of the fission of plutonium-239, the notion of building a reactor and the notion of building a nuclear weapon seemed like completely separate potential uses of nuclear energy. If one built a nuclear reactor to make power, then that reactor would be consuming scarce uranium-235 that could otherwise be isotopically separated and used in a weapon. It was an either/or proposition, and if one had nuclear weapons as a top priority, then the investigation of nuclear reactors for power was an expensive diversion that did nothing

more than consume scarce uranium-235. But the discovery that plutonium was fissile changed all that. It was the bridge between reactors and weapons. Now one could imagine why building a uranium-fueled nuclear reactor would be an essential step in producing the plutonium that could be used in a weapon. Enrico Fermi, working at Columbia University, began to consider the practical difficulties of building a uranium-graphite pile. [1, pg.42]

For Seaborg, the summer of 1941 brought something far more practical to his research: funding. Seaborg flew to Washington D.C. on August 3 to meet with Briggs and other officials at the National Bureau of Standards. While he was there, Berkeley was notified that his new project had been assigned "A-1-a" priority by the Priorities Division of the Office of Production Management. That meant that they would have top priority in purchasing key items for their project in competition with other national demands for needed items.

2.7 Does Uranium-233 Fission?

One of the important acquisitions that Seaborg made with his new funding was a young postdoctoral chemist named Raymond Stoughton, who had recently graduated from Berkeley. [6, pg.56] Stoughton was born in 1916 and had grown up in California where he had attended Berkeley as a chemistry student, obtaining his bachelor's degree in 1937 and his doctorate in 1940. He had gone on to teach chemistry for a year at Texas A&M University before returning to Berkeley to join Seaborg's group. [13, pg.54]

Seaborg intended for Stoughton to improve the chemical separation techniques for protactinium and uranium from thorium. If they could discover that uranium-233 was fissile, much like plutonium-239 was fissile, then Seaborg could add the potential of thorium to the national effort to build an atomic weapon. The thorium/uranium-233 effort was dwarfed by the rapidly growing effort in plutonium chemical separation, but it was, for the time, partaking of the bounty of funding and interest that the plutonium discovery had generated.

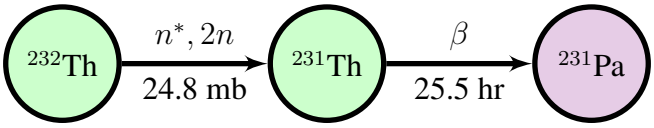
Stoughton, with Gofman supporting him, continued the work of isolating protactinium chemically from an irradiated thorium sample. By September, they had found promise in a technique where protactinium is co-precipitated with manganese dioxide. They also intended to apply electrolysis techniques that would prepare thin samples of Pa-233. Their intent was to bombard a much larger sample of thorium with neutrons from the cyclotron in order to produce enough U-233 to test for its fissionability by slow neutrons. [6, pg.59]

Using previously-exposed thorium samples, they also tested separation techniques using zirconium phosphate. Stoughton's chemical expertise was helping the U-233 project to move along much more rapidly than when Gofman had to act alone. By October they felt confident in their chemical techniques and began to bombard a 5000 gram thorium sample in the cyclotron beginning on October 30. This time the thorium would already be dissolved as a nitrate salt in water.

After 12 days and 14,250 microampere-hours of deuteron bombardment on beryllium to produce the neutrons needed for their thorium sample, they removed the sample on November 10, 1941 and immediately began their chemical extraction procedure. It took a week of chemical effort, using nitric acid, manganous chloride, and potassium permanganate to effect various chemical shifts in the tiny amount of protactinium present in the sample. [6, pg.69]

Figure 11: Dr. Raymond Stoughton, chemist and co-discoverer of the fissile properties of uranium-233. Photo courtesy of Oak Ridge National Laboratory, U.S. Department of Energy.

The next day they began looking for the alpha radiation that would tell them that uranium-233 was forming in their protactinium sample (from the decay of Pa-233) and it immediately showed an alpha-particle count. It was too early for U-233 to have formed in sufficient quantities, and they realized that another isotope of protactinium, Pa-231, had formed in the sample from the action of fast neutrons on thorium. [6, pg.70-71] Pa-231 was an alpha emitter but Pa-233 was a beta emitter. The reaction that formed the Pa-231 was



Pa-231 is the only natural form of protactinium. It has a half-life of 32,000 years and had been independently discovered in 1917-18 by two groups of scientists, Otto Hahn and Lise Meitner of Germany and Frederick Soddy and John Cranston of Great Britain. Since it was an alpha-emitter like U-233 it would confuse the measurement they were trying to make. But fortunately, its long half-life and the very short half-life of Pa-233 meant that the uranium-233 they desired could be separated chemically from their protactinium sample if they would simply wait for a period of time.

And so the generation of U-233 became a waiting game, whose rules were set by the roughly one-month half-life of Pa-233. Throughout the winter of 1941-42, while most of Seaborg's lab worked with haste on the new plutonium, Stoughton and Gofman waited for their ever-so-tiny amount of protactinium to gradually decay into uranium-233.

While they patiently waited, things were changing. Fermi was making experimental measurements of how neutrons would be multiplied in a pile of graphite and natural uranium, which would be the form a plutonium production reactor might take. Eugene Wigner of Princeton was also thinking about how a graphite-uranium reactor could make plutonium, and later he would design the machine that would. Results of US and British studies were being disseminated in government channels indicating that a nuclear weapon using uranium-235 or plutonium-239 could be feasible and likely would be small enough to be carried by an aircraft. And then the world changed. On December 7th, 1941, the Japanese attacked the US Navy base at Pearl Harbor, Hawaii with 353 aircraft launched from six aircraft carriers. 2402 Americans were killed in the attack and 1282 were wounded. Four battleships were sunk and four more damaged. The Japanese also sank or destroyed other ships and 188 aircraft.

The United States was shocked. The next day war was declared on Japan, and later on Germany. The effort to build a nuclear weapon took on added priority. Seaborg wrote in his diary:

> **A greatly expanded effort is getting underway at many places on the overall project of the preparation of fissionable material for use in a nuclear weapon.** This now has a high national priority and a large contract ($405,000 for six months) in the name of Lawrence is going into effect here at the University of California, Berkeley. This covers all of the work that I am doing, other than a couple of my specific contracts, as well as Lawrence's ambitious program for the separation of U-235 by the

> electromagnetic process, to which Kennedy and Segre are shifting their efforts. Similarly, a large program is getting underway at the University of Chicago under Arthur H. Compton concerned with the production of the isotope [plutonium]-239 from the nuclear chain reaction with natural uranium; this will be the center for this research program which has been given the code name **Metallurgical Project**, with the main component of this, at the University of Chicago, to be known as the **Metallurgical Laboratory**... I am assuming leave status from my position as Assistant Professor of Chemistry in order to devote full time to this high priority research program. My group is being substantially expanded in order to broaden the scope of my work. [6, pg.82-83]

Seaborg now realized that it was only a matter of time before he would be leaving Berkeley to join the growing group of researchers being gathered from around the country to join the "Metallurgical Laboratory" in Chicago, one of the obliquely code-named branches of the effort we now know as the Manhattan Project. All of the key plutonium researchers would be relocating from Berkeley to Chicago. Would uranium-233 be of sufficient importance to bring that small group as well?

The answer drew nearer as Stoughton and Gofman began the first chemical separations of uranium from protactinium in January 1942. Near the end of the month their pace of work accelerated considerably, with Gofman working nearly day and night to complete chemical separations. He could sense that the goal was near and was almost feverish in his pursuit. On February 2nd, with a freshly purified sample of uranium that he had finished only the night before, he began to bombard the sample with neutrons produced from a radium-beryllium source.

And there it was–at 9:44pm he found 39 counts per hour that could be attributed to the slow neutron fission of uranium-233. [6, pg.100] It was a rough realization, since there was still no knowledge of exactly how much uranium-233 they had laid down on the sample, nor the relative probability of U-233 fission to other isotopes, but the broad conclusion was obvious. Uranium-233 underwent fission in slow neutrons, just like uranium-235, just like plutonium-239. Thorium, a material three to four times more common than uranium, could be made into a nuclear fuel through neutron absorption. It was an epic discovery with profound implications for the future of mankind, but its significance was overshadowed by the haste of war and the focus on plutonium. The manufacture of plutonium in nuclear reactors and its chemical extraction and purification were already the central objectives of the "Metallurgical Laboratory". Seaborg assumed that the creation of uranium-233 and its extraction from thorium would attain equal priority and significance. But that did not happen, for a variety of reasons.

2.8 Preparing to Move to Chicago

Within just a few days from February 2, Gofman and Stoughton had confirmed that uranium-233's properties in fission came exclusively from neutrons. When they removed the neutron source (the radium-beryllium neutron generator) the fission counts stopped, and when they returned it, the fission counts started again. This is an important consideration for potential weapons use, since spontaneous fission was a very undesirable property in fissile material used in a weapon.

Figure 12: John Gofman, Glenn Seaborg, and Raymond Stoughton hold a plaque commemorating the 25th anniversary of the discovery of the fissile nature of uranium-233, in 1967. Photo courtesy of Lawrence Berkeley National Laboratory.

Seaborg's mind was largely occupied with his impending move to Chicago and the dawning realization of the magnitude of the challenge that plutonium production and separation would be. On February 3 he had left for a meeting in Chicago, and on February 7 he wrote in his journal:

> As a result of this meeting I now fully realize the magnitude of the project being planned around our new element [plutonium] and the enormity of the chemical separation problem of isolating [plutonium]-239 from large amounts of uranium and almost fantastic intensities of fission products. When Compton asked me if I thought I could devise very soon a chemical process for separating [plutonium] from such uranium and fission products—a process that could be successfully scaled up for actual use in

> a chemical extraction plant—I indicated that I thought I could, but I must confess to some misgivings. [6, pg.104]

In order to determine the fissile properties of uranium-233, it was necessary to determine just how much there was in the samples they were measuring. This was a very difficult task, and Stoughton labored mightily to coordinate the decays he was seeing in the original protactinium sample back to an estimate of just how much there was in the first place. From these estimates, along with a knowledge of just how long they had waited to make an extraction of the uranium, they could estimate the amount of uranium present on the sample. And with this estimate and the counting rates they were seeing from fission, they could make an intelligent guess at the actual behavior of uranium-233 relative to its fissile cousins, uranium-235 and plutonium-239. This required precision calibration of electroscopes, even accounting for known measurement errors that particular units had experienced in the past. It must have been very tedious and frustrating work. Gofman in particular was still searching for spontaneous fission, and was so bothered by electrical interference in his lab at Berkeley that he relocated the uranium-233, the radium-beryllium neutron source, and the ionization chamber to his apartment. Perhaps he was finally able to get some sleep as he waited for the single count that might indicate that uranium-233 had some measure of spontaneous fission. [6, pg.113]

By March 16, Stoughton had an estimation of the mass of uranium-233 on their samples, and from that difficult extrapolation he produced a half-life estimate for the material of 120,000 years, much closer to the modern calculation of 159,200 years. Stoughton estimated that the mass of uranium-233 in their tiny sample was about 0.8 micrograms, about the mass of a single particle of baking powder. One of the remarkable aspects of radioactive material is that this incredibly tiny sample of uranium-233 was still sufficient to begin to tease its secrets from nature. Their estimates for the half-life of Pa-233 were improving as well. Based on their latest work they calculated a half-life of 27.4 days.[1] [6, pg.121-123]

On March 18, 1942, Seaborg dictated a paper where he first suggested the name "plutonium" be assigned to element 94, after the planet Pluto that had been discovered in 1930 and named after the Roman god of the underworld. This continued the pattern established by naming uranium after the planet Uranus, and element 93 neptunium after the planet Neptune.[2]

Calculating the mass of U-233 in one sample also allowed them to calculate masses in other samples they had extracted, by using the ratio of their alpha-particle emission activity. Another sample, which they called "Sample J", had a mass of 3.8 micrograms by their estimation. Gofman had been studying Sample J in his apartment for several weeks, searching for any sign that it was undergoing spontaneous fission. By March 30 he had recorded about 175 hours of effective counting and had not observed any spontaneous fission events. This meant that if uranium-233 had spontaneous fission, the half-life of that fission must be greater than 100 trillion years, and far

[1] The modern accepted value of the half-life of protactinium-233 is 26.275 days.

[2] In 1942 Pluto was considered a planet, but the discovery of the dwarf planet Eris in January 2005 led to a crisis in the planetary science community that led to a formalized definition of a planet, which had previously not been explicitly defined. In 2006 Pluto was demoted to the status of a "dwarf planet" along with several other solar system bodies. Eris was named after the Greek goddess of discord but "erisium" rather than "plutonium" might have been an appropriate name for element 94, which has caused a great deal of discord.

beyond any level that would be of concern to the weapons program. Uranium-233 was looking more and more appealing to Seaborg.

On March 31, Seaborg composed a letter to Dr. Compton where he proposed "Investigations Which Might Be Considered in Case A Very Large Chemical Program Were Undertaken" in his absence at Berkeley. One of those investigations was "The development of chemical methods for separating kilograms of U-233 from large amounts of thorium and fission products in case U-233 should prove to be a useful isotope."

Meanwhile, in his much larger plutonium group, two of his students, Spofford English and Arthur Wahl, were investigating a promising technique called "fluoride volatility" in an attempt to separate plutonium from uranium and fission products. Fluorine is the most electronegative element on the periodic table, and nearly all other elements would convert to a fluoride chemical state in preference to all others. This made fluorination of materials an interesting option for chemical separation. Uranium tetrafluoride (UF_4) was one form of fluorinated uranium, but there was another, uranium hexafluoride (UF_6) that was a gas at mildly elevated temperatures. What English and Wahl were attempting to uncover was whether plutonium had fluoride states that were less volatile than UF_6. If there were chemical forms of plutonium that were less volatile than UF_6 then perhaps it could be a separation technique. But their early experiments were not terribly promising. Later the techniques of fluorination and fluoride volatility would play an important role in the development of the thorium-fueled molten-salt reactor.

By April 2, Gofman had given up the search for spontaneous fission and turned his attention back to neutron fission measurements of uranium-233. With his 3.8 microgram Sample J, he used the radium-beryllium neutron source to stimulate fission in the tiny U-233 sample. As he measured the counts, the picture that began to emerge was that uranium-233 was even better than uranium-235 in slow fission. It had about 1.3 times the "effective slow neutron cross section" than its natural cousin. [6, pg.139] This was another promising development for the use of uranium-233 in nuclear reactors. But nuclear weapons do not use slow neutrons for fission—they use fast neutrons, and the same measurement would need to be made for fast neutrons. Gofman began working on the fast-neutron-cross-section problem on April 9 but ran into problems immediately. Radon gas in his neutron source began to leak, contaminating the ionization chamber. [6, pg.142] Within a few days he got the leak fixed and resumed his experiments, but time was running out for Seaborg at Berkeley. He was in the final stages of his departure and was attempting to write up his results in a series of reports. One of those would be "Properties of U-233"[3] which would report that with a 3.8 microgram sample of U-233 they determined that U-233 had a slow neutron cross section 1.25 times greater than U-235, that it had a spontaneous fission half-life greater than 100 trillion years, and that it decayed by the emission of alpha particles with a half-life of 120,000 years. [6, pg.144]

Each of these important numbers had been discovered through countless hours of hard work by Gofman and Stoughton under the leadership of Seaborg. Each represented an incredible amount of planning, calculation, and experimentation. Fundamentally, they had been made possible in the

[3]The paper "Properties of U-233" (abstract) was issued by the S-1 Committee in Washington as report number A-153 dated April 13, 1942 with authors G. T. Seaborg, J. W. Gofman and R. W. Stoughton. After the war it was published under the changed title "Nuclear Propertics of U-233: A New Fissionable Isotope of Uranium" by G. T. Seaborq, J. W. Gofman and R. W. Stoughton in Phys. Rev. 71, 378 (1947).

first place by Lawrence's far-sighted decision to build larger and larger cyclotrons at Berkeley. No where else in the world would it have been possible to smash deuterons on a target with enough energy and intensity to produce such a flux of neutrons. And without that flux of neutrons it would have been impossible to make enough uranium-233 from thorium to determine its emission properties, its long half-life, and its superior slow neutron properties. It was an amazing accomplishment, achieved at almost the first moment in human history when it would have been possible.

On April 17, Seaborg and his colleague Isadore Perlman boarded the steamliner "City of San Francisco" and set out for the University of Chicago to join the new "Metallurgical Laboratory". [6, pg.148-149] There they would determine how to separate plutonium produced in a nuclear reactor from uranium and fission products. That plutonium would go on to be detonated in the first nuclear explosion in New Mexico in July 1945 and over the city of Nagasaki on August 9, 1945. Their efforts would change the world.

2.9 Alvin Weinberg at the University of Chicago

Alvin Weinberg was born on April 20, 1915 in Chicago and had grown up in its Albany Park district. His father abandoned his family [11, pg.1-2] when Alvin was 15 just as the Depression was beginning and Alvin saw little of him in the years to come. When he was 16 he began studying at the University of Chicago, first in chemistry, and later in physics. He credited the breadth of his education there to a new plan implemented by the university president, Robert Hutchins, to expose each student to elements of humanities, the social, biological, and physical sciences to much of his later success as a scientific adminstrator.

Although he claimed that he found physics to be a difficult subject, Weinberg showed quite an aptitude for it, and stayed at the University of Chicago to study under Professor Carl Eckart. Weinberg's research concerned mathematical biophysics, an ambitious attempt to model biological performance based on mathematical models of nerves and cellular metabolism. As part of his research Weinberg became adept at using a family of challenging mathematical functions called Bessel functions. Bessel functions were used to model electrical conduction along a long nerve ending.

Weinberg was awarded his Ph.D in the winter of 1939 on almost exactly the same day when German scientists were announcing their discovery of nuclear fission. Weinberg was awarded a National Research Council fellowship to work on nerve biology with one of the nation's top biophysicists at Columbia University, but that professor ended up coming to the University of Chicago to work on the new Manhattan Project. Eckart knew of Weinberg's capabilities and began to enlist him to make calculations for various aspects of neutron-multiplying "piles" of uranium and graphite or beryllium. These calculations made special use of Weinberg's skill with Bessel functions that he had acquired from his modeling of nerve endings. By the fall of 1941 as he was reporting his results, Eckart announced that he would be leaving Chicago for war-related work at the Undersea Naval Laboratory in La Jolla, California. Weinberg protested that he would not be able to continue his work without Eckart's guidance. Eckart simply said, "Don't worry. Eugene Wigner is coming to Chicago." [11, pg.10]

Eugene Paul Wigner had been born in Budapest, Hungary on November 17, 1902 to middle

Figure 13: Young Alvin Weinberg in his Manhattan Project-era days.

class Jewish parents. Wigner began studying at the Technische Hochschule in Berlin in 1921 at the age of 19, where he met such luminaries as Max Planck, Werner Heisenberg, Wolfgang Pauli, and Albert Einstein. He also met Leo Szilard who became his closest friend. In 1930 he was recruited to Princeton and emigrated to America, becoming a naturalized citizen in January 1937. His friend Leo Szilard had become very concerned about German activities in nuclear energy and at Szilard's prompting, Wigner drove him to a meeting with Albert Einstein. Szilard crafted a letter for President Roosevelt which Einstein signed. That letter is considered the origin of the Manhattan Project.

Weinberg met Wigner in February of 1942, around the same time that Gofman and Stoughton were discovering that U-233 was fissile. Wigner was commuting from Princeton every month and had been working loosely with Enrico Fermi on the problem on neutron multiplication in a pile of uranium and graphite. The cyclotron at Berkeley had been able to produce individual neutrons from accelerated deuterons, but even the most magnificent cyclotron in the world could only produce a few micrograms of plutonium or uranium-233. But with the multiplication of neutrons from fission, it could be possible to produce much more. If uranium piles could achieve the state of "criticality", where each fission led to exactly one more fission, then truly impressive production rates might be possible. This was the heart of Wigner's research. Weinberg had not known the purpose of the research when he began work in Eckart's group in Chicago, but it was whispered to him by a younger colleague one day in an unguarded moment.

Wigner had experienced Nazism in Germany and Hungary and was convinced that the Nazis would prevail over the free world unless the Metallurgical Laboratory could achieve its goals in the shortest possible time. [11, pg.23] He and Szilard had been pushing the government to action since 1939, and in 1942 they were convinced that precious years had been wasted because the project had not been moving fast enough. Weinberg said "Wigner after all counted each day until a bomb was made as an unnecessary gift to the Nazis." [11, pg.30]

3 Relevant Aspects of a Sustained Chain Reaction

3.1 Plutonium's Advantage in a Production Reactor

Although Seaborg's small U-233 team had shown that uranium-233 had many of the same advantages that plutonium-239 did as a fissile material, plutonium had a surpassing advantage over uranium-233 as a material for a nuclear weapons program. It was simply this: **uranium, the parent material of plutonium, already contained the fissile isotope needed to enable a reactor to achieve criticality**.

Thorium, the parent material of uranium-233, contained no such fissile isotope. A pile of natural thorium could never achieve criticality, no matter what its size, no matter what moderator material was used, because there was no fissile material present. This meant that any thorium pile would have to be supplied with fissile material[4] in order to achieve criticality and begin operation. But the entire objective of the Metallurgical Laboratory was to **create** fissile material in a pure and

[4]Either uranium-235, plutonium-239, or uranium-233.

chemically separable form. Creating pure fissile material and then utilizing it to start a thorium pile made no sense whatsoever. The very material that would be used to start the pile would be the material whose creation would be the goal of the pile!

The fact that uranium was composed predominantly of the parent material of plutonium-239 with just a small amount of fissile material meant that a production reactor to make plutonium could be achieved from the outset of the nuclear endeavour, using natural uranium. A chain-reacting pile need not depend on any uranium enrichment, only on the proper choice of moderator material and geometric arrangement.

But any pile with thorium as a fuel would have no fissile material and no potential for multiplication, to say nothing of actually achieving a chain reaction. **Thus, no matter what properties Seaborg and his clever chemists would find in uranium-233, there was no way that a thorium production reactor would ever achieve the same significance as a uranium production reactor.**

Why didn't natural thorium have any fissile isotopes? Well, several thorium isotopes have appreciable fission cross-sections in thermal neutrons. Further research would show that thorium-227 and thorium-231 had a fissile cross-sections about 38% and 21% those of uranium-235. But thorium-231 had a half-life of only one day, and thorium-227 only 19 years. In the eons of time that had passed since the heavy elements were formed in the cores of stars neither of these thorium isotopes existed on Earth in any appreciable amount. **Only thorium-232, with its 14 billion year half-life, about the age of the universe, persisted to the present day.**

Uranium also had other interesting isotopes, such as the uranium-233 that Seaborg and his group had discovered. But the only isotopes of uranium with truly long half-lives were uranium-238 and uranium-235, with half-lives of 4.5 billion and 700 million years, respectively. Only U-238 and U-235 persisted to the present day, and of all fissile isotopes since discovered, U-235 by far has the longest half-life. Thus natural uranium was the only material in 1942 that could be the basis of a chain-reacting pile. Since the difficult isotopic separation of uranium was the goal of another branch of the Manhattan Project, it was left to the physicists and chemists of the Metallurgical Laboratory to construct a chain-reacting pile based on uranium, and that pile would only make one chemically-separable fissile isotope in any quantity: plutonium-239.

This fundamental fact about the nature of uranium and thorium has consequences that persist to this day. If a nation or organization wishes to build the simplest nuclear reactor to produce chemically-separable fissile material, they will undoubtedly choose uranium as the reactor fuel and plutonium as the product. This is a basic reason why thorium and uranium-233 were of diminished consequence to the Metallurgical Laboratory and have been of little interest to nations or groups who have desired to develop nuclear weapons ever since.

3.2 The Priorities of the Metallurgical Laboratory

The priorities of the Metallurgical Laboratory in Chicago in 1942 had nothing to do with the eventual use of nuclear energy to produce safe and efficient civilian electrical power. They were entirely focused around the creation and purification of material for nuclear weapons. To this end, each group had particular objectives. Eugene Wigner and his group, including Alvin Weinberg,

were attempting to design the large plutonium-producing reactors that would eventually be built in Hanford, Washington. Glenn Seaborg and his chemists were attempting to design techniques to remove plutonium from uranium and fission products in a chemically pure form. And Enrico Fermi and his team were attempting to achieve the first controlled chain reaction in a pile of graphite and natural uranium there in Chicago. If Fermi was successful, it would aid Wigner's group in confirming the nuclear calculations that they were using as the basis of their designs. And without Seaborg's success, none of it would be useful.

Seaborg continued to correspond with his former Berkeley colleagues Raymond Stoughton and John Gofman after his arrival in Chicago. He also promoted uranium-233 as an alternative fissile material to plutonium-239, should the need arise. [7, pg.8] Since chemical separation would be an important part of any uranium-233 production scheme, by June Seaborg was corresponding with Stoughton about the potential of fluorination as a separation technique.

Fluorination was a clever chemical technique that took advantage of a basic chemical difference between thorium and uranium. Each element has a variety of "valence states" which describe how many bonds they will make with other elements. Thorium has a very simple valence structure, called "+4". This means that thorium will share four electrons to form four chemical bonds with other elements. Some elements typically form more than one bond, like oxygen. Oxygen will usually share two electrons to form two bonds with other elements, and with thorium two oxygen atoms will form a very chemically stable configuration: thorium dioxide (ThO_2), which is the chemical form thorium is usually found in nature.

Fluorine forms just one bond with other elements, so it requires four fluorine atoms to form a stable molecule with thorium. That molecule is called thorium tetrafluoride (ThF_4).

Uranium has two main valence states, +4 and +6. Depending on the conditions, uranium will share either four or six electrons with other atoms. This means that an atom of uranium could chemically bond with two or three atoms of oxygen (UO_2 and UO_3), or with four or six atoms of fluorine (UF_4 and UF_6). When uranium bonds with six atoms of fluorine it forms uranium hexafluoride, and this compound turns to a gas at relatively low temperatures.

The theory behind fluorination as a chemical separation technique is that a mixture of thorium and uranium tetrafluorides (ThF_4 and UF_4) can be separated from one another by adding more fluorine to the mixture. Two fluorine atoms will bond with each atom of uranium, causing uranium tetrafluoride to become uranium hexafluoride, which is typically gaseous at the temperatures of interest. Uranium hexafluoride, as a gas, will bubble out of a liquid mixture of ThF_4 and UF_4. Thorium does not form a gaseous hexafluoride like uranium does, so it is left behind in a fluorination process.

Seaborg's interest in fluorination as a separation technique came from the fact that thorium, exposed to neutrons in a reactor, would absorb some of those neutrons and form small amounts of uranium-233. If the thorium and uranium were chemically converted to tetrafluorides, which was a rather simple chemical step, and if that mixture was further fluorinated to convert the uranium to a hexafluoride, then it would be rather easy to separate uranium from thorium.

In late June Seaborg and Stoughton exchanged samples of thorium tetrafluoride that had been exposed to neutrons in the 60-inch cyclotron. Stoughton irradiated the thorium tetrafluoride in the cyclotron, then sent it to Seaborg in Chicago. Seaborg's chemists fluorinated it and then sent

the resulting material back to Stoughton for his analysis. By July 17, Stoughton reported that he had recovered 25-35% of the uranium-233 through fluorination. [7, pg.125] The basic idea of fluorination seemed sound. Later work would improve its efficiency considerably.

Despite his interest in thorium and uranium-233, Seaborg's overwhelming priority was the chemical separation of plutonium from uranium and fission products in the production reactors that would be built in Hanford. Fluorination was not a very effective technique for separating uranium from plutonium, because plutonium had a variety of valence states and could also form a gaseous hexafluoride. If plutonium hexafluoride formed in preference to uranium hexafluoride then there might have been interest, but just the opposite took place. During fluorination all of the uranium would react with the fluorine (forming gaseous UF_6) before the plutonium would, meaning that an entire volume of uranium parent material would need to be fluorinated to UF_6 before the plutonium would begin to fluorinate to PuF_6. This was undesirable, since they estimated that the Hanford reactors would have a million grams of uranium in their exposed fuel for every thirty grams of plutonium.

The fact that uranium would fluorinate to an easily-separable gas and thorium wouldn't made fluorination an attractive chemical separation technique for a thorium-uranium-233 reactor, but not attractive for a uranium-plutonium reactor. Since the priority of the Metallurgical Laboratory was chemical separation of plutonium, fluorination assumed a role of lesser importance over time.

3.3 The Significance of Achieving Criticality

Seaborg and his team at the University of California, Berkeley had originally begun their research into plutonium and uranium-233 using neutrons generated from a cyclotron. Why then was there such a strong interest in building a critical nuclear reactor when they had cyclotrons?

It was simply this—with the cyclotron there was a substantial cost associated with producing a neutron but with a critical nuclear reactor that cost dropped by orders-of-magnitude.

To produce kilograms of plutonium for a nuclear weapon it would be necessary to bombard natural uranium-238 with neutrons to form plutonium-239. To produce one kilogram of plutonium-239 from one kilogram of uranium-238 would require 2.5 trillion trillion neutrons (2.5×10^{24} neutrons). The 60-inch cyclotron that Seaborg and his students had used at Berkeley was the most powerful in the world and could generate high-energy deuteron beams that would smash into phosphorus, causing the ejection of neutrons. At the beam currents that had been used in previous experiments at the cyclotron (~35 micro-amperes) the cyclotron was only producing 200 trillion neutrons per second. At that rate, it would have taken 400 years to produce a single kilogram of plutonium-239. Since the Manhattan Project estimated that they would need tens of kilograms of plutonium for their mission, even a fleet of cyclotrons operating continuously would take far too long to produce the amounts of material needed.

Each neutron produced in a cyclotron represented a cost in terms of energy. It took energy to accelerate the deuteron to sufficient speeds to smash it into the phosphorus target, producing a neutron. The relationship between the cost in energy and the number of neutrons needed was direct and rather immutable.

Fission was another way to obtain the trillions of trillions of neutrons needed to make pluto-

nium. Each fission reaction released two or three neutrons and about 200 MeV of energy. The leadership of the Manhattan Project wasn't interested in the energies that would be released in fission (unless they were in an explosive); rather they were interested in the neutrons, which could then be used to create plutonium-239 from uranium-238. If each neutron could be "multiplied" to produce another and yet another, the cost of those additional neutrons would be reduced. And if a neutron could be multiplied an infinite number of times, then its cost plummeted relative to the cyclotron.

A critical reactor was a compelling option for producing large numbers of neutrons that held much more promise than cyclotron technology. The basic idea was that neutrons could be multiplied if you assembled fissile material, with the right moderator, in the right configuration. There was only one option at that time for the fissile material—uranium. It alone contained a fissile isotope (uranium-235) although U-235 was a tiny fraction of natural uranium. Only one uranium atom in 137 was uranium-235. The other 136 were neutron-absorbing uranium-238 atoms, which would not fission nor would they help to multiply neutrons significantly.

Enrico Fermi had been one of the pioneers in assembling piles of material that would multiply neutrons. Since 1939 he had been researching the notion of the "exponential pile", an arrangement of uranium and graphite that would try to demonstrate that each fission event could lead to another and yet another.

This notion of a neutron causing a fission reaction, which would lead to the emission of more neutrons, which would lead to more fission reactions, was very compelling because it promised to provide far more neutrons for the conversion of uranium to plutonium than external techniques like the cyclotron. But the basic challenge was the fact that uranium-235 was such a rare component of natural uranium.

The physicists expressed their accomplishment in terms of a single, non-dimensional number which they called "k", or the multiplication constant. The inverse of $(1 - k)$ was the number of additional fission events that would be triggered, including the original. If k was 0.5, then each fission event would lead to another. A k of 0.8 meant a fission event would lead to four more. A multiplication constant of 0.95 meant a fission event would lead to a total of twenty before the process ended. At k = 0.99 a hundred fissions would result. And at k = 1.0 something amazing happened. A fission would be multiplied an infinite number of times and the reactions would continue and continue without end. That magic number, k = 1.0, was called "criticality"; a value of k less than one was called "subcritical"; and a value of k greater than one was called "supercritical". All of the exponential piles that had been built had been subcritical, but they were getting closer and closer to a critical pile. Arranging a pile of uranium and graphite to achieve criticality was the goal of Fermi's group, first at Columbia University and later at the Met Lab in Chicago. In a supercritical pile each fission would lead to more than one additional fission and theoretically the power level would grow without bound.

Of course, the physicists couldn't simply wish for the multiplication constant to equal one and thus it was so. They had to arrange material carefully, studying the parameters of uranium and the graphite that surrounded it, to see which changes improved k and which reduced it.

Alvin Weinberg recalled his skepticism that criticality could actually be achieved:

> Fermi had performed the first exponential experiment in August 1941. From his sec-

ond experiment, in the fall, Fermi found a value k = 0.87—some 0.13 short of the magical k = 1. To increase k, three improvements were available: optimizing the dimensions of the lattice; shifting from uranium oxide to uranium metal; and ridding the graphite and uranium of tramp elements, such as boron, that absorbed neutrons. Thus Fermi brought to Chicago a program of successive exponential experiments, each embodying one or another improvement—purer graphite and uranium; uranium metal instead of oxide; and more optimal lattice spacing...in those early days I was skeptical. **The idea of a nuclear chain reaction simply seemed too miraculous, too unbelievable.** The distance from k = 0.87 to k > 1 struck me as a God-given barrier that could not be overcome. Even Johnny Wheeler's patient tabulation of the improvements in k provided by increased purity, optimized lattices, and the shift from oxide to metal failed to convince me. [11, pg.15-16]

An analogy might be made between the multiplication constant and skipping a stone on a pond. A skilled stone-skipper can cause a flat stone to skip many times before it falls into the water. This could be likened to a fission reaction in a subcritical multiplying pile, with each skip of the stone representing a fission reaction. But if one could imagine skipping a stone that keeps skipping forever, never slowing down or falling into the water, then that could be likened unto a fission reaction in a multiplying pile that has achieved criticality. Of course, in the real world, a stone cannot skip across a lake forever. But criticality can be achieved in a pile of natural uranium and graphite; with great care, high-purity materials, and a penetrating insight into the problem. These are the gifts that Enrico Fermi and his team brought to the challenge of building the world's first critical assembly of material.

3.4 The Relationship between Criticality and Power Level

By March 1942, experiments with the first exponential pile in Chicago were yielding a multiplication constant of 0.94, with a margin of error of plus-or-minus 0.02. That meant that a fission reaction was leading to roughly twenty others before it was quenched in that pile. By mid-May Fermi was reporting that he had achieved k = 0.995 in an improved pile, meaning that each fission was being multiplied about 200 times before the reaction stopped. Confidence was growing that criticality was actually possible.

The implications of a critical pile of material were significant. To produce plutonium for weapons in kilogram-scale quantities require trillions of trillions of neutrons. In a critical nuclear reactor, each fission would lead to another fission indefinitely. But would it produce enough neutrons?

One of the remarkable aspects about criticality, as a physical principle of a nuclear reactor, is it is very different than the power level of the reactor. Criticality can be achieved even when the thermal power generation of the reactor is at a billionth, or even a trillionth of a watt. Explained another way, stating that a reactor has achieved criticality tells you nothing at all about how many nuclear reactions are taking place at a given moment inside the reactor. All that it explains is that each fission reaction is leading to another one. It is not difficult to imagine a nuclear reactor critical

on a single neutron, so long as that neutron is causing a fission that leads to another fission, that state of affairs is correctly described as a reactor that has achieved criticality. But in such a reactor, the power that would be generated from the fission reaction would be far, far too small to measure.

What is important, then, is that a sufficiently large number of fission reactions are taking place at a given moment. The reactors that Wigner's group at the Met Lab were designing were intended to produce plutonium, and for that mission that they needed a great number of fission reactions. Wigner's original design for the Hanford reactors intended for each of them to operate at 500 megawatts of thermal power. Assuming that each fission releases about 200 MeV of energy, that implies that each of those reactors needed to be producing 16 billion billion (16×10^{18}) fission reactions each second. If each of those fission reactions also led to the formation of a plutonium atom, then a 500-MW Hanford reactor could produce a kilogram of plutonium-239 in less than two days.

In principle, there was no difference between a reactor that was critical on 16 billion billion fission reactions per second or a reactor that was critical on a single reaction per second. Both were perfectly valid expressions of the principle of criticality. But from a practical standpoint there was a world of difference between a reactor that could make the material needed at a rate of half-a-kilogram per day and one that took hundreds of thousands of years.

So if there is no relationship between criticality and power level, why was there so much interest in developing the first critical assembly of materials? It was this—if a reactor could achieve criticality then it could likely achieve a very small degree of supercriticality ($k > 1$), and if a reactor was operated at a supercritical state its power level would increase. One fission would lead to more than one fission. In this manner, a reactor that was originally critical at a vanishing small power level (perhaps as low as a trillionth of a watt) could be increased in power until it reached the desired level. Then by adjusting the multiplication of the pile, through the insertion or removal of neutron-absorbing material, it would be possible to "level off" the reactor (from supercriticality back to criticality) at the power level desired. That was one of the responsibilities of the reactor operator—to take a reactor from subcriticality to criticality at some very low power level, then to take the reactor slightly supercritical to increase its power, then to return to criticality to hold the power steady at the desired level for production of plutonium.

An analogy might be made between an airplane flight and a reactor moving from subcriticality to supercriticality. The airplane rolling down the runway and accelerating could be likened to a subcritical reactor whose multiplication constant is increasing from zero to one. When the airplane first lifts off the runway, that could be likened to the reactor achieving criticality. As the airplane climbs in altitude, that could be likened to a supercritical state, where the reactor is increasing in power level. When the airplane reaches the desired altitude, it levels off and maintains altitude. That would be analogous to a reactor returning to criticality from supercriticality at the desired power level.

3.5 The Important Role of the Moderator in a Reactor

When a fission reaction takes place, two fission products and several neutrons are released. Each of these has a large amount of kinetic energy. Although it might seem that a high-speed neutron

would be more likely to cause another fission reaction than a low-speed neutron, just the opposite is true. Neutrons that have been slowed down are much more likely to cause a fission reaction in a suitable nucleus.

The process of slowing neutrons down, or "thermalizing" them, falls to a class of material that is just as important in a nuclear reactor as the fuel. This material is called the "moderator" and its role is to slow down neutrons through collisions with the nuclei of the moderator material without absorbing them.

Since neutrons are slowed down by colliding with atomic nuclei, the most effective moderator for slowing down neutrons is a single proton. The proton and neutron are almost exactly the same size and there is no electric repulsion between them because the neutron has no electric charge. In theory, a neutron can lose all of its kinetic energy in a single collision with a proton, provided it strikes at precisely the right angle. But in reality, since the angle of impact is random, each collision between a high-energy neutron and a low-energy proton only results in about 63% of the kinetic energy of the neutron being lost.

An atomic nucleus with only a single proton is not difficult to find, in fact it is the most common type in the universe. A single proton is the description of the most common isotope of hydrogen, hydrogen-1. Hydrogen is the only element where each of its isotopes have been assigned their own distinct names. Hydrogen-1 is called "protium", hydrogen-2 is called "deuterium", and hydrogen-3 is called "tritium". Each has a single proton and zero, one, or two neutrons.

Protium is the most effective material for slowing-down neutrons, but it has a serious flaw as a moderator. Protium has a significant appetite for absorbing the neutron rather than simply letting it bounce off in a collision. In an ideal moderator material there would be no neutron absorption at all by the moderator. A material that comes much closer to that ideal is protium's rare sister isotope deuterium, sometimes called "heavy hydrogen". A deuterium atom is only present in one out of 6400 atoms of protium in a sample of hydrogen, so while it is naturally-occurring it is very dilute.

Deuterium is not as effective as protium in slowing down a neutron, removing only 52% of the kinetic energy of a high-energy neutron in each collision, but it makes up for that disadvantage by its almost complete lack of neutron absorption, far less than protium.

Any material that contains hydrogen then can potentially serve as a moderator to slow down neutrons. Water, paraffin, and polyethylene are all compounds that contain large amounts of hydrogen that have been used as moderating materials. Although each of these compounds contain other elements, like oxygen or carbon, it is the hydrogen they contain that is responsible for the vast majority of their ability to slow down neutrons. In each case, if the hydrogen in these compounds is replaced by hydrogen where the deuterium content has been increased considerably, the compound is referred to as "heavy", while the same compound using a natural mixture of hydrogen isotopes is called "light". Thus water with unaltered hydrogen is called "light water" and water with highly-enriched hydrogen is called "heavy water". Light water is very effective at slowing down neutrons in a short distance but it consumes some of them. Heavy water is somewhat less effective at slowing down neutrons in a short distance but will hardly absorb any neutrons.

Atomic nuclei heavier than a single proton become less and less effective at slowing down neutrons through collisions, but other materials have been used successfully as moderators because

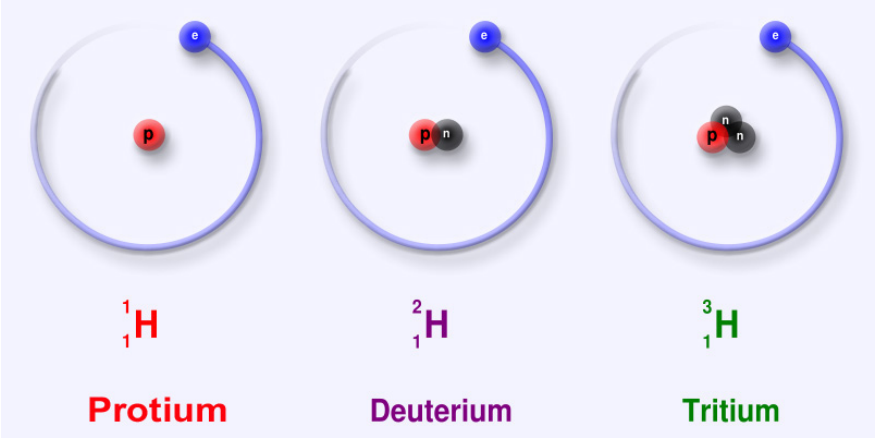


Figure 14: Isotopes of hydrogen. Tritium is artificially produced.

they have low neutron absorption potential. Beryllium is an element that consists of a single isotope, beryllium-9, with four protons and five neutrons. Each collision with beryllium-9 only slows a high-energy neutron by 19%, but beryllium also has a very low propensity to absorb neutrons, and it is an effective moderator in high-temperature applications where water or another hydrogeneous compound is unsuitable. Natural carbon has two main isotopes, carbon-12 which comprises 99% of natural carbon and carbon-13 which comprises nearly all of the rest. Carbon-12 is less effective than hydrogen or beryllium at slowing down neutrons, and each collision between a high-energy neutron and a carbon-12 nucleus only removes 15% of the energy of the neutron. But carbon-12, like deuterium, has a very very low propensity to absorb the neutron, and carbon in the form of graphite can be used at high temperatures.

isotope	energy removed from a high-energy neutron per collision	neutron absorption (relative to deuterium)
hydrogen-1	63.2%	604
hydrogen-2	51.6%	1
beryllium-9	18.7%	17
carbon-12	14.6%	6
nitrogen-14	12.7%	3458
oxygen-16	11.3%	0.4
fluorine-19	9.7%	17

Beyond carbon, few materials are considered as moderators since their effectiveness falls off rapidly with increasing mass. Other light elements are rarely considered as moderators for other reasons. Helium-4 is light and has almost no neutron absorption, but as a low-density gas it is not an effective moderator since it is very difficult to confine enough helium nuclei in a given

volume to appreciably slow neutrons. Lithium has a common isotope (lithium-6) that is a strong neutron absorber, as does boron (boron-11). Nitrogen also has two isotopes, but the common one (nitrogen-14) is quite absorptive of neutrons. Oxygen has little neutron absorption but it is difficult to form compounds with a high density of oxygen. Thus in the first eight elements of the periodic table only three (hydrogen, beryllium, and carbon) are realistically considered as moderators, with the two natural isotopes of hydrogen generally considered separately, leading to the description of "light" and "heavy" compounds of materials that contain hydrogen.

For Fermi's efforts to achieve criticality in an exponential pile with natural uranium, neutron absorption by the moderator had to minimized to the maximum degree possible. In large part this was due to the fact that natural uranium consisted mostly of absorptive material (uranium-238) with very little fissile material (uranium-235). This swiftly ruled out normal hydrogen as a moderator material—protium was simply too absorptive of neutrons. Beryllium was too expensive and rare. The remaining credible options were highly enriched hydrogen in the form of heavy water and carbon in the form of graphite. Graphite was cheap and effective and it was chosen for the exponential pile experiments, even though heavy water was more effective. The cost and scarcity of heavy water ruled it out for early consideration, although Eugene Wigner investigated heavy-water moderators reactors shortly after Fermi achieved success with his first critical assembly.

Early descriptions of nuclear reactors as "piles" came about because Fermi's early experiment literally involved stacking bricks of graphite and cylinders or spheres of uranium in piles under the west stands of the former stadium at the University of Chicago. Later as reactors used liquid water as a moderator the description of a reactor as a "pile" fell into disuse.

4 The Metallurgical Laboratory at the University of Chicago

4.1 Achieving Criticality for the First Time

Wigner's group continued to make progress on the design of the plutonium-producing reactors for Hanford, considering all manner of coolant options and core geometries. In July 1942 Fermi's most recent calculations predicted that one of his geometries could achieve $k = 1.007$. There was light at the end of the tunnel, particularly for young Weinberg, who had wondered whether $k < 1$ was some sort of cosmic barrier preventing the creation of reactors. Weinberg was electrified. [11, pg.16]

Those were heady times for the young physicist, working on the most challenging and important projects in the world.

> We were writing on a tabula rasa. Everything was new and untried. Discoveries came easily. Since graphite was the moderator of choice for the plutonium-producing reactor, most of our computations were devoted to combinations of graphite and uranium, or uranium oxide. In Fermi's exponential pile the uranium oxide was disposed as roughly spherical lumps about 6 centimeters in diameter. But in a high-power reactor, which required a coolant such as swiftly-flowing helium, or air, or even water, such an arrangement was awkward since each uranium lump was separated from its

> neighbor by several inches of graphite. Much better was an arrangement in which the uranium was disposed as long rods bathed in flowing coolant that traversed the entire pile. The rods were regularly spaced to form a two-dimensional lattice. To calculate the multiplying properties of such a two-dimensional cylindrical lattice required some familiarity with Bessel functions. I had encountered these in my work ... (since nerves are long, very thin cylinders), and so I became a sort of minor expert on Bessel functions for Wigner's group.
>
> Under Wigner's guidance we examined lattices moderated by heavy water, light water, and beryllium, as well as by graphite. We quickly realized that the k in an optimal lattice moderated by heavy water was at least 10% higher than one moderated by the purest graphite. Indeed, as we learned in 1945, the German uranium project focused entirely on heavy-water-moderated piles. But heavy water required an isotopic separation of deuterium (heavy hydrogen) from ordinary hydrogen. This separation was much easier than separation of uranium isotopes, but it was still formidable. Although plants for producing heavy water were built as insurance in both Canada and the U.S. in case graphite moderation proved infeasible, heavy water played no role in the production of plutonium during the war.
>
> Life for Wigner's band of theorists was filled with great excitement and seven-day work weeks. [11, pg.18-19]

Fermi's group was getting closer and closer to being able to build a chain-reacting pile, meanwhile Wigner's group was designing the plutonium production reactors for Hanford as if they were certain that Fermi would succeed. Weinberg reported that both groups got along well.

Success for Fermi's group came on December 2, 1942, when their graphite piles, fueled by lumps of natural uranium, first achieved criticality. Seaborg recorded in his journal:

> The first committee member I saw today was Greenewalt. We met late this afternoon in the corridor of Eckhart Hall. As he approached me, I could see from his demeanor he was bursting with good news. The aura of cheerfulness and excitement that he carried with him and the way he held out his hand in greeting told me that this signified more than just taking pleasure in seeing me again. Then when I heard him say he had just come from the West Stands, I understood the reason for his jubilation. **Fermi has produced a chain reaction—the pile is a success!** Greenewalt said that Fermi, Whitaker, Zinn and their crew started the experiment this morning. As the control rods were cautiously withdrawn throughout the day a few inches at a time, Fermi would take the new meter readings, and using his slide rule, would calculate the multiplication factor. Greenewalt said that he and Compton stood on the balcony, alongside Fermi and most of his crew, watching the proceedings. Then at 3:20 p.m., Fermi called for a few more centimeters. The pile became self-sustaining; in a few minutes the output of the chain reaction rose to one watt. Fermi ordered the reaction stopped, and everyone was tremendously relieved to see that the activity could be extinguished by shoving the rods back into place. The reaction took place without requiring as much

material as Fermi earlier anticipated (and thus he was two weeks ahead of schedule), and it was not necessary to use the balloon to exclude air from the pile. However, he did use almost six tons of uranium metal, 50 tons of uranium oxide, and 400 tons of graphite, nearly as much as predicted. (I learned later that Zinn, Anderson and their pile builders reached a point where they knew the pile would be self-sustaining very early this morning but did not proceed to this historic point, leaving this dramatic act for a more convenient time later in the day.)

Of course we have no way of knowing if this is the first time a sustained chain reaction has been achieved. The Germans may have beaten us to it. I wonder, are they aware that U-233 can be made from Th232 and [plutonium]-239 from U-238 in a chain-reacting pile and that either of these isotopes can be used in a fission bomb? And if they have a pile that chain-reacts, would they use it to generate power or to produce vast amounts of radioactivity as a military weapon? One thing is certain; although Fermi has demonstrated that we now have a means of manufacturing [plutonium]-239 in copious amounts, it is the responsibility of chemists to show that the [plutonium] can be extracted and purified to the degree required for a working bomb. [7, pg.390-391]

Fermi had achieved the goal of the first controlled nuclear reactor. But Seaborg was wise enough to wonder if they had actually been the first. In the final days of the war, the Alsos Mission was an Allied team deployed to Germany to find any evidence of a nuclear weapons program. In the small town of Haigerloch, in the basement of a castle, they discovered an odd arrangement of uranium metal cubes suspended by wires in heavy water. It was never even close to successful operation and certainly not comparable to Fermi's achievement in Chicago.

4.2 Oak Ridge and the X-10 Reactor

Although the reactor group under Wigner and the chemical separation group under Seaborg labored in the Metallurgical Laboratory at the University of Chicago, the headquarters for the Manhattan Project was located in northeastern Tennessee, not far from Knoxville. It was here that the Army established a secret city, now called Oak Ridge, where the primary efforts of isotopic separation of uranium would be concentrated. Another aspect to their efforts was to construct a much larger pile of graphite and natural uranium than the rather small one that Fermi had briefly built in Chicago. This pile is known today in Oak Ridge simply as the "Graphite Reactor" but during the war was referred to by the code-name of its coordinates, X-10.

The X-10 pile was intended to operate at a power level vastly greater than Fermi's pile, and to operate for an extended period of time. It was meant to produce enough plutonium in its uranium fuel in order to test Seaborg's ideas about plutonium separation. The land that would host the reactor had been acquired by the Army in the fall of 1942 through condemnation and eviction of the farmers who lived there. Construction began shortly thereafter. In late September 1942, Seaborg took his wife Helen on a vacation in eastern Tennessee and as part of that trip they toured the land that would soon host "Pile Number 2", or the X-10 reactor. In correspondence on October 26, Seaborg instructed that space be made available in the new pile to irradiate samples of thorium

to produce uranium-233. [7, pg.314] He anticipated that the uranium-233 produced would be on the order of milligrams rather than the micrograms that they could produce in the Berkeley cyclotron, a thousand-fold improvement, but still a miniscule amount of material. To produce these small amounts of uranium-233 would require exposing several tons of thorium to the neutrons of the pile.

Figure 15: The X-10 "Graphite Reactor" under construction at Oak Ridge in 1943. This reactor would produce the first milligram-level quantities of uranium-233 that allowed understanding of its nuclear characteristics, and the realization that thermal breeding—and the nearly complete utilization of thorium as an energy resource—was possible.

In early November, Seaborg had grown very worried about the purification levels that would be required in the new plutonium that they would be producing for the war effort. Impurities in the plutonium, particularly among the light elements like boron or beryllium, could lead to neutron emission as alpha particles from plutonium decay smashed into the impurity nuclei. He was so concerned that he wrote to Robert Oppenheimer. [7, pg.325-326] A week later Oppenheimer wrote back, and his words were not particularly reassuring. [7, pg.337-338] Due to the far greater radioactivity of plutonium-239 relative to uranium-235, the tolerance to impurities would be orders of magnitude lower than in uranium-235. Oppenheimer asked "how bad is 23?" referring to uranium-233 by its code name. It is doubtful that uranium-233 would have offered much improvement over plutonium-239 in this situation, since its half-life was only about six times less. What

wasn't mentioned then, but would later be discovered, is that the U-232 contaminant of U-233 would shower any sample with far more alpha particles than plutonium-239 did.

By December 9, 1942, Seaborg was still excited from Fermi's recent accomplishments in building the first chain-reacting pile and wrote to Stoughton in anticipation of producing larger quantities of uranium-233 by loading thorium in the periphery of the X-10 graphite pile. The production rate would be orders of magnitude greater than they had achieved heretofore from the cyclotron, but still only a tiny fraction of the production rate of plutonium in the pile. Seaborg continued to view uranium-233 production as a credible alternative to plutonium-239 production, despite its lower production rates. The main advantage of U-233 in his mind was that, as uranium, its chemistry and metallurgy were well understood whereas those properties in plutonium remained a great mystery. He was also far more confident that uranium could be extracted from a pile at high purity than plutonium. [7, pg.405]

By early February, Stoughton had left Berkeley to join Seaborg's Metallurgical Laboratory team in Chicago. Seaborg had hopes that Stoughton would carry on his U-233 research, but noted in his diary that plutonium work would likely take priority initially. [7, pg.512]

On Febrary 16, 1943, Seaborg sent another memorandum to Oak Ridge that space be left in the new pile for "50-100 pounds of thorium salt, which would produce enough U-233 to make possible further studies of its nuclear properties." [7, pg.527] Despite Stoughton's addition to the small U-233 effort in Chicago, progress slowed even as it was accelerating on plutonium. Most of the chemical group's effort consisted of preparing thorium slugs for irradiation in the new "Pile number 2" that was being built in Oak Ridge (the X-10 reactor).

4.3 Wigner Accelerates his Effort

As the new year of 1943 dawned, Wigner's reactor group was finishing their design for a 500-megawatt water-cooled reactor that could be built at the Hanford site in Washington ("Site W") and would produce half-a-kilogram of plutonium per day. Their design report was called CE-407, "Preliminary Process Design of Liquid-Cooled Power Plant Producing 500,000kw" and it was issued on January 9, 1943. After that point, the design was essentially handed-off to the DuPont Company for the actual detailed design, construction, and operation of the new plutonium-producing reactors. [11, pg.26]

Seaborg noted on May 26 that Wigner had called the group's attention to articles in the scientific literature that indicated that the Germans were "running neck and neck with us in 1941 and 1942" in the investigation of neutronic behavior in various materials—a definite precursor to any work on nuclear reactors. [8, pg.32] Wigner would simply not allow himself to entertain the possibility that Germany was doing anything less than proceeding at maximum effort towards a nuclear weapon, and he was driven by the unrelenting need to protect the free world by achieving the technology first. As Weinberg recorded:

> In the summer of 1942, Compton called Fermi, Wigner, and me to his office. The issue: when could we expect the German atomic bomb? Wigner went to the board and figured about six months to build the reactor, three months to extract the plutonium,

> two months to build the bomb, and maybe a few months for slippage. **We might receive the German bomb sometime in 1943!** Wigner's error was to assume the German project to be manned entirely by Wigners. [11, pg.30-31]

Wigner was very frustrated by DuPont and what he perceived as their sloth and indifference in implementing his design for the new reactors. Having largely completed his design of the Hanford plutonium-production reactors, Wigner turned his attention to other ways he could accelerate the war effort through new reactors. Natural uranium was their only feasible reactor fuel; with this restriction the feasible choices for moderating material were small and one had already been used for Fermi's pile and for the Hanford reactors—graphite. The other option was heavy water. Heavy water was exceptionally expensive but it was a more efficient moderator than graphite, and plutonium production piles using heavy water could be much smaller than similar graphite piles. Furthermore, there would not be a need to physically segregate the uranium from the moderator, as was done in graphite. Heavy water was such a good moderator that a uranium compound that was soluble in water could be dissolved in the heavy water and used directly. This had profound advantages over the current techniques that they had proposed, where uranium metal would be welded in an aluminum can and physically pushed through the structure of the Hanford reactor graphite block.

This was the origin of the "aqueous homogeneous reactor" which was the first idea for a reactor with fluid fuel rather than with solid fuel. Weinberg enthused about the potential of such a reactor:

> We calculated the k and the critical size of both homogeneous and heterogeneous (lattice) piles moderated by [heavy water]. Since heavy water is a much better moderator than graphite, many fewer neutrons are lost by capture in ^{238}U while slowing down in [heavy water] than are lost in graphite. It is therefore unnecessary in a [heavy water] pile to remove the neutrons during their moderation away from the uranium fuel—that is, it is unnecessary to arrange the uranium in a lattice. The uranium can be mixed homogeneously with the heavy water. We estimated that a homogeneous mixture of [heavy water] and natural uranium would have a k of about 1.08, compared to a k of about 1.2 in an optimized lattice.
>
> Both Urey and Wigner were intrigued by the possibility of a plutonium-producing pile consisting merely of a solution or slurry of uranium in heavy water. Gone would be the thousands of carefully machined uranium slugs; gone, too, the intricate system of piping required to feed water into each process tube. Experiments were therefore undertaken at Chicago during 1943 and 1944 to find either a stable solution or a slurry of uranium in water. Alas, this was not easy, although aqueous homogeneous piles based on uranyl sulfate, on uranyl nitrate, and on stable uranium dioxide slurries were developed several years later. **I myself became bitten with the "homogeneous" bug, a fixation I have never fully recovered from!** [11, pg.32-33]

The project to develop a heavy-water plutonium production reactor, code-named "P-9", began in early 1943 but did not reach a conclusion during the war years. Nevertheless, it was the beginning of thought by Wigner about fluid-fueled reactors and their advantages over solid-fueled

reactors. Later these thoughts, when coupled with Seaborg's discoveries about uranium-233 and his research with Stoughton on the fluorination of thorium, would form the basis for the invention of the thorium-fueled molten-salt reactor.

4.4 The X-10 Reactor Achieves Criticality

Even as the first criticality of the X-10 reactor drew near, Dr. Compton, the leader of the Metallurgical Laboratory, was reducing the already-small emphasis on uranium-233. He felt it was not a significant priority because there was no way to make militarily-significant quantities of uranium-233 before 1947. [8, pg.134] This was because the only way to make any amount of uranium-233 would be to expose thorium to neutrons in one of the plutonium-producing piles. The amount of "spare" neutrons available in these piles was incredible compared to the tiny amount that had been produced by the Berkeley cyclotron, but very small compared to the amount that would be producing plutonium. There was simply no comparison between the rates of production. If thorium had had a naturally-fissile isotope, akin to uranium-235 in natural uranium, then dedicated thorium piles producing U-233 might have been possible. But this was simply not the case, and thorium/U-233 research by necessity took a back seat to the development of plutonium.

On November 4, criticality was achieved for the first time in the X-10 reactor at Oak Ridge. It had a far greater thermal power than Fermi's Chicago pile a year before, and the X-10 reactor would be the first to make gram-scale quantities of plutonium. [8, pg.234] Within the month, thorium carbonate slugs had been loaded into various locations in the X-10 reactor and were projected to produce about one milligram of uranium-233, vastly more than had ever been previously available. [8, pg.254] But even on the edge of this exciting development, the Metallurgical Lab was pulling back even further from uranium-233. Dr. Compton addressed the group on November 17, commending their achievement in getting the X-10 reactor operational, but emphasizing the need to conserve manpower throughout the program. He said that due to the cancellation of Wigner's heavy-water "P-9" project, there would be no way to get useful quantities of uranium-233 and so the research work would be discontinued. [8, pg.258]

Seaborg was not pleased with this development. He wrote a memo to a colleague at the Clinton (Oak Ridge) Labs where he speculated that Germany was developing weapons based on U-233 rather than on plutonium. The German interest in heavy water was part of the reason he believed this was taking place.

Even as he was worrying about German uranium-233, Seaborg was worried about the myriad ways that the Chicago project's emphasis on plutonium could run aground. On November 25, he ran into Leo Szilard and mentioned his concern that not every interaction between plutonium-239 and a neutron would result in fission. Sometimes, he anticipated, the plutonium-239 nucleus would simply absorb the nucleus and form plutonium-240, and this nucleus, with its even number of nucleons, would have a greater probability of spontaneous fission. Spontaneous fission released neutrons that could cause nuclear weapons to "fizzle" before achieving the ideal geometry for detonation. Szilard expressed doubt that the Pu-240 potentiality was a problem. [8, pg.266-267] It turned out that he was very wrong, for Seaborg was correctly anticipating the most severe issue that would face the Metallurgical Laboratory project, and the new X-10 reactor would soon uncover it.

Seaborg may have seen uranium-233 as a back-up plan to the plutonium effort, but the leaders of the Met Lab did not share his view. By December 15, Compton had informed Seaborg that General Groves had directed that the Met Lab halt until further notice all work directed toward the development of a uranium-233 process and discontinue all work on its chemical separation. [8, pg.289] There was some worry about the thorium carbonate cans that were in the X-10 reactor—should they be removed? But it was decided to leave them in the neutron flux until April, when they anticipated that approximately 15 milligrams of uranium-233 would have been produced. [8, pg.301]

Seaborg tried once again to persuade Manhattan Project leadership that a secondary effort in uranium-233 to receive serious consideration. On January 27, 1944, he wrote an extended memo to Dr. Compton giving his reasons why he thought that the Germans might produce a nuclear weapon from uranium-233 in the near future, and suggested that these reasons be communicated to the highest levels of command in the Allied powers. He reasoned that (1) the Germans were continuing to procure heavy water from a manufacturing facility in Norway; (2) the chemical separation of uranium from thorium was easier to research and accomplish than the separation of plutonium from uranium; (3) uranium-233 would have a greater tolerance for impurities due to lower alpha activity than Pu-239; and (4) thorium raw materials were likely available to the Germans. [8, pg.398-399]

One might ask, why was Seaborg pushing so hard for a material development program based on uranium-233? There were several reasons. The first was that the chemistry of uranium was well-understood. Seaborg realized that there would be no difference between the chemistry of inexpensive natural uranium and scarce uranium-233, therefore he could develop all of his chemical separations processes with natural uranium at low cost and with great confidence. At this time, only a few months after the X-10 reactor achieved criticality, plutonium was an exceedingly scarce commodity, available only in the microgram quantity from the Berkeley cyclotron. Surely this availability of plutonium must have been a dominant concern to a chemist like Seaborg, even if the potential for manufacturing multi-kilogram quantities of plutonium was far better than for uranium-233.

Another concern was the purity that would be required of the plutonium, both chemically and isotopically. It was terribly difficult to assess the performance of a chemical separations process when one was dealing with micrograms of a substance versus kilograms. Opening another chemical "front", so to speak, with uranium-233 meant that Seaborg's group could immediately begin developing separations processes between thorium and uranium without having to wait for any products to emerge from the X-10 reactor.

Finally, Seaborg saw in the German attempt to obtain heavy-water supplies an echo of Wigner's P-9 experiments, and a way to make a much more compact reactor than with graphite and natural uranium. He knew that a P-9 style pile would be almost necessary in order to produce any meaningful quantity of uranium-233 from thorium. But the same advantages of P-9 style pile would also accrue to plutonium development, and indeed, many years later the United States produced considerable quantities of plutonium from the heavy-water-moderated production reactors at Savannah River in South Carolina.

By February 29, Compton had written back to Seaborg, indicating that he had communicated

the contents of the memo on potential German U-233 production to General Leslie Groves, the head of the Manhattan Project, as well as others in the Army command. But Compton also added his opinion that any U-233 production effort by the Germans would probably require more time and effort than a Pu-239 production attempt, and that he saw no strong reasons to believe that the Germans were at that time actually engaged in a program of U-233 production. [8, pg.456] In this case, Compton's suppositions were entirely correct. The Germans had no effort to develop U-233 as a weapons material. They were trying to procure heavy water, but it was for their own low-scale effort to create a critical assembly of uranium and heavy water in order to produce plutonium.

This rare misstep by Seaborg may be explained by considering his approach to the problem. As a chemist, he was tackling the challenge of separating a newly-discovered element from scores of others at high purity. The prospect of shifting that challenge to an already-understood element like uranium, available in large quantities, must have been tempting. But others like Wigner and perhaps even Compton would have seen it differently. To them, the challenge of producing fissile material from natural uranium, which contained both the fissile material to sustain the reaction (uranium-235) and the fertile material to create the product (plutonium-239) they desired (uranium-238) would have seemed much more feasible than to attempt the same feat with thorium, which had no fissile isotope. Any attempt to create significant quantities of uranium-233 would have to rely on neutrons created from the fission of uranium-235, and these neutrons would be far more likely to create plutonium-239 than uranium-233 in any practical pile arrangement using natural uranium as a fuel.

Nonetheless, Compton did encourage Seaborg to continue to interact with another group of researchers in Canada that were looking at uranium-233 as a fissile material. [8, pg.456]

Seaborg's concerns about plutonium were not without a basis; in fact, one could argue that the plutonium situation that actually developed was far worse than Seaborg feared at the time. But the challenge from plutonium that actually materialized was far more fundamental than extreme chemical purification, and could not be addressed by Seaborg's considerable talent in chemistry.

4.5 Conceiving the Breeder Reactor

In the spring of 1944, Alvin Weinberg remembered that the mood in Wigner's reactor design group was expansive because they had completed their primary responsibility of designing the plutonium production reactors at Hanford. They began to speculate about "new piles" and formed an informal group which they called the "New Piles Committee." [11, pg.39]

A particularly notable meeting of this group took place on the morning of April 26, 1944, when Enrico Fermi began to describe his ideas for a nuclear reactor that would produce more nuclear fuel than it consumed. [14, pg.1] His argument was centered in the nuclear properties of plutonium-239 and uranium-235 as they were then known. Through a mathematical derivation, Fermi indicated that he thought it possible to build a reactor using plutonium where enough neutrons would be produced through fission to not only replace the fuel consumed, but to generate additional fuel. Fermi referred to "mother plants" that would produce excess plutonium in order to feed a series of smaller plants. He indicated that there might be a concern that during the shipment of plutonium it might fall into the wrong hands, but those concerns were beyond the scope of their discussion.

All of the reactors that Fermi and Wigner's group had worked on up to that point had been reactors where moderating materials such as graphite or water had been used to slow down neutrons so as to maximize the probability that fission would take place. Reactors that used moderators to intentionally slow neutrons are now called "thermal-spectrum" reactors, because the energies of their neutrons follow a spectrum of values that correspond to thermal equilibrium in the reactor. Put another way, the neutrons are at the same temperature as the materials around them, bouncing about until they are consumed through absorption.

Another reason why a thermal-spectrum reactor was the only option for the Manhattan Project was the use of natural uranium as a nuclear fuel. Its very low fissile content (only one atom out of 137 in uranium is fissile) meant that every attempt had to be made to maximize the probability of fission, and that dictated that neutrons be slowed-down, or "thermalized," for in this condition the probability of fission was maximized.

Fermi imagined two large departures from the current state of reactor design. The first was that he proposed to use pure plutonium as the fuel in the reactor. This would necessitate a significant diversion of material from the weapons program in order to simply start the reactor. The other departure he hoped would make it worth the cost. The reactor would not slow down the neutrons produced in fission; instead, it would make every effort to keep them at the high speeds with which they are created. This concept would come to be known as a "fast-spectrum" reactor, and the reason to take this approach would be that when plutonium is struck by fast neutrons, it fissions nearly every time. Non-productive creation of plutonium-240 would be drastically reduced, and the fact that each plutonium fission produces nearly three neutrons would enable the fast reactor to make twice as much plutonium as it consumed.

In this way, through the use of plutonium fuel and intentionally fast neutrons, Fermi hoped to build a reactor that would double the inventory of plutonium inside it on a regular basis. This interval of time that would be necessary to double its plutonium inventory was called the "doubling time," and Fermi and others hoped it would be as short as two years.

Admittedly, the engineering challenges of creating a fast-spectrum reactor were far greater than the thermal-spectrum reactors at X-10 or Hanford. Any coolant that might slow down the neutrons significantly could not be used. That ruled out water as a coolant. Instead, early attention turned to metallic bismuth as a coolant, or possibly, molten metallic sodium. Liquid sodium metal had a number of advantages over bismuth, but like other pure alkali metals[5] it was very dangerous to work with since it would burst into fire upon contact with air or water. Its advantages were that it was highly thermally conductive and could operate at temperatures high enough to drive a steam turbine yet stay at low pressure.

A sodium-cooled fast reactor could therefore be a power-generating reactor, unlike the huge reactors under construction at Hanford. These mighty machines, despite their impressive thermal output of 250 megawatts, used water coolant that was circulated at such a rate that its temperature at discharge was far too low to raise steam. Thus all of the thermal power produced was wasted, and no fraction of it could be turned to electricity.

The sodium-cooled fast reactor might be able to make power at the same time it was doubling

[5]Metallic lithium, sodium, potassium, rubidium, and cesium are all highly chemically reactive with air and water, with the reactivity increasing from lithium to cesium, as the outermost electron is more loosely bound.

its plutonium inventory. Fermi doubted that such a reactor would be an economically competitive source of power, although he said that he looked upon the use of nuclear power to heat cities with sympathy. Producing more plutonium was a far more compelling objective.

Fast reactors had a basic disadvantage over thermal reactors, however. The cross-sections of nuclides were far smaller to fast neutrons than thermal neutrons; achieving the same rate of power generation would require far more atoms of fissile plutonium in a fast-spectrum reactor than in a thermal-spectrum reactor. A graphical depiction of this disparity is shown in figure 16, where the thermal fission cross section of a single atom of Pu-239 (1303 barns) is compared to 698 atoms of Pu-239 with their fast fission cross sections of 1.87 barns each.

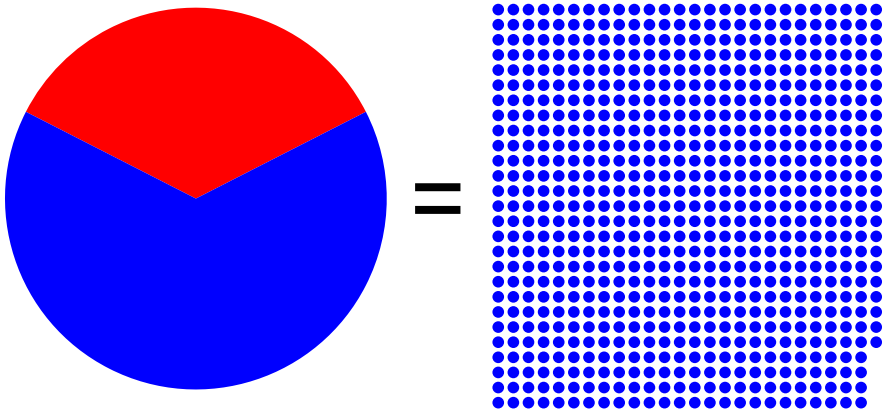


Figure 16: A single plutonium-239 atom has the same fission cross section to a thermal neutron as nearly 700 plutonium-239 atoms to a fast neutron.

That evening, Fermi continued to expound his ideas about fast-breeder reactors, as Alvin Weinberg remembered:

> Although the idea of the breeder was in the air almost from the beginning, it was "officially" recognized in an evening lecture Fermi gave to the entire staff in the Eckhart Hall auditorium. Fermi explained the principle of breeding and noted that breeding was more likely if it was based on fast neutrons, as in a bomb, because the number of neutrons per fission exceeded 2 by a very comfortable 0.5 or more if the fissions were induced by fast neutrons. None of this was really new to Wigner's group, but as usual Fermi held his audience with his lucid exposition. [11, pg.39]

Soon this innovative reactor concept received the name by which it has come to be known:

> [Szilard], Wigner, and I were discussing Fermi's lecture while walking along the sidewalk that ran from Eckhart Hall to Jones Laboratory. I can't remember who said that we needed a name for a pile that produced more fissile material than it burned; it was probably Wigner. Szilard thought for a moment and said something like, "Let's call it a breeder." And "breeder" it has been ever since. [11, pg.39]

Two days later, on April 28, the New Piles Committee met again to discuss the availability of uranium and thorium, and how the prospect of a "breeder" reactor might affect the prospects for future utilization of energy. [15, pg.1] Morrison felt that a fission reactor merely as a source of "unspecialized energy" would have great difficulty competing economically against existing sources of energy, particularly inexpensive hydropower and coal.

The crust of the Earth had about 4 parts-per-million (ppm) uranium and three times as much thorium, which represented a huge yet diffuse resource. Seawater contained dissolved uranium but no thorium of significance, due to a difference in their chemical properties and solubilities. A breeder reactor would make it economical to recover uranium or thorium from the crust of the Earth and consume it for energy at a net gain. Morrison used a comparison with the costs involved in gold mining, a far more scarce resource. Several countries contained good uranium resources, including the Congo, Czechoslovakia, Canada, the United States, and likely Russia. Thorium was practically everywhere.

But if only the rare uranium-235 was used for energy, as was the case for thermal-spectrum nuclear reactors fueled by natural or enriched uranium, then the uranium inventories of the US and Canada would only provide enough energy for nine months of demand. The group came to the inescapable conclusion that only breeder reactors would enable nuclear energy to be a sustained option as an energy source.

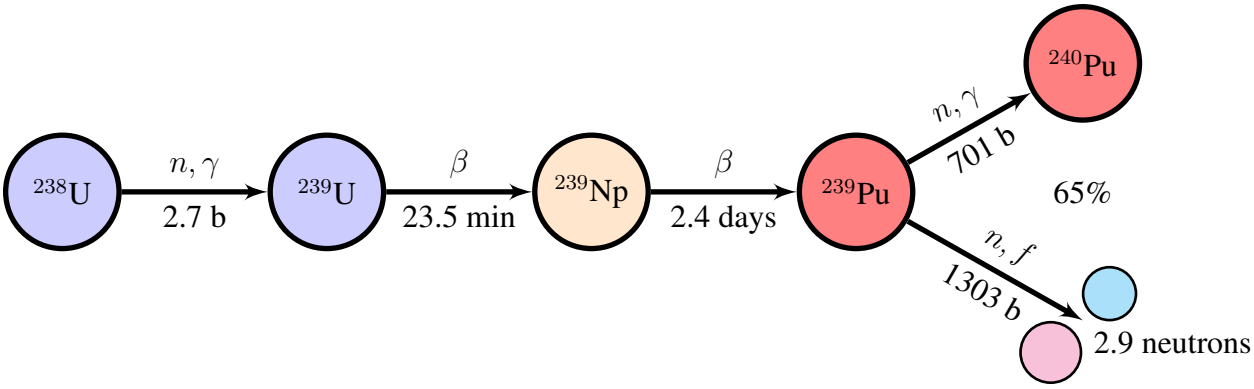
Although the excitement about breeder reactors was significant, much of it was premature. Physicists were still deciphering the important nuclear properties of each of these materials, particularly their propensity to absorb a neutron rather than fission, to such a degree that committing to any path was premature. At that time, the idea that only a fast-spectrum reactor would be able to consume all of the energy of uranium or thorium was strongly believed, and time would bear that out to some degree. But a remarkable property of thorium—one that would substantially distinguish it from uranium—still awaited discovery, and this discovery would open up opportunities for the realization of energy from thorium that simply were not possible with uranium.

4.6 The Plutonium-240 Scare

On January 1, 1944, Glenn Seaborg recorded some of his feelings about the status of his work and its prospects for the future:

> The beginning of 1944 finds our Project deep in the problems of plutonium production, extraction, and purification. This vast involvement with a secret, synthetic element unheard of not much longer than two years ago and unseen until sixteen months ago in August 1942, would seem incredible to the outside world. Moreover, the means of producing plutonium in copious quantities—the chain-reacting pile—became operational just one year ago. I thought about these matters today when we received our first shipment of plutonium from Clinton Laboratories—1,500 micrograms! It equals almost the total amount of plutonium produced by all previous cyclotron bombardments. It is hard for me to remain nonchalant when I realize that before the end of February, production of plutonium will increase more than a thousandfold and gram quantities will then become available. [8, pg.326]

The X-10 reactor had the ability to make roughly a million times more plutonium, over a given time, than the Berkeley cyclotrons, and by early 1944 there was enough chemically-extracted plutonium for chemists at Los Alamos to extract it and to assess its nuclear properties. To their horror, they discovered that Seaborg's previous fears about plutonium-240 had been well-justified. The first concrete evidence of the problem was reported by Enrico Fermi in a meeting held on May 19. Fermi had been measuring the nuclear cross-sections of U-235 and Pu-239. He now had one gram of plutonium dioxide that had been produced by the X-10 reactor. [9, pg.25-26]



The basic issue concerned a quantity called the "branching ratio" by the Chicago group, which dealt with the fact that a fission reaction was not always the outcome when a neutron struck a fissile nucleus. Sometimes the fissile nucleus simply absorbed the neutron and grew by one atomic mass. Fermi had wanted to know how often this happened in plutonium-239, and the answer was very troubling. In the environment of the X-10 reactor, or in the reactors that were under construction at Hanford that would produce the plutonium needed for weapons, the plutonium-239 would simply absorb a neutron in roughly one out of every three neutron interactions. Plutonium-239 would become plutonium-240, which is not fissile and likely had a relatively high rate of spontaneous fission due to its even atomic mass number.

Ironically, within just a few weeks of this meeting Seaborg and his colleague John Willard traveled to Hanford, Washington, and toured the areas where the huge plutonium-producing reactors were under construction. Seaborg reported:

> It is an awe-inspiring experience to see the thousands of workmen busily engaged in the building of these complicated edifices. These are located in a vast expanse of area (almost 500,000 acres in all) with the piles near the Columbia River for cooling purposes and the extraction plants somewhat removed. To reach these areas we drove over some of the flattest, most lonesome territory I have ever seen.
>
> For lunch we were taken into the Hanford Camp where the construction workers live in rows of barracks, tents, and trailers stretched out in all directions. We ate in the largest mess hall I have ever been in—a sea of faces all being well-fed in shifts. We were told that there are some 40,000 residing at Hanford... [9, pg.41]

The success of this titanic construction effort was now in jeopardy based on the results of Fermi's measurements of the properties of plutonium.

Figure 17: Construction of the first plutonium-producing reactor (B reactor) at the Hanford site in Washington state in 1944.

While Seaborg and the other Met Lab personnel continued to wait for further results from Fermi's team, a small amount of research continued on uranium-233, sustained to some degree by continuing Canadian interest in the topic. A delegation of Canadian nuclear scientists visited Chicago in early June and General Groves had determined that Met Lab personnel would be permitted to discuss the properties and possibilities of uranium-233 with them, although any discussion of plutonium and its manufacture, properties, and purification was still forbidden. [9, pg.286] The Canadian group, code-named "Evergreen", was particularly interested in obtaining slugs of thorium carbonate that were currently being irradiated in the X-10 reactor in Oak Ridge. [9, pg.8] They intended to attempt the extraction of either protactinium or uranium from these unique samples.

In early July, General Groves issued a directive to the Metallurgical Project to freeze their current level of personnel and to consider ways to reduce personnel requirements. This directive threatened any attempt by Seaborg to open up another line of chemical investigation into uranium-233 extraction and purification. Groves informed the Chicago group that personnel reductions would begin in September and would be completed by January. Even as these directives were being communicated to the group, Groves sent another letter specifically asking for recommendations about the use of uranium-233. At that time, about sixty "slugs" of thorium carbonate were in the X-10 reactor undergoing exposure to neutrons. Fermi wanted extracted uranium-233 so that he could conduct similar measurements on it to those that he had been doing with plutonium-239. There was discussion as to where the chemical extraction would take place—at the chemical facilities in Oak Ridge, or would it be done by the Evergreen research team in Canada? [9, pg.124-125]

Seaborg's tiny team for uranium-233 research had been decimated by the cuts that Compton had specified at Christmas 1943, and now consisted only a single man, Leonard Katzin. [8, pg.296] But on July 10, Katzin began preparing for the chemical extraction of the uranium-233 that they anticipated had been accumulating in the thorium slugs being irradiated in the X-10 reactor. [9, pg.128] Katzin planned to extract the uranium directly, without any attempt to remove the protactinium precursor. [9, pg.136] There was a potential concern with this plan. Despite their best attempts, the thorium that had been loaded into the reactor had not been completely purified of any natural uranium. Whatever tiny quantity of natural uranium had been present in the thorium slugs would be removed with the uranium-233 that had accumulated during the time in the reactor. Assessing how much of the uranium was uranium-233 (from natural thorium) and how much was uranium-238 (from natural uranium) would be important.

Construction progressed at the Hanford site, with the anticipation that each ton of uranium would yield between 5 and 50 grams of plutonium. They anticipated that the first production reactor at Hanford would go into operation in September, and that it would not take long for the reactors to achieve their full rated power levels.

On July 17, an evening meeting of the Metallurgical Project brought devastating news for all the personnel involved. Seaborg reported:

> The meeting, however, concentrated on something that came up which was much more immediate. Robert Oppenheimer, who was attending the Board Meeting from Los Alamos, announced that E. Segre, O. Chamberlain, and G. W. Farwell have found strong evidence for the existence of the plutonium isotope Pu-240, which undergoes decay by spontaneous fission! This was found in the neutron-irradiated Pu-239 from the Clinton pile that we had purified for them.
>
> It should be noted that this disclosure came as a great shock to everyone. However, over a year ago in my "Report of Month Ending March 15, 1943, Special Chemistry of 94," (CK-514), I had written about the possibility of an n, γ reaction on Pu-239 giving the isotope Pu-240. I had said that a possible spontaneous fission decay of this isotope would seriously impinge upon our ability to use Pu-239 as intended. Now it was learned that indeed this reaction takes place; and that since the neutron flux in the Hanford piles would be so high, Pu-240 would be produced in so great a relative

> abundance that the neutrons resulting from its spontaneous fission would overshadow those from any α, n reactions on impurities that might be present in Pu-239.
>
> Because of this new development, Site Y [Los Alamos] will now have to rethink how it will proceed in the design of a plutonium bomb. Furthermore, it was decided at this Board Meeting to demobilize Thomas's staff which is handling the coordination and general direction of the chemistry, purification, and final metallurgy of Pu-239 because the planned extreme purification of plutonium would be futile—this could not prevent the emission of the unwanted neutrons. This meant that there would be no further monthly meetings at the Met Lab on this aspect of the chemical program. [9, pg.145]

Alvin Weinberg also remembered the events of this time:

> ^{240}Pu undergoes fission spontaneously and therefore emits neutrons. Thus, the plutonium produced at W was bathed in a background of stray neutrons. A "gun-type" bomb in which the separate parts of plutonium were brought together rather slowly would likely be triggered by a neutron from the ^{240}Pu before it reached the compact state needed for a maximum explosion. A plutonium bomb would therefore always predetonate! The whole approach to a bomb based on plutonium was imperiled by the spontaneous fission in ^{240}Pu. [11, pg.37]

This was precisely what Seaborg had feared, and the actual situation was far worse than his previous fears about plutonium purification. In that instance, in November 1942, he had been concerned that light elements impurities with the plutonium would emit neutrons under alpha-particle bombardment from plutonium decay. Hopefully, an effective strategy of plutonium purification would address this concern. But the discovery of plutonium-240 was far more troubling. Here was a contaminant that could never be eliminated, for it had precisely the same chemical composition as the desired plutonium-239. Seaborg and his group despaired that the central effort of the Met Lab had been in vain, and that the output of the future Hanford reactors would be unusable.

None of the efforts of the Met Lab could salvage the situation, but an innovation in weapon design at Los Alamos was the only hope. In 1943, Seth Neddermeyer, a physicist from Caltech, had proposed using an implosion technique to detonate a nuclear weapon rather than the technique which was being pursued at the time, which was to bring two subcritical pieces of fissile material together rapidly to form a critical mass. This latter technique was called the "gun-type" method and was considered to be suitable for both highly-enriched uranium and plutonium. Fermi's results on plutonium-240 contamination indicated that there was little chance that the plutonium that would be produced by the Hanford reactors would be suitable for the gun-type technique.

The implosion technique used shaped explosive charges surrounding a spherical mass of fissile material to compress it extremely rapidly into a supercritical configuration that would detonate. It was challenging, but it offered the promise of being far less susceptible to difficulties from plutonium-240 contamination. Another important advantage was that it required far less fissile material for a given yield. It would require the Los Alamos weapons design team to become experts in the precision arrangement and detonation of shaped explosive charges. [1, pg.245-247]

But for Seaborg's chemical purification team, there was little to be done. On July 19th, he was approached by two of the leaders of the Met Lab and told that his purification program was no longer needed.

> I was standing in the hall in front of my office as they approached. They said that Compton had agreed I should be given the reason but that I was not to tell others. I said they didn't need to tell me the reason—**I assumed the spontaneous fission rate of Pu-240 has been found to be so high as to overshadow the neutrons from the α, n reaction.** I went on to say that, since no one has given me this information, I feel free to pass my interpretation on to my men. [9, pg.147]

On July 28th, the director of the Met Lab, Samuel Allison, sent a memo to Compton reviewing the situation on the Met Lab and its personnel:

> "Probably next highest priority will be accorded studies of the basic chemistry of plutonium and to methods of extraction of 23 studied on a laboratory scale. **The recent abandonment of attempts to reach high purity in our product has, in my opinion, increased somewhat the interest in 23. [uranium-233]** This is because we are now committed to only one method of final assembly; and if this should fail, the military usefulness of our product is doubtful. **Under these conditions we would immediately give consideration to the conversion of our product to 23.** Work on extraction of a few milligrams of 23 to be tested at the Argonne Laboratory will probably carry over into the post-October period. [9, pg.163]

The "failure" of the plutonium-manufacturing project, as then seen by the Met Lab personnel, was increasing interest in uranium-233 as a possible fall-back position. Met Lab personnel had little insight into the techniques for implosion-type weapons that were being developed at Los Alamos, and they could not foresee that the successful development of this approach would lead to successful use of the plutonium that they had helped produce.

Seaborg confided to his collegue Roy Post the futility of the plutonium situation as seen from the Chicago perspective on February 28, 1945:

> ...as to how much Pu-240 could be tolerated in purified plutonium for Los Alamos, I said they have no choice in the matter; whatever is produced must be tolerated. [9, pg.521]

4.7 Realizing the Unlimited Potential of Thorium

Leonard Katzin and his very small team of chemists began working after hours on August 2, 1944, to extract the small amount of uranium-233 from the thorium carbonate slugs that had been irradiated in the X-10 reactor for many months. [9, pg.169] By August 9th they were nearly finished. They anticipated recovering about 5 milligrams of uranium, which despite its seemingly tiny mass represented a thousand-fold increase in the amount of uranium-233 that could be produced in the

Berkeley cyclotron. [9, pg.181] On August 12th Katzin's team had completed their extraction and had six milligrams of uranium. [9, pg.189]

There was a degree of uncertainty associated with just how much uranium-233 they had extracted from the pile, since their technique for assessing the mass was based on an assumption that the half-life of uranium-233 was 120,000 years, when in reality it is 159,000 years. W. C. Johnson pointed out to them that there was an initial natural uranium impurity in the thorium that was loaded into the reactor of 0.05 parts per million. Despite this incredibly tiny value, the amount of uranium-233 that had formed in the thorium during its residence in the reactor was also incredibly tiny. The result was that when all of the uranium, both the natural isotopes and the 233 isotope, was chemically extracted from the thorium, the natural uranium composed about 10% of the uranium in the sample. This was throwing off their calculations of mass, half-life, and nearly every other nuclear property that they meant to measure. As they factored this correction into their calculations, they came up with a new half-life of 153,000 years for uranium-233, much closer to the actual value of 159,000 years. [9, pg.209]

By September 9th, a mass spectrographic analysis of the uranium extracted from the thorium nailed down its relative proportions: uranium-233 was $85 \pm 1.5\%$ of the total uranium in the sample. [9, pg.232]

Seaborg took another trip to Montreal in late September 1944 to visit with the Canadian "Evergreen" group that was interested in uranium-233. They discussed the proper chemical forms to produce uranium-233 in an X-10 type reactor and the chemical processes that could be used to extract uranium, including a technique that used fluorine as an extraction method.

On November 1st, 1944, sufficient work had been done on the measurements of the cross-sections of uranium-233 to allow Seaborg to ponder some of their implications. To the best of our knowledge, he was the first to realize the profound potential of thorium as an energy source:

> I reviewed recent results on the physical constants of U-233 it appears that the fission cross section is about the same as for U-235 (550 barns); the total neutron absorption is only around 620 barns, giving a favorably small value for the parameter alpha (0.12). The number of neutrons per fission is 1.07 times the U-235 value. The number of neutrons given off per neutron absorbed is 1.13 times the U-235 value. **These values are within the range to enable U-233 to be made from thorium by a chain reaction on the U-233 (*i.e.*, to make breeding possible)—extremely important because it may make it possible to be independent of uranium once a supply of U-233 for starting purposes is on hand.** [9, pg.320]

Starting with Seaborg's first musings about thorium in 1940, and culminating with the realization of this day, Seaborg had come to a conclusion of lasting importance, whose ramifications echo even to the present day and likely far beyond. He had discovered that thorium could be made into a fuel; he had discovered that fuel could be made to fission, and now he had discovered something of surpassing importance about that fuel, something that distinguished it from all others.

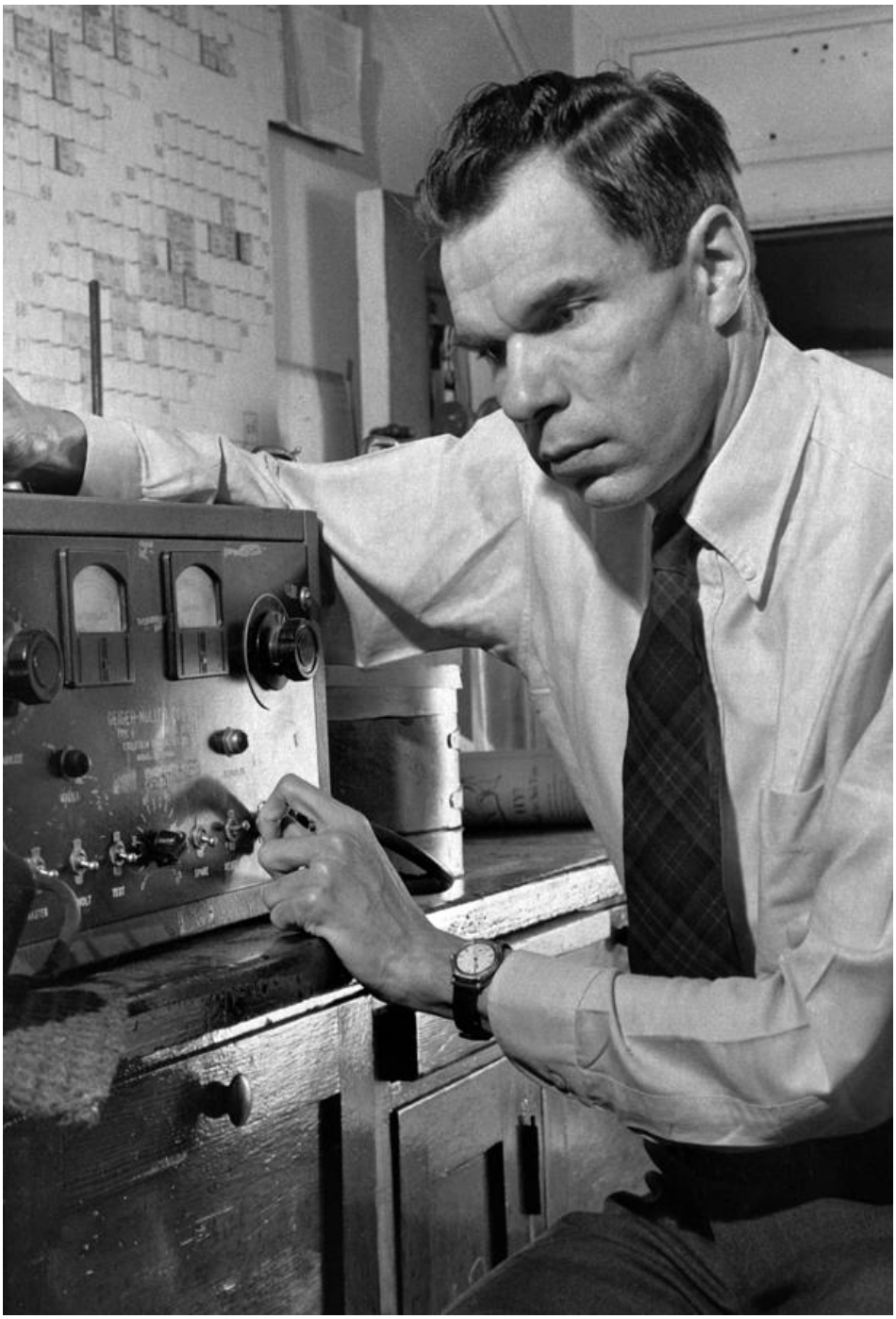
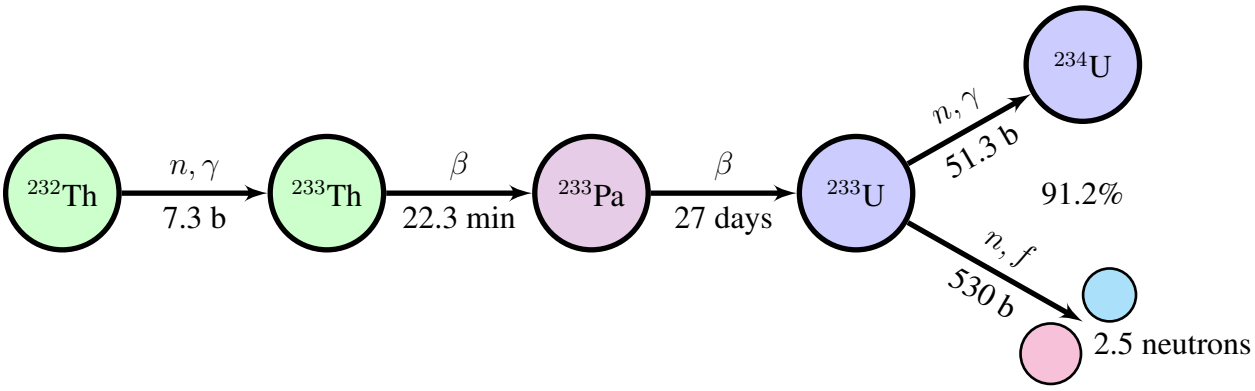


Figure 18: Glenn Seaborg's diligent research of the properties of uranium-233 led to the discovery that it had the capability of sustaining the nearly complete consumption of thorium as an energy resource. This realization transformed thorium into perhaps the world's greatest practical energy resource.



Uranium-233 had a high propensity to fission when it was struck by thermal neutrons, far more than plutonium-239. And after it fissioned, it gave off sufficient neutrons to continue the chain reaction (which would require that one neutron find another fissile nucleus) and to create new fuel (which would require that another neutron be absorbed in thorium). There was even margin for the loss of neutrons from any sort of practical reactor.

This was the first realization of the potential of a breeder reactor—a reactor that could make as much, if not more, fuel than it consumed. A breeder reactor based on thorium, which would later go on to dominate the thoughts of young Alvin Weinberg, was born that day in the mind of Glenn Seaborg, as he studied the relative cross section values of uranium-233 and realized that inexpensive, abundant thorium could be an essentially inexhaustible energy source. Seaborg is said to have remarked many years later that this was a "fifty-quadrillion-dollar discovery". [16]

4.8 Xenon-135 and the Dramatic Hanford Startup

In the fall of 1944, the titanic construction effort that was taking place at the Hanford site in the western deserts of Washington state was bearing its first fruit—the completion of the first reactor, called the "B-reactor". Several other reactors were also under construction and would soon follow B-reactor into operation. Uranium slugs were first loaded into B-reactor on September 13, and on September 27, at a few minutes after midnight, the reactor achieved criticality for the first time. Within two hours, the B-reactor at Hanford was producing more thermal power than any reactor before it ever had, and it still had a long way to go to reach its rated capacity.

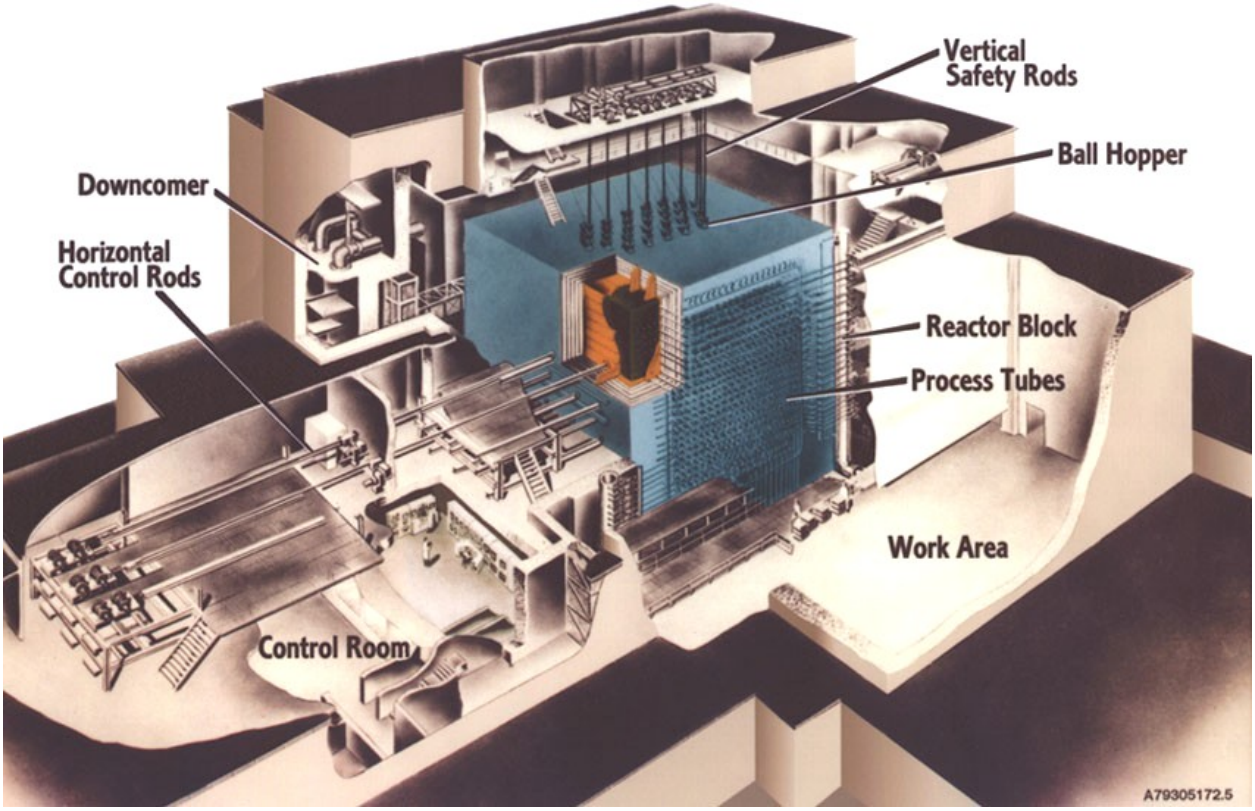


Figure 19: Cutaway view of the interior of the first plutonium-producing reactor (B reactor) at the Hanford site in Washington state. The reactor essentially consists of a huge block of graphite, through which slugs of natural uranium were pushed through. The uranium contained both the fissile material to sustain the nuclear reaction (U-235) and the fertile material (U-238) that could transmute to plutonium under neutron bombardment. The reactor was cooled by water pumped through the fuel channels. Early operation of the reactor was plagued by the buildup of a fission product that strongly absorbed neutrons, later found to be a noble gas, xenon-135. Additional uranium fuel was loaded into fuel channels that overrided the poisoning effects of xenon-135 and the reactor went on to produce the plutonium used in the first nuclear test (Trinity) and in the nuclear attack on Nagasaki, Japan.

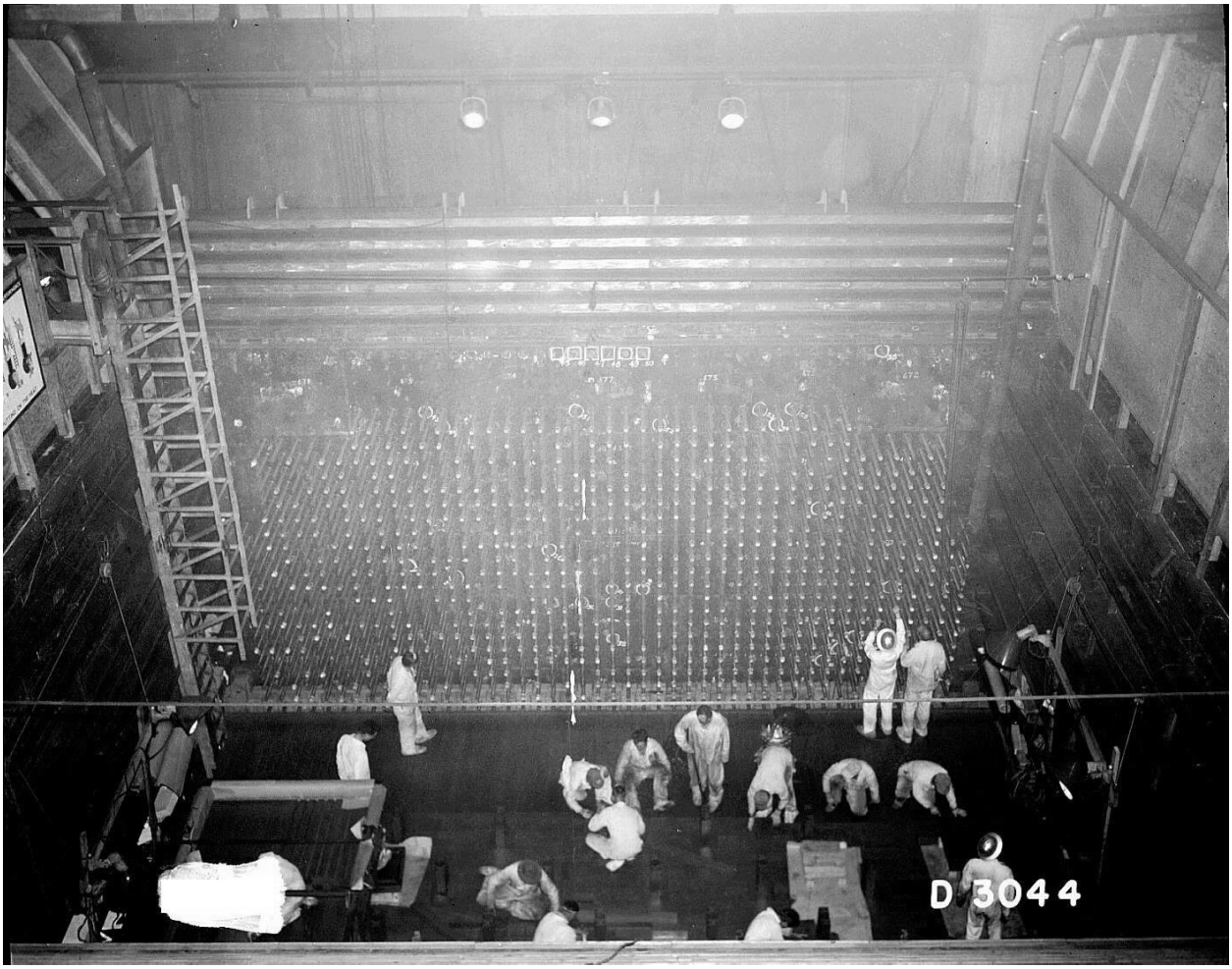


Figure 20: The loading face of the B reactor at the Hanford site. Much like the X-10 reactor in Oak Ridge, uranium slugs were loaded from one end of the reactor and extracted from the other, where they contained small amounts of plutonium that was chemically extracted.

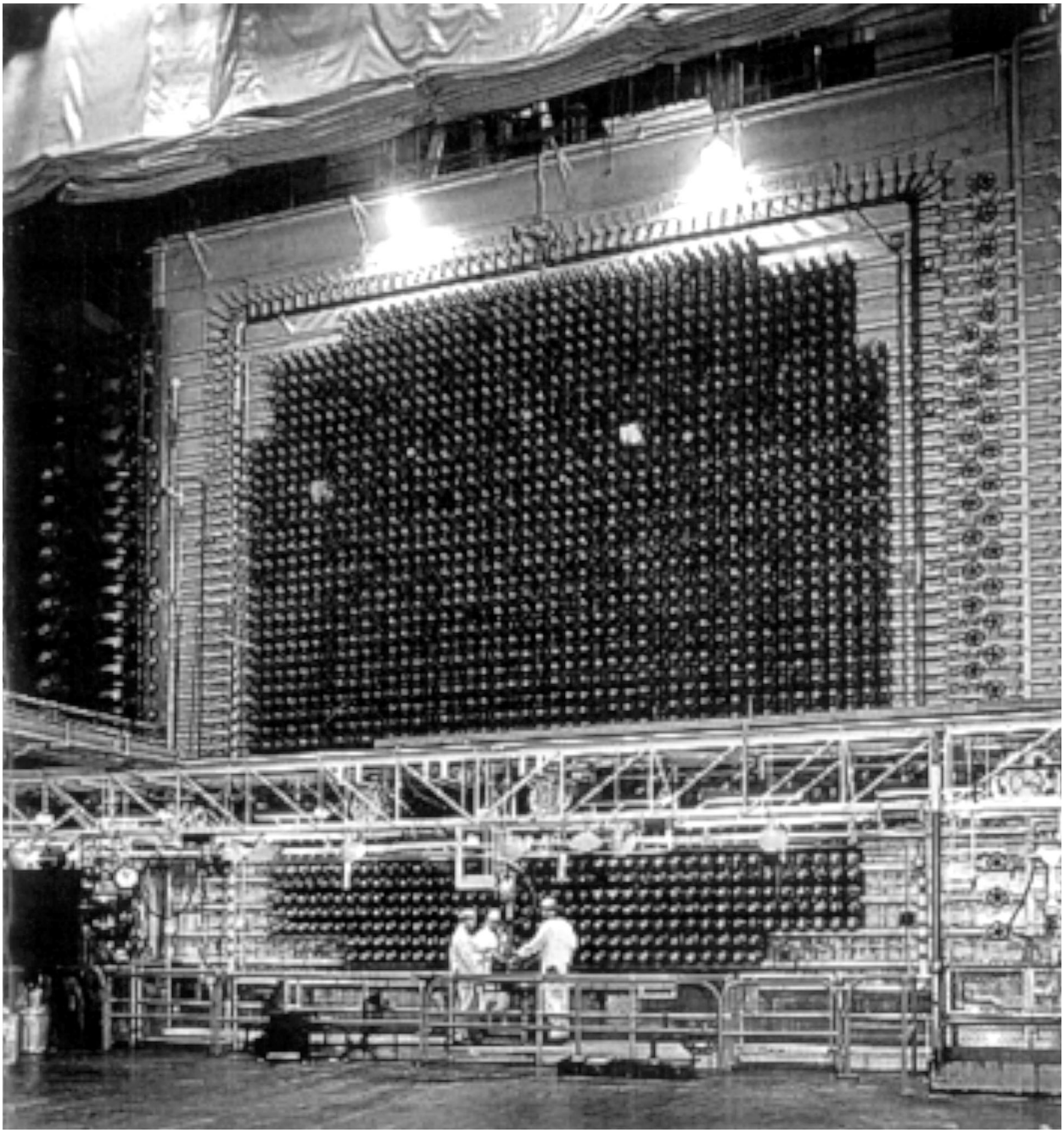


Figure 21: Another view of the loading face of the B reactor at the Hanford site.

Once again, Enrico Fermi was supervising the startup of B-reactor, just as he had done at the first pile in Chicago nearly two years before. But B was an absolutely huge reactor, intended to produce 500 megawatts of thermal power, over a billion times more powerful than the pile in the squash court in Chicago, and hundreds of times more powerful than the X-10 reactor at Oak Ridge.

Within three hours of startup, something unexpected happened to the B-reactor. Its power level began to fall slightly. Reactor operators had made no change in the controls, yet the power level of the reactor continued to fall. For hours the decline in power continued, dropping faster and faster as time went on, until by six-thirty the reactor had shut off completely. This strange behavior had never been observed in a reactor before, and Fermi's group had prided itself on how well they had predicted the behavior of reactors in the past. On the B-reactor they had predicted the amount of uranium needed to achieve criticality within 1%. What could possibly be going on with the reactor?

A variety of hypotheses were put forward. Perhaps coolant tubes were leaking, or something was coming off the fuel to "poison" the reaction. But early Thursday morning, a little more than a day after the reactor had first been brought to power, the counters showed that the reactivity of the core was increasing again. The reactor slowly began to rise in power, just as inexplicably as it had lost power before, faster and faster. By seven am on Thursday the reactor was at half-power, and twelve hours later it had attained the same power rating it had achieved on the first startup. But then it began to decline in power again, falling and falling until it shut down once more.

This was the clue that the physicists needed to begin to deduce the problem. Something was happening in the fission reaction that was causing the reactor to shut down. Then, after it shut down, that same something was going away. Once it had gone away, the reactor would start again, but the operation of the reactor must be creating that same something yet again, which would lead to its shutdown. The most logical conclusion was that there was a fission product—one of the scores of different isotopes created in fission and decay—that was highly absorptive of neutrons. Yet this same fission product must be radioactive as well, because its effect diminished over time.

By Friday morning they had accumulated enough data on the rise and fall of power in the reactor to make some educated guesses about this isotope and its properties. From the rise of reactor power they deduced that it had a half-life of about nine hours, and from the fall in reactivity they concluded that it must have a gargantual appetite for neutrons—something vastly more absorptive than any isotope previously encountered. The data was pointing to an isotope of the noble gas xenon, with a mass number of 135. The neutron absorption cross section of xenon-135 relative to uranium-235 is depicted in Figure 22.

To be sure, the group at Hanford enlisted the help of Walter Zinn's reactor group at Argonne. Using their heavy-water pile CP-3, they ran the reactor at high power to see if they could duplicate the effect that was seen at the B-reactor at Hanford. It didn't take long, once they knew what to look for, and within twelve hours Zinn was reporting back that he was highly confident that xenon-135 was the culprit behind the strange behavior at Hanford.

To understand how xenon forms in a reactor, it is necessary to understand a little more about the actual process of nuclear fission. Fission is the process whereby a neutron strikes an atomic nucleus and causes it to split into two pieces. It is tempting but erroneous to think that these two pieces are the same size, and the reason why they are not is enlightening. Recall that the stability

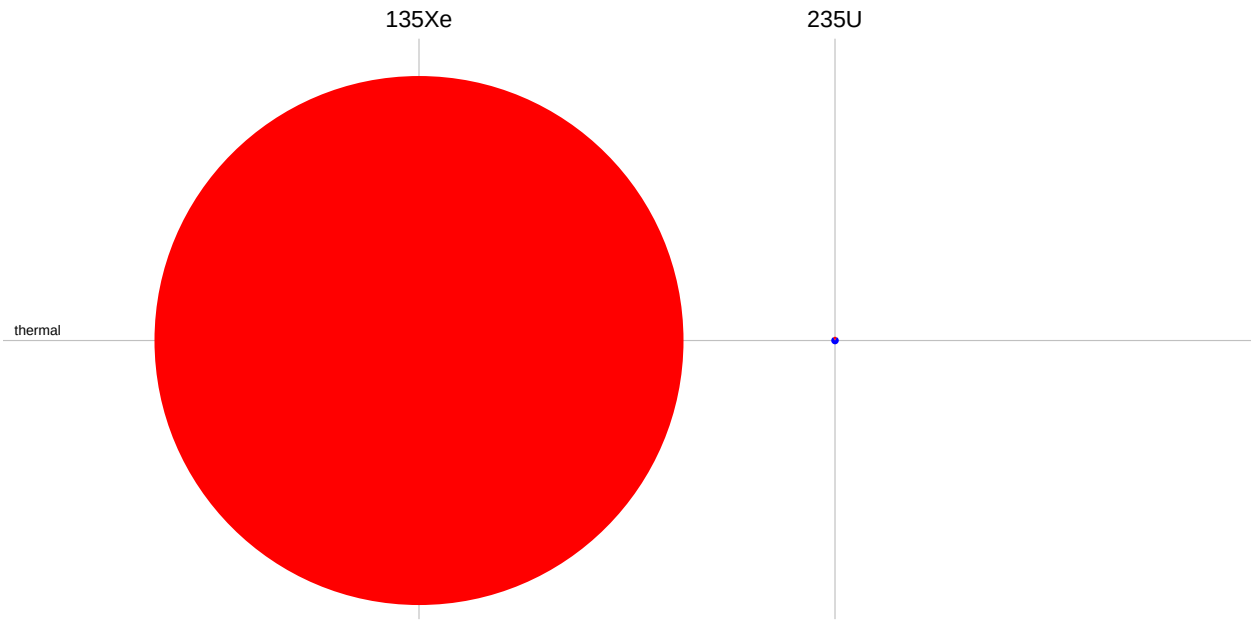


Figure 22: The relative sizes of the thermal cross-sections of xenon-135 and uranium-235. Xe-135, with a total thermal neutron absorption cross-section of 3.085 million barns, dwarfs U-235, with a total thermal absorption neutron cross-section of 630 barns, by a ratio of roughly 4900 to one.

of the nucleus is a constant struggle between the nuclear force, which tends to bind protons and neutrons together, and the electromagnetic force, which is repelling protons from one another and trying to rip the nucleus apart. This is why some atomic nuclei are radioactive—they do not have a stable configuration of protons and neutrons to keep these forces from changing them. In an atomic nucleus that is about to fission, the nuclear force and the electromagnetic force are again fighting one another.

As the nucleus begins to pull apart in fission, the size of the smaller piece is determined by the interplay between the two forces. A small piece, below about 10% of the mass of the original, simply doesn't form because the nuclear force is too strong to let a piece that small be ejected by the electromagnetic force. The largest piece that could form in fission would have 50% of the mass of the original nucleus, but in real fission reactions that rarely happens, because the electromagnetic force rips the two pieces apart before a fragment of that size can form. So what happens in reality is that two pieces form, and that the smaller piece has about 35-44% of the mass of the original. If the smaller piece is this size, then it stands to reason that the larger piece is about 56-65% the size of the original nucleus, and that is indeed the case.

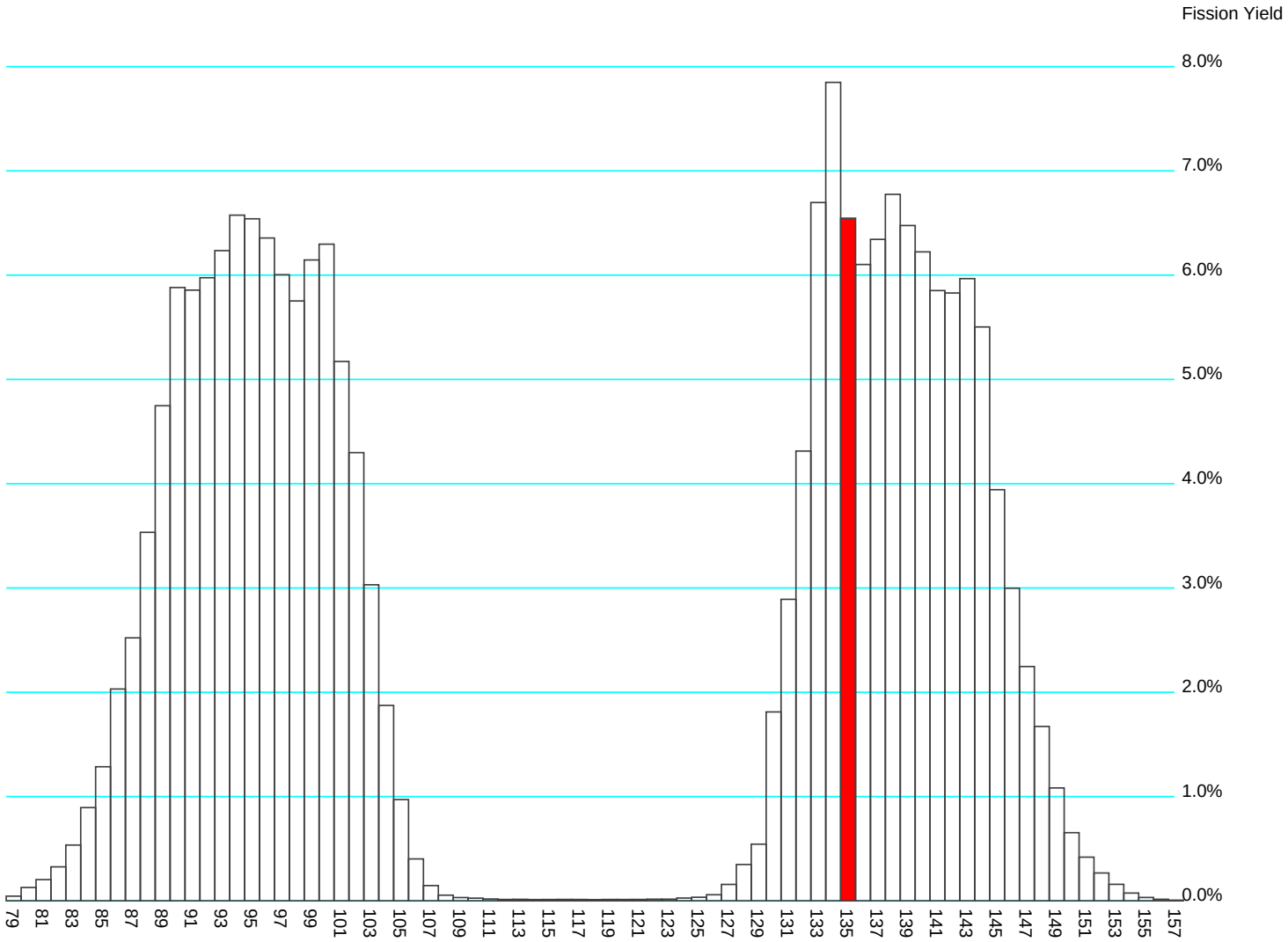


Figure 23: The mass distribution of products from the fission of uranium-235, with mass number 135 highlighted for clarity. Xenon-135 was part of the radioactive decay of this chain, and it had a tremendous propensity to absorb neutrons.

Real fission reactions—and there are many possible ways for a nucleus to fission—result in two fission products whose mass follows a double-humped distribution, when production is plotted by atomic mass. For the smaller fission product, the peak of the hump is centered around zirconium on the periodic table, and for the larger fission product, the peak is roughly centered around xenon.

The fission product distribution shows the creation of xenon-135 from fission and subsequent decay is not a rare outcome—rather it is one of the most common. And a fission fragment with 135 atomic masses is very close to the second peak of the fission product distribution, as shown in Figure 23. So fission makes a relatively large amount of fission products with mass number 135, and in the course of their decay they pass through a stage where they become highly absorptive xenon-135. This causes problems in the operation of nuclear reactors, as the team at Hanford was discovering in late September 1944.

This discovery was conveyed to General Groves on October 3, and he was not pleased. He had ordered the CP-3 reactor at Hanford run around the clock to detect problems like this. Now they were showing up in the big plutonium production reactor at Hanford—the reactor that really had to work for the Manhattan Project to be successful. All of the other reactors had been but preludes to this moment, yet fundamental problems were being discovered at Hanford instead of the test reactors.

By mid-October, researchers were generally satisfied that xenon-135 was indeed the cause of the behavior of the Hanford reactor, and the issues associated with startup were eventually resolved by loading more fuel into extra channels that had been built into the design. This extra uranium increased the multiplication constant sufficiently to overcome the effects of xenon poisoning, and allowed the B-reactor to achieve full power by mid-December. As 1945 dawned, the Hanford reactors were now producing plutonium through fission that would be chemically removed using the steps that Seaborg and his Met Lab team had carefully worked out. After being separated, it would be sent to Los Alamos to be fabricated into the first nuclear explosive device.

There was another important lesson from the xenon scare at Hanford. Xenon was a noble gas. It did not have any propensity to chemically bond to other materials. In the solid fuel slugs that fed the Hanford reactors, it was trapped and could not escape. But in a reactor that used liquid fuel, there was every reason to believe that xenon would bubble out of the fuel mixture and be removed. Since all fission reactions created sizeable amounts of xenon-135, a fluid-fueled reactor could be a way to get around the thorny problem of xenon in future designs.

4.9 The Plutonium-to-Uranium-233 Converter Reactor

In the fall of 1944, the mood of the scientists at the Metallurgical Laboratory in Chicago was sour. There was great uncertainty about the value of the work they had done, and some even went so far as to consider their efforts to develop plutonium for nuclear weapons a failure. Plutonium-239 could not be produced at high purity despite their best chemical efforts, and it was still unknown whether what could be made was actually good enough for weapons use.

On October 10, Charles Cooper presented a discussion on "New Goals for the Metallurgical Project" where he pointed out the possibility that the Met Lab's primary responsibility might already be complete, and that there would be a need to find a new role and responsibility for the

talented group of researchers to pursue. He summarized the current state of the project in less-than-enthusiastic terms:

> By some two years of great effort under emergency conditions we have arrived at a situation which may be summarized thus. A process has been developed which very certainly will produce quantities of 49 [plutonium-239] adequate for experimental purposes and probably sufficient to meet situations immediately foreseen. The present process is very complicated and unduly expensive, both as to manpower requirements and the materials which are involved. **The present approach is not adaptable to generation of power nor is it designed to manufacture or recover effectively possible byproducts of operation.** The product will be contaminated with undesired isotopes which could possibly be eliminated if other methods, as yet only sketchily developed, were to be employed. We cannot be entirely certain until final trials have been made that the product will be as useful as anticipated, and it may well develop that other elements or isotopes will turn out in the end to be much more useful than the one we have prepared to manufacture.

Cooper could see that the efforts of Wigner and others to build production reactors had led them in a direction that was not compatible with the eventual goal of power production from nuclear reactors. The central reason for this was because production reactors viewed nuclear heating as an undesirable byproduct of reactor operation. Their only goal was to produce fissile plutonium from natural uranium, not to produce thermal power suitable for operating a power conversion system like a steam turbine. To that end, the reactors at Hanford and at Oak Ridge simply threw thermal energy away, putting it to no use at all. Nor was there any prospect of redesigning those reactors to produce a useful form of thermal power. They were too large to be pressurized, which at any rate would involve putting thick metal channels in them that would absorb too many neutrons to allow criticality. They were doomed to being low-pressure machines that could not raise steam or make electrical power. They could only make plutonium.

A reactor that could make useful electrical power would be an attractive goal, and Cooper pressed for that goal, which he thought "poses a problem much more difficult technically than the one which we have had to solve in connection with our present manufacturing objectives." Seaborg also noted that power reactors would have additional problems in the areas of high temperatures, corrosion resistance, the need for new materials and alloys for fuel fabrication, structural purposes, and shielding. [9, pg.277-278]

A power-generating reactor would be a noble goal, but Eugene Wigner was still as determined as ever to deliver useful fissile material to defeat the Nazis. Despite his focus on that goal, his group was about to propose ideas that would one day lead to power-generating nuclear reactors, as well as advanced nuclear reactors using fluid fuels.

It began with the proposal to design a new reactor that would consume the plutonium that would be produced in fission, and use the neutrons from that fission to convert thorium to uranium-233 that would be used in weapons. The reactor had the potential to be cooled and moderated by normal (light) water, since the plutonium would have a high fissile content (unlike natural uranium).

Weinberg described the proposed "converter" reactor:

> Wigner's converter consisted of two concentric regions. In the central region were a large number of parallel aluminum plates each containing plutonium. Surrounding the central region was a ring of aluminum plates each containing thorium. The entire array of plates was moderated and cooled by swiftly flowing water. The chain reaction occurred in the central, plutonium-containing region. About half of the neutrons produced by fissioning plutonium leaked into the thorium and converted the thorium into ^{233}U. [11, pg.38]

This reactor would take plutonium that would be produced by the Hanford reactors and form it into solid-fuel elements. These would be immersed in ordinary (light) water, which was a feasible moderator in this case because the plutonium contained a high fraction of fissile material, unlike natural uranium. By surrounding the plutonium fuel elements with thorium, uranium-233 would be generated which could be chemically extracted and hopefully incorporated into the wartime effort.

This general direction was sustained in a November 22 meeting that Seaborg attended:

> Compton asked that those present consider whether the work from here on should continue to be organized around the immediate future or whether some other objective should be followed. In the discussion of possible objectives that followed, **Jeffries pointed out that the objective of Chicago has been achieved only in part since plutonium does not have all the desirable properties that had been anticipated. The same apparently holds for U-235. Perhaps, then, the only way to provide adequate insurance that the objective really be achieved would be to be ready to convert Pu-239 over to U-233.** This could provide a clear-cut objective. [9, pg.352]

The theory of a plutonium-uranium-233 converter reactor was much more straightforward than a production reactor that had to operate on natural uranium since the fissile content of plutonium was hundreds of times greater than natural uranium, but the bulk of the problem fell on Seaborg's chemical research group. In this respect the "fuel" of the proposed converter reactor was vastly more expensive than the natural uranium used to fuel Hanford. They would not be able to afford to waste it as they were at Hanford. They would have to figure out how to dissolve the solid plutonium fuel elements, separate plutonium from fission products, and reconstitute new plutonium fuel elements, all the while minimizing the loss of valuable plutonium at each step. Laying out the core design of the reactor with plutonium and thorium (Wigner's task) was doable; figuring out the nearly-perfect chemical processing scheme (Seaborg's task) was nearly impossible.

Despite the challenges, a reactor development task of significance was seen as something that would be necessary to hold the Met Lab team together for the months until the war was won. The Met Lab's report for November 1944 stated in its summary:

> It is assumed that by June 30, 1945, the purpose for which this Laboratory was established—namely, that of aiding the Hanford Engineering Works to establish production—will have been fulfilled. The work from now until June 30, will be divided into three parts...during all this time a competent staff of personnel must be maintained which

> is qualified and willing at a moment's notice to devote its entire attention to any unforeseen problems which may arise at the Hanford Engineer Works or Y. **To maintain such a staff intact with a sufficient degree of morale, it is necessary that they work on some forward-thinking problem. Most promising of the problems at the present time seems to be in the field of (1) 'converter' piles and 'breeder' piles...** [9, pg.368-369]

On December 19, the leadership group over Seaborg's work met and considered the probability that the converter pile could be made to work effectively. They knew that only 7 times out of 10 would plutonium-239 fission with thermal neutrons, so there was a built-in 30% loss of fuel from the outset. From there, things got progressively worse depending on the frequency at which the fuel elements would need to be processed and the anticipated loss of plutonium material at each step of processing. Processing recovery factors of 100% (perfect recovery) down to 90% were considered, and the overall results were very poor. Essentially, a chemical process would be needed that achieved a 99.9% recovery factor, and Seaborg said "**an efficiency of 99.9% for a decontamination by a factor of some** 10^7 **followed by refabrication of an aluminum sandwich is totally impossible by any known process and probably also by any conceivable process.**" [9, pg.406]

The problem that Seaborg was describing was not unique to the converter reactor that they were attempting to design. In fact, it is a problem that persists in every nuclear reactor design to this day, particularly those that would attempt to recycle plutonium fuel in a thermal-spectrum reactor. Every single time the solid fuel is dissolved, there are inevitable losses of fissile material, and every single time the liquefied fuel is refabricated, there are more losses. It would be analogous to going to a currency exchange that takes some fraction of your money each time you convert from one currency to another. The percentage might seem low, but given frequent conversions, a large fraction of the value of your money can quickly be lost.

Seaborg put forward some hope for the bleak situation in the form of a reactor that used liquid fuel. This was not the first time such a reactor had been considered—in fact, it had been considered off and on for many months by the reactor design team at the Met Lab—but in this scenario it was being put forward as a potential solution to the fundamental issue of material losses each time solid fuel was converted to liquid for processing and separation, and then converted back again.

> I also mention, as an alternative possibility, a homogeneous reactor with ordinary water that would require a minimum of decontamination (only to remove fission product poisons) and would avoid the hold-up due to fuel fabrication operations; however, little is known about this approach, and hence this could require the most research work.

At a meeting the next day, Wigner was disappointed but unsurprised to find out that a solid-fueled plutonium-uranium-233 converter reactor would have very low yields due to losses in chemical transitions. As the meeting progressed, the group generally began to believe that a converter reactor based on fluid fuel, with a reasonable approach to chemical processing, could produce fissile uranium-233 with a 50% yield, and that they should orient their efforts around making a

case for this reactor to project leadership in an upcoming meeting. If they were able to obtain a go-ahead, then it would give the Met Lab a new goal and a new focus, and would put them back in the business of directly contributing to the war effort.

A 50% yield was a disappointing objective however. The notion that plutonium would be produced at extraordinary cost from the Hanford reactors, only to be immediately introduced into yet another reactor where it would be consumed to generate uranium-233, was not a satisfying prospect. It added more layers, more costs, and more time to a project that was already struggling to meet its overall goal.

As 1944 ended, Seaborg reported that the main effort of the Met Lab had been to prepare "a report to the Project Office on the feasibility of a conversion of 49 [plutonium-239] into 23 [uranium-233] as a war-time insurance measure;" and the "preparation of a report to the Project Office making recommendations for a research and development program for the Laboratory from June 30, 1945, to the end of the war." [9, pg.430-431]

On January 3, 1945, Seaborg noted another problem with the idea of a converter reactor:

> A further problem, not solvable by chemical processes, I went on, is that for every 140 grams of Pu-239 present there are 40 g of Pu-240 formed in addition to 100 g of fission products. If Pu-240 will not fission with slow neutrons, the result will be that in successive decontaminations the dead isotope will be carried along, taking up valuable space, and contributing nothing.

This issue continues to be a problem for thermal-spectrum reactors that intend to use recycled plutonium as a fuel.

Stacking up, one by one, each of these issues contributed to the eventual cessation of effort on the development of a plutonium-uranium-233 converter reactor. On January 15, Seaborg records:

> Compton wrote to Stearns to thank him for sending the report on the feasibility of the 23 converter pile. The matter has been placed in the hands of the Army with the statement that further active steps along this line will await a request from them. **Compton also mentioned that in discussing the problem with Colonel Nichols, the impression has been gained that it is unlikely that we shall receive a request to proceed further with the 23 converter pile.** Nichols has expressed himself, however, as actively interested in a research and development program that would lead to the more efficient use of uranium and thorium in producing a useful product. [9, pg.453]

Although Wigner's notion of a solid-fueled plutonium-uranium-233 converter reactor came to an end after only a few months of examination, Alvin Weinberg noted that it had far-reaching implications for the future of nuclear energy:

> Wigner's converter was never taken much beyond rough conceptual design, but in a way it has had enormous influence on the development of nuclear energy. For this was the first detailed description of a heterogeneous, plate-type, highly enriched reactor, cooled and moderated by water...**most research reactors, almost all naval reactors, and most commercial nuclear reactors have been water-moderated and cooled, more or less along the lines of Wigner's converter.** [11, pg.38]

Although Wigner's converter reactor was never built, it still represented a great leap forward in reactor development. It is worth considering that the pressurized-water, uranium-fueled reactors that dominate nuclear energy around the world today did not evolve from the gas-cooled X-10 reactor at Oak Ridge, nor did they evolve from the huge graphite-moderated, water-cooled reactors at Hanford. They evolved from Wigner's efforts to convert plutonium-239 into uranium-233 in the fall and winter of 1944.

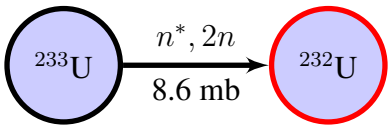
4.10 Uranium-232, the Fatal Flaw for Weapons

On December 28, 1944, Seaborg completed a ten-page report entitled "Conversion of Pu-239 to U-233" and sent it to project leadership. He gave a more detailed description of the problems that any converter reactor would face as it attempted to consume plutonium-239 to generate uranium-233. But for the first time it also detailed a much more insidious problem that would eventually close the book on the practical consideration of uranium-233 as a material for nuclear weapons.

Seaborg was acutely aware of the problems that had been caused by the Manhattan Project's failure to appreciate the real behavior of plutonium-239 in reactors. It didn't always fission, and when it did not fission it formed plutonium-240 which had a significant probability of spontaneous fission. Spontaneous fission produced neutrons that could cause a nuclear weapon to pre-detonate, and it made plutonium unsuitable for use in the simple "gun-type" weapon that was the baseline design of the Manhattan Project.

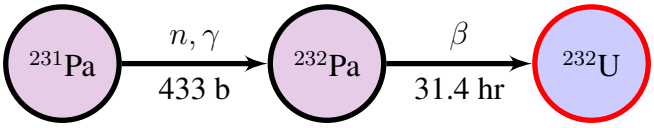
If uranium-233 was to be considered as a material for weapons, Seaborg did not want a repeat of the issues with plutonium, and he was on the hunt for other side-reactions that might also cause trouble. U-233 was much more likely to fission than plutonium-239, with only one reaction in ten leading to the formation of uranium-234. Uranium-234 was not nearly the problem that plutonium-240 was, but uranium-233 had a different problem. In the production techniques they envisioned, there were several different ways that a new isotope, uranium-232, could form. Uranium-232 had a relatively short half life (less than 70 years) and its rapid decay would produce a background of alpha particles.

Uranium-232 formed in several different ways. One of them was when uranium-233 was struck by a high-energy neutron. Sometimes rather than fissioning it knocked loose a neutron in addition to the one that struck it. This caused uranium-233 to turn directly into uranium-232. This process required high-energy neutrons,[6] but each fission neutron begins its existence with a great deal of kinetic energy (before it is moderated) and so this process is possible in either fast or thermal-spectrum reactors.

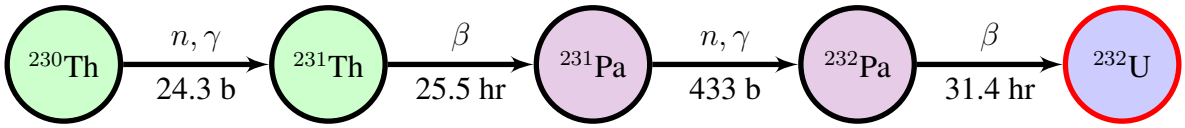


[6] In the reactions depicted in the graphics, fast neutron reactions are indicated by n^* (such as $n^*, 2n$) while thermal neutron reactions are simply indicated by n (such as n, γ). Cross-sections for fast-neutron reactions are generally very small, measured in millibarns (mb) rather than barns (b).

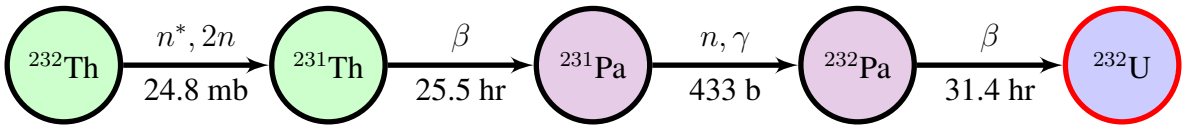
Another was the uranium-232 that formed when protactinium-231 absorbed a neutron. High purity thorium could minimize the presence of Pa-231, but it couldn't be eliminated because there were two ways in which it could be formed in thorium itself.



One of them was when the rare but natural isotope thorium-230 (also sometimes called "ionium") would absorb a neutron, forming Pa-231 which absorbed another neutron to form U-232. Th-230 was a natural decay product of abundant uranium-238 and was chemically identical to thorium-232, so any thorium deposit that was found near uranium (which was common) tended to pick some up.



Another formation process came from thorium-232 itself. Sometimes when it was struck by a high-energy neutron it also kicked out a neutron in addition to the one that struck it, forming Th-231 which rapidly decayed to Pa-231. Then the Pa-231 absorbed another neutron and formed U-232.



These three formation pathways made it extremely difficult to make uranium-233 that was completely free of uranium-232 contamination, and Seaborg was determined to raise these issues as early as possible with project leadership. Describing his memo, he said:

> **I begin by revealing the complications that will be introduced by the nuclear side-reactions in relation to the problem of final chemical purification of U-233.** I point out that if care is not taken to control small amounts of heavy isotope impurities, **the final material may have the same limitations as the material (plutonium) whose deficiencies are being remedied.** At the time the material is worked up the thorium will contain elements 90, 91, 92, and in some cases, 93 and 94. In addition to U-233, there will be produced as relatively stable long-lived isotopes, Pa-231, U-232, U-234, and, in the case where 94 is present, U-235, Pu-239, Pu-240, and Pu-241. **Of particular concern is the presence of U-232 that has four short-lived (hence long-range) alpha-particle decay products, which means that the problem of purifying the material from light element impurities assumes importance again as it originally did in the case of Pu-239.** There is also the yet unanswered question of spontaneous

> fission in U-232 or one of its daughters. All of these factors make it seem extremely important that the factor of possible fast neutron reaction (the source of U-232) in the thorium mass be minimized, including the possible necessity of processing the thorium at shorter intervals than other considerations might dictate. [9, pg.426-427]

Seaborg then described the prospects that he saw for success in a converter reactor. He estimated that it would take 100 men and at least 6 months to develop a process that could achieve 98% recovery of plutonium and thus 50% conversion efficiency of plutonium-239 to uranium-233. Such resources were not forthcoming in the Met Lab as currently constituted. After detailing the formation pathways for uranium-232, he issued a negative view on the value of proceeding with such an effort:

> U-232 has a short half-life (approximately 50 years) giving off alpha rays and producing decay elements which fit into the natural thorium decay series. This series includes several short-lived alpha-emitters. Among these alpha-emitters is ThC' which gives off alpha-rays of exceedingly high energy. This fact introduces complications, in that the tolerance limits for some of the lighter elements may have to be made more exacting for the ultimate use. **U-233 appears to be better than Pu-239 and purification from light elements would seem to be less exacting by a factor of ten or so. However, this advantage may be completely nullified and, in fact, the situation may be worse if appreciable amounts of U-232 are present in the U-233.** The possibility of high spontaneous fission rates for U-232 and its decay products must also be taken into account. These possibilities must be considered in designing piles for converting plutonium into U-233. It seems likely that the formation of U-232 can be sufficiently minimized by placing the thorium only at the outside of the pile where there are no fast neutrons and by reducing the length of exposure before the U-233 is extracted from the thorium." [9, pg.429]

Plutonium-239 had been a disappointment to the Met Lab chemical researchers because of its propensity to form plutonium-240. But given the formation of uranium-232 in converter reactors, Seaborg was of the opinion that uranium-233 might be an even worse material for nuclear weapons. He was right, and Alvin Weinberg recalled that Seaborg's foresight probably saved a great deal of otherwise wasted time and effort:

> At the time, however, there was no assurance that the implosion would work; and if it didn't work, the whole Chicago project was a failure! What to do, what to do? Arthur Compton convened a staff meeting to discuss possible ways to retrieve the situation. The meeting was dominated by Wigner. He proposed that the ^{240}Pu-contaminated plutonium from W be converted into the fissile isotope ^{233}U in a special "converter reactor." ^{233}U is produced from thorium-232 when the latter absorbs a neutron, much as ^{239}Pu is produced when ^{238}U absorbs a neutron. ^{233}U is fissile, and could possibly be used in a bomb with little chance of predetonation. (**Glenn Seaborg, however, pointed out that ^{233}U manufactured in Wigner's converter was contaminated with**

> **^{232}U, which probably fissioned spontaneously and also emitted many alpha particles. A bomb made of ^{233}U would therefore have to be carefully cleansed of any tramp light elements such as beryllium, since alpha particles interacting with beryllium would produce neutrons; and if spontaneous fission of ^{232}U was sufficiently probable, the ^{233}U gun-type weapon was unworkable.** I was greatly impressed by Glenn's insight into this difficulty, since at the time little was known about either ^{233}U or ^{232}U.) [11, pg.37-38]

By the spring of 1945, serious consideration of uranium-233 as a material for nuclear weapons had essentially come to an end. It was much more difficult to make a unit of uranium-233 than plutonium-239, and the uranium-233 was contaminated with uranium-232 that could not be separated chemically from uranium-233, much like plutonium-239 was contaminated with plutonium-240. The chemists and reactor designers of the Met Lab were far more excited about the prospects of using uranium-233 in a breeder reactor.

4.11 Fluid-Fueled Thorium Breeder Reactors

On January 17, 1945, within two days of his decision to stop working on the plutonium-uranium-233 converter, Arthur Compton and Colonel Nichols were speaking optimistically about the prospect of turning the attention of the Met Lab to thorium breeder reactors instead. Seaborg notes:

> With regard to [research and development], Compton said that he was interested to find that three different laboratories have proposed breeder piles for U-233. He stated that while we have not been authorized to carry out the program activities as proposed, the indications are that the level of support will be relatively constant and we will have a wide range for making decisions about their priority.
>
> Compton reported on a discussion with [Colonel] Nichols at Clinton on the possibilities of a U-233 converter, based on the report prepared early this month which describes the feasibility of such a converter. Compton said that he told Nichols we prefer the breeder, whereupon Nichols said he thinks it is more important that we go ahead on the [thorium-U-233] breeder than on the [plutonium-uranium-233] converter and to lay our plans along this line. [9, pg.457]

Chemistry Division director Farrington Daniels had solicited ideas about "breeder" reactors, and the consensus was emerging of a reactor that would be fueled by a solution of a uranium compound dissolved in heavy water. [9, pg.488] This would eliminate the considerable difficulty of fabricating solid fueled elements and dissolving them to remove fission products. Several uranium compounds appeared suitable for dissolution in water, mostly notably uranium nitrate and uranium sulfate. Some of Seaborg's chemist colleagues, particularly Davidson and Katzin, pointed out the challenges that could be expected in a "homogeneous" reactor with fuel dissolved in water. One of them would be the formation of hydrogen and oxygen gas as radiation split apart the water molecule. Unless the hydrogen and oxygen was "recombined" back into water, it might form an

explosive gaseous mixture somewhere in the reactor. Another challenge was the chemical stability of nitrate compounds. [9, pg.492] This seemed to favor using sulfates rather than nitrates, but much of the chemical processing techniques were based on the use of nitrates. A chemical shift from sulfate to nitrate might be necessary to process the fuel regularly, and that would be undesirable.

Seaborg noted in his summary of the activities of the Met Lab for February 1945 that [9, pg.518]

> ...most of the research and development work of the Laboratory is exploratory work in connection with breeder piles. Consideration is being given to the homogeneous thermal breeder and the heterogeneous resonance and high energy breeder type of pile.

Alvin Weinberg and Eugene Wigner were also looking at the "aqueous homogeneous" reactor concept, but considering it more from the neutronic angle rather than the chemical perspective of Seaborg's group. They recognized that for each absorption of a neutron in uranium-233, it would produce more than 2 neutrons. This was the key to its ability to operate as a breeder reactor. But how much more than two? It mattered because it gave them a sense of the "margin" they might enjoy in an actual, practical reactor. How many neutrons could they afford to lose to xenon, or to neutron absorption in other fission products? How much would be lost to leakage? The answer to that question would have a great deal to do with the minimum size such a reactor could assume.

Weinberg and Wigner knew that a small fraction of the neutrons from fission were delayed. In a reactor with circulating fuel, some of these would be emitted outside of the core. With uranium-233 as the fuel, there was also a different grouping and number of delayed neutrons than in uranium-235. And although uranium-233 had better performance in a thermal reactor than uranium-235 or plutonium-239, about 10% of the time it would simply absorb a neutron too, forming uranium-234. Was uranium-234 fissile, or would it simply absorb neutrons? How much fissile material would be lost in chemical processing?

Each of these questions needed answers if a realistic aqueous homogeneous "breeder" reactor was to be built, and in many cases Weinberg and Wigner could do little more than pose the question and frame the experiments that would need to be done to find the answer. Each one of these mechanisms of neutron loss would eat away at the "margin" that uranium-233 had as a breeder fuel, and for an organization obsessed with producing and increasing the supply of separated fissile material, these were very important questions.

Weinberg and Wigner made the first detailed description of their aqueous homogeneous thorium breeder reactor ideas in an internal report titled "Preliminary Calculations on a Breeder with Circulating Fuel," issued in Chicago on May 17, 1945. [11, pg.116] In the paper, they painstakingly calculated, to the best of their knowledge and computational abilities at the time, the neutronic losses that such a reactor design would undergo, and concluded that they were confident that the losses could be held within the "margin" of neutrons that their breeder design would allow.

While it appeared that an aqueous thorium breeder might be possible and feasible, it was fairly clear that such a reactor would only breed enough new fissile U-233 to make up for the amount consumed. It would not be a "net" breeder, producting significantly more fissile but rather an "isobreeder", producing just enough for its needs. In such a design the reactor would be able to

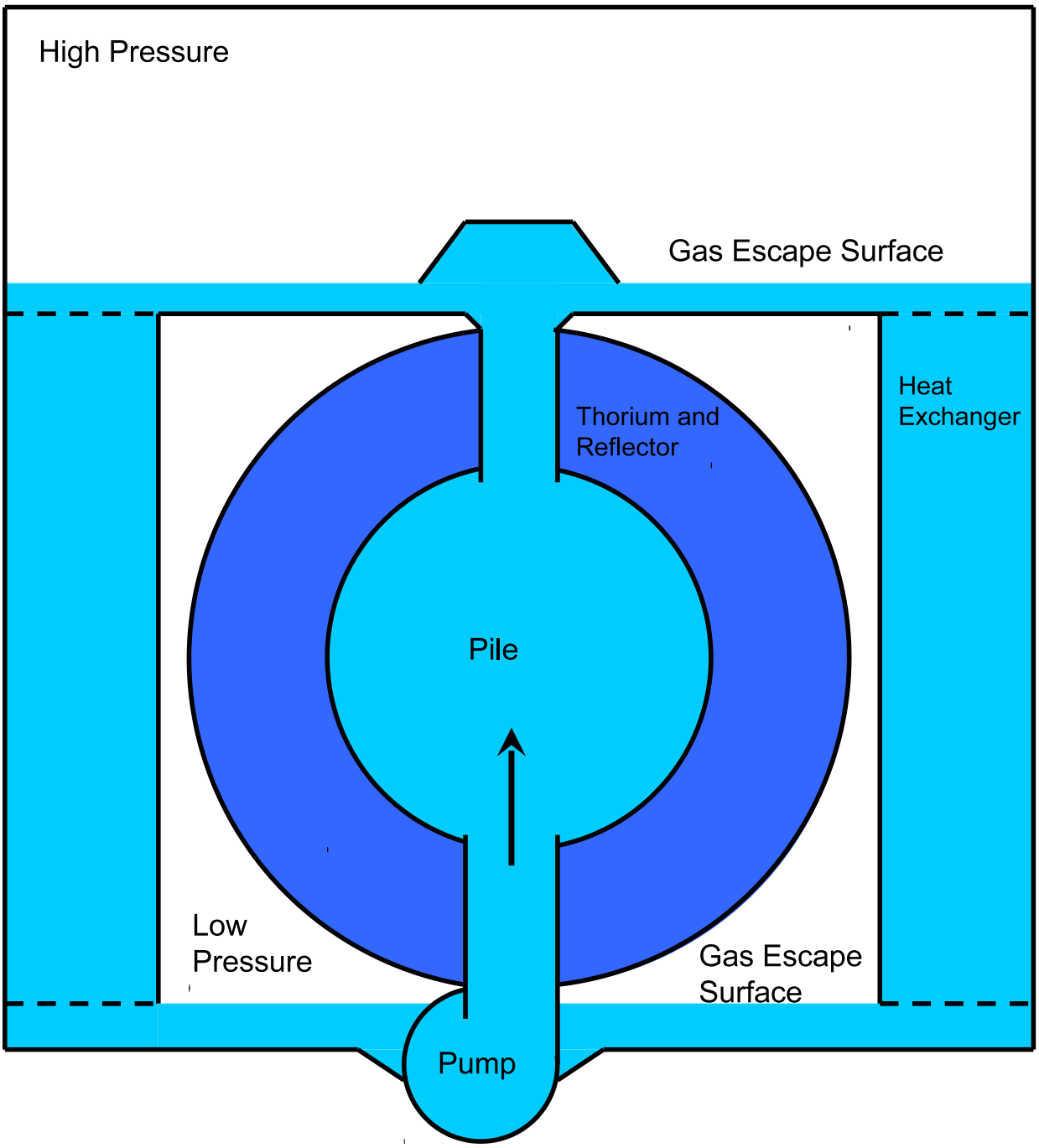


Figure 24: A recent adaptation of the thorium breeder reactor concept described in the Wigner-Weinberg report of May 17, 1945. The original caption states: "The degassing method is only illustrative."

consume thorium at high efficiency, but the fissile material that started the reactor would have to come from somewhere else. The search was still on for a breeder reactor that would have significant breeding gain, in other words, would produce substantially more fissile material than it consumed.

There was also the issue of relative value of uranium-233 versus plutonium-239. Interest in U-233 for weapons had dropped off considerably from the excitement of just a few months before. But would the impure plutonium that Hanford was producing be suitable for the first nuclear weapons? Few at the Met Lab in Chicago had insight into what was going on at Los Alamos that would conclusively answer that question.

Even if a thorium breeder reactor would not have significant breeding gain, its feasibility transformed the potential value of natural thorium from worthless to potentially the planet's greatest natural energy source. General Groves realized this could have profound geopolitical consequences. To that end, he directed that the entire supply of uranium-233 that the Met Lab had generated and separated be sent to Argonne for experiments to conclusively determine how many neutrons were produced in fission from each neutron absorbed in U-233. [9, pg.556] In late March, the Met Lab sent 60 milligrams to Argonne for further testing. Within a few days General Groves demanded to know why they had not sent the entire 100 milligrams in their possession for testing, and they scrambled to placate him. [9, pg.556] Groves was insistent that that "eta", the Greek letter that the physicists had chosen to denote the number of neutrons produced in fission per neutron absorbed, be known to high accuracy for uranium-233. If eta was greater than two, a breeder was theoretically possible. If it was modestly greater than two, such as 2.3 to 2.5, then a practical breeder was possible from an engineering perspective. It was very important to know the value of eta for uranium-233.

Seaborg and his team realized that they could get more U-233 to Argonne for measurement if they processed more of the thorium carbonate cans that had been exposed in the X-10 reactor at Oak Ridge. To that end, they entered into a "mass-production" effort culminating in a total of 200 milligrams of U-233 that they made available for analysis by April 12. [9, pg.580] Their success came just as they received disheartening news: President Roosevelt had died in Warm Springs, Georgia.

Using the 200 mg of U-233 from the Met Lab, the value for eta for U-233 was reported to General Groves on April 17 as being 2.35. A thermal breeder reactor using thorium, started by U-233, was possible. [9, pg.586] By June 27, all data relating to the eta of U-233 was deemed "top secret". [10, pg.79] Only the United States would possess the knowledge that uranium-233 could, for all practical purposes, "catalyze" energy release from a supply of thorium indefinitely.

For Alvin Weinberg, his time at the Metallurgical Laboratory in Chicago had come to an end. On a hot May weekend in 1945, Weinberg, his wife Marge and 2-year-old son David, boarded a train for Knoxville and from there on to the secret city of Oak Ridge, Tennessee, where Weinberg would spend the rest of his life, until his death in 2006. [11, pg.45-46]

Weinberg viewed his move from Chicago to Oak Ridge not as the end of his work on fluid-fueled thorium reactors, but rather as the beginning. He described his migration to Oak Ridge in these terms: [11, pg.116-117]

> The aqueous homogeneous circulating-fuel power breeder based on the ^{232}Th-^{233}U cycle became a kind of obsession for me. I came to Oak Ridge convinced that the

> laboratory, with its many chemists and chemical engineers, was the ideal place to develop Wigner's aqueous homogeneous power breeder.

Indeed, in the years to come after the war, Weinberg would supervise the successful construction and operation of two aqueous homogeneous reactors, intended to lead to thermal-spectrum thorium breeder reactors. He would also oversee the development, construction, and operation of two molten-salt reactors, another fluid-fueled reactor that overcame many of the limitations of the aqueous homogeneous reactor, and which many consider to be the ideal implementation of the thorium breeder reactor.

5 Ending the War with Nuclear Weapons

5.1 Technical Triumph at Trinity

By design, General Groves had structured the Manhattan Project so that information did not flow readily from one part of the project to the others. He and a few others kept the larger view of their objectives closely guarded. For this reason, the Metallurgical Laboratory group in Chicago only had snippets of information about what was going on at Los Alamos amongst the group of researchers who were receiving the plutonium from Hanford and trying to make it into a workable nuclear device.

As had been previously mentioned, there was a feeling among some of the Chicago Met Lab team that their efforts had been a failure. Despite their attempts to provide pure plutonium to the group at Los Alamos, they discovered that the plutonium was contaminated with an undesirable isotope—plutonium-240—that might make it unusable. They had worked to design a reactor to convert plutonium to uranium-233 only to discover that they could not build a solid-fueled reactor that would be sufficiently effective for this task. Furthermore, they discovered that any uranium-233 they might make would be contaminated with uranium-232, and would likely be an even worse material for a nuclear weapon than the plutonium from Hanford.

All of these setbacks make what was accomplished at Los Alamos all the more remarkable. Quietly and with tremendous determination, they had developed an entirely new approach to a nuclear weapon based on the technique of implosion, and throughout the winter of 1944 and 1945 they had been systematically testing the implosion technique. If it could work, it would be a much more effective use of plutonium than the previous idea for a gun-type bomb. Because of its efficiency it would require far less fissile material for a given explosive yield. It would also work even with the plutonium contaminated by plutonium-240. But it was a technologically difficult challenge, and there was no fallback position if it failed. Without the development of implosion technology all of the efforts of Fermi, Wigner, Seaborg, and their teams would have been for naught.

At the center of Los Alamos was J. Robert Oppenheimer, who effected a sweeping reorganization of the lab in the summer of 1944 to focus on the scientific issues associated with developing practical weapons. To lead efforts on implosion he chose Robert Bacher and George Kistiakowsky. [1, pg.311] For implosion to work, the secret would be shaped high-explosive charge, called "explosive lenses" which had to detonate at precisely the same time, forcing the plutonium "pit" inside them to be compressed tremendously and quickly. Supercritical multiplication of the neutrons in the pit would cause it to explode like nothing the world had ever seen.

To test the implosion technique, Bacher's team used radioactive lanthanum-140 that had been produced in the X-10 graphite reactor and shipped to Los Alamos by truck. Since the La-140 only had a half-life of 40 hours, it was imperative that it be produced, separated, and transported quickly for use. La-140 decayed to stable cerium-140 and did not pose any long-term radiohazard, but while it was present it emitted high-energy gamma rays. By placing radioactive lanthanum in the center of devices meant to test the implosion principle, various configurations could be tested and assessed.

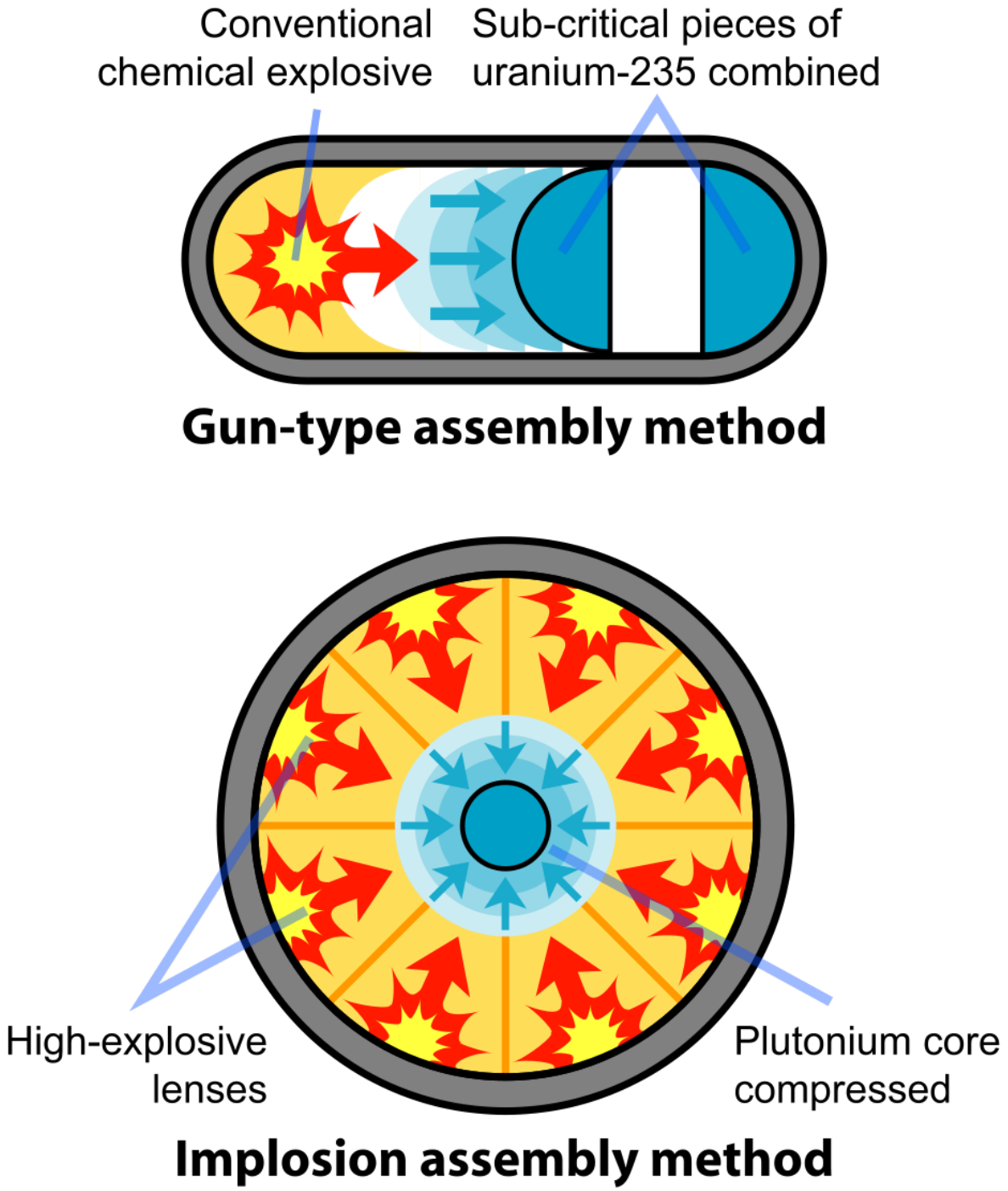


Figure 25: Two types of assembly methods were developed at Los Alamos for the Manhattan Project. Only the implosion technique was suitable for the plutonium that had been produced at Hanford.

The first preliminary tests were conducted in the summer of 1944 but it wasn't until December 14 that one of Bacher's group leaders, Bruno Rossi, saw definite evidence of successful implosion. Time was running out and General Groves was getting very nervous about the prospects for the entire reactor-plutonium-implosion branch of the Manhattan Project. The effort connected with uranium isotope separation and a gun-type bomb seemed far more likely to succeed, but it came with a different cost. The gun-type bomb required roughly ten times the fissile material that the implosion-type bomb needed for the same explosive yield. Nevertheless, Groves committed the entire output of highly-enriched uranium, worth nearly a billion dollars, to the gun-type weapon design.

Further implosion tests in early 1945 were promising, and the arrival of Samuel Allison from the Metallurgical Lab in Chicago sped and focused the work at Los Alamos. Soon there was a focus on having an implosion device ready for test with real plutonium by July 4. It would be called "Project Trinity" and it would be the world's first nuclear detonation. The decision to use precious plutonium in a test was controversial. Some thought that all material should be committed to an actual wartime weapon, but Oppenheimer and most of his division considered the test essential. They felt that the step from theory to reality was too great to risk without a test first, and that the prospect of dropping a plutonium weapon on enemy territory that failed to detonate would give the enemy the chance to collect the plutonium and make their own weapon, without going to the trouble of building expensive production reactors and chemical purification facilities.

The test site for Project Trinity was the Jornada del Muerto (Journey of Death) Valley in the northwest corner of the Alamogordo Bombing Range about 100 miles south of Albuquerque, New Mexico.

On February 5, 1945, the first delivery of plutonium to Los Alamos from Hanford was made, and the second plutonium-producing reactor (D-reactor) reached its rated power level. [9, pg.488] On February 16, the third Hanford plutonium production reactor (F-reactor) was charged with uranium slugs, [9, pg.501] and achieved criticality on February 25. [9, pg.515] On March 9, F-reactor also reached its rated power level. [9, pg.534] These two reactors and the original B-reactor would produce the plutonium used in the Trinity test and later in the weapon used on Nagasaki.

Fabricating the plutonium nitrate that was delivered from Hanford into plutonium metal for the weapons was challenging. Plutonium was very toxic and great care had to be taken against inhalation. It also had a bewildering number of different metallic states, each with a different density and varying properties. By April 1945 the prospects for successful implosion continued to improve as the radioactive lanthanum experiments continued.

One of the final prepartions for the Project Trinity test came on May 7, when Kenneth Bainbridge's task force detonated 100 tons of high explosive on a tower similar to the one that would be used for the actual weapon test. The explosives were spiked with 1000 curies of fission products from Hanford. The explosion was seen sixty miles away.

General Groves had ordered a bit of insurance against failure in the Trinity test by having a giant steel canister called "Jumbo" constructed that would be fitted around the weapon. If the high explosives detonated but failed to trigger the nuclear detonation, then it was thought that Jumbo would contain the fragments of the device and allow the valuable plutonium to be recovered. If the test was successful, however, Jumbo would be incinerated.

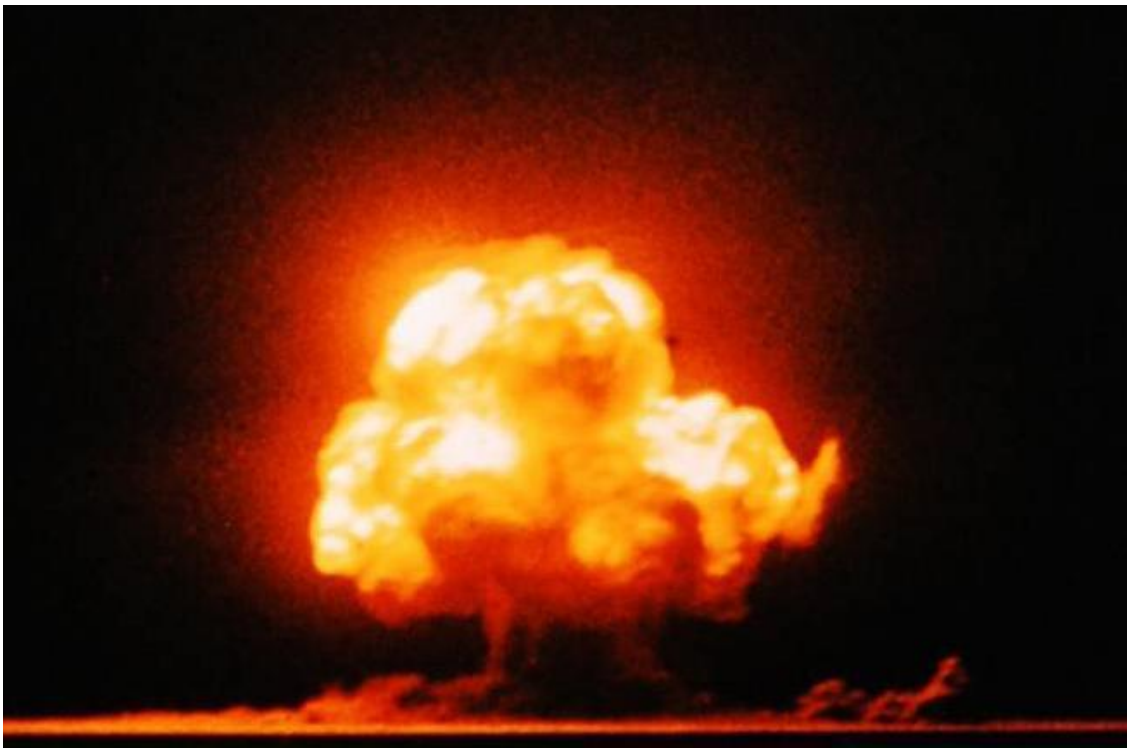


Figure 26: The detonation of the first nuclear device on July 16, 1945.

As the July 4 test date drew closer, it was clear that the date would slip, first to July 13, and then to July 16. On the morning of July 16, very early, a crucial weather report indicated that the test could proceed. Scientists, engineers, and military officials were watching the test from a site about five miles away. A twenty-minute countdown began, and at 5:29am the nuclear device exploded with an explosive equivalent of about 20 kilotons of TNT. The scientists had become confident that Jumbo would not be needed, and had it installed on a tower 800 yards away from the tower that held the device, nicknamed "the Gadget". The explosive force destroyed Jumbo's tower, but Jumbo survived.

The explosive shock wave took 40 seconds to reach the observers five miles away and was ultimately felt for a hundred miles. Oppenheimer later said that as he watched the test he was reminded of a Hindu scripture, "Now I am become Death, the destroyer of worlds."

The next day, Tuesday, July 17, the Chicago Sun dutifully reported the cover story issued by the Army on page 3 of their newspaper:

> SHELL DUMP EXPLODES
> Alamogordo, New Mexico, July 16 - (UP) - A magazine containing high explosives, gas and other shells blew up at the Army base here today with a pyrotechnic display visible 200 miles away. No one was injured. [10, pg.112]

The remoteness of the Jornada del Muerto Valley had successfully hidden America's secret.

Implosion had worked. Through incredible work and determination, Los Alamos had rescued the efforts of Hanford and the Met Lab in Chicago and now stood ready to deliver a nuclear weapon based on plutonium to the Japanese.

5.2 Scientists Concerns about Nuclear Energy

For many months, the scientists and engineers of the Met Lab had been worrying about how the world would respond to the first use of a nuclear weapon.

Leo Szilard, the original worrier who had drafted the letter to President Roosevelt that he had convinced Albert Einstein to sign, was ahead of the rest in his list of concerns for a future world with nuclear weapons. In an afternoon meeting on March 10, 1945, Seaborg recorded:

> Szilard spoke about his concern that the first bomb that we detonate may start a race in atomic armaments between us and other nations, in particular, Russia. He believes that for the safety of this nation it is necessary for us, within the next couple of years, to produce ten times as much fissionable material as is now planned (*i.e.*, ten tons instead of one ton) in order to insure an agreement with Russia to prevent its future use in war. [9, pg.536]

A few days later, Szilard prepared a draft memorandum where he proposed that the United States produce "ten tons of heavy elements within the next few years" in order to convince the Russians that they would have no hope of catching up to US technology in this area. He believed that by convincing the Russians of overwhelming US superiority that they would agree to a system of international control on the manufacture of "heavy elements" anywhere in the world. [9, pg.536-537] Farrington Daniels, an advocate for high-temperature, gas-cooled, pebble-bed reactors, agreed with Szilard and called for substantial expansion of fissile production capability. [9, pg.537]

On May 29, James Nickson, an M.D. with the Met Lab who was part of the Met Lab's Committee of Social and Political Implications, wrote a memo to his section chief, Leon Jacobson, in which he stated:

> Therefore it seems to me that one of the most important functions at hand is the education of the general public to the fact that the era of atomic power is at hand. Further, the implications of this fact must become general knowledge. If the public, as a whole, is not entirely conversant with the broad implications of the field of nuclear physics, it seems reasonably certain that the public will not accept any organization which attempts to take into account the implications of these facts. [10, pg.38]

Seaborg was also a member of this committee, and on June 6 he asked the opinions of the men in his group about possible alternatives in the use of the bomb.

> (1) not use the weapon in this war but simply announce at the end of the war that we have it; (2) tell the world immediately that we have it and give a demonstration at

which representatives of all countries, including Japan, are invited to see it; (3) make bombs as fast as possible and use them on Japan. I added that the committee's opinion is to follow the second procedure.

In the discussion which followed, one man said he would like to see no. 3 used on the basis that we should treat it as just another weapon. I asked if there was not a certain moral aspect that we must consider. **If we are the first to use the weapon and then want to build up a world organization that would never allow its use again, our position would be very much weakened.** I also mentioned that it might be important to give a demonstration from the point of view of keeping our allies' good will; this would be particularly important in the case of Russia who is our most likely future antagonist in view of Russia's great future potentialities as an industrial power.

The division of opinion of the people present was mostly between the possibilities of no. 2 and no. 3. Most people agreed that a strong international organization is necessary but that some precaution must be taken to prevent capturing the whole available supply of fissionable material by that organization. Some expressed pessimism to the possibility of forming a sufficiently strong organization to control the problem, but everyone agreed that it was essential to make the effort. [10, pg.51-52]

Their committee issued a report on June 11, and its summary stated:

The development of nuclear power not only constitutes an important addition to the technological and military power of the United States but also creates grave political and economic problems for the future of this country.

Nuclear bombs cannot possibly remain a "secret weapon" at the exclusive disposal of this country for more than a few years. The scientific facts on which their construction is based are well known to scientists of other countries. Unless an effective international control of nuclear explosives is instituted, a race for nuclear armaments is certain to ensue following the first revelation of our possession of nuclear weapons to the world. Within ten years other countries may have bombs, each of which, weighing less than a ton, could destroy an urban area of more than ten square miles. In the war to such an armaments race is likely to lead, the United States, with its agglomeration of population and industry in comparatively few metropolitan districts, will be at a disadvantage compared to nations whose population and industry are scattered over large areas.

We believe that these considerations make the use of nuclear bombs for an early unannounced attack on Japan inadvisable. If the United States were to be the first to release this new means of indiscriminate destruction upon mankind, she would sacrifice public support throughout the world, precipitate the race for armaments, and prejudice the possibility of reaching an international agreement on the future control of such weapons.

> Much more favorable conditions for the eventual achievement of such an agreement could be created if nuclear bombs were first revealed to the world by a demonstration in an appropriately selected uninhabited area.
>
> In case chances for the establishment of an effective international control of nuclear weapons should have to be considered slight at the present time, then not only the use of these weapons against Japan, but even their early demonstration, may be contrary to the interests of this country. A postponement of such a demonstration will have in this case the advantage of delaying the beginning of the nuclear armaments race as long as possible. If, during the time gained, ample support can be made available for further development of the field in this country, the postponement will substantially increase the lead which we have established during the present war, and our position in an armament race or in any later attempt at international agreement would thus be strengthened.
>
> On the other hand, if no adequate public support for the development of nucleonics will be available without a demonstration, the postponement of the latter may be deemed inadvisable, because enough information might leak out to cause other nations to start the armament race, in which we would then be at a disadvantage. There is also the possibility that the distrust of other nations may be aroused if they know that we are conducting a development under cover of secrecy, and that this will make it more difficult eventually to reach an agreement with them.
>
> If the government should decide in favor of an early demonstration of nuclear weapons, it will then have the possibility of taking into account the public opinion of this country and of the other nations before deciding whether these weapons should be used in the war against Japan. In this way, other nations may assume a share of responsibility for such a fateful decision.
>
> To sum up, we urge that the use of nuclear bombs in this war be considered as a problem of long-range national policy rather than of military expediency, and that this policy be directed primarily to the achievement of an agreement permitting an effective international control of the means of nuclear warfare.
>
> The vital importance of such a control for our country is obvious from the fact that the only effective alternative method of protecting this country appears to be a dispersal of our major cities and essential industries. [10, pg.57-59]

On June 13, Seaborg sent Ernest Lawrence his opinions and suggestions on the course to be taken for nuclear weapons in the immediate future and on the question of the postwar future. He indicated that the opinions he expressed were shared almost unanimously by the people associated with him in his division of the Met Lab.

> (1) The basic facts concerning the successful release of nuclear energy and its immense destructive possibilities should be made public very soon, both to the general public

and to scientific channels. I propose withholding only information with respect to the actual detailed designs of the major manufacturing installations.

(2) With respect to use in the present war, we propose not to use the weapon on Japan without warning but to demonstrate the weapon in the presence of all leading countries including Japan.

(3) On postwar control, I indicate we favor free research in nucleonics throughout the world and control through an international organization. Probably the best method of control lies in the control of the raw materials. Perhaps the only method of maintaining these controls would involve world-wide pooling to form a stockpile of fissionable material to be used by the international organization for policing purposes. **I mention that, as suggested by Szilard, perhaps control could be effected by denaturization, *i.e.*, by mixing it with suitable isotopes to spoil its use for explosive purposes without interfering too much with its use for research purposes such as power pile developments.**

(4) With respect to the organization of postwar research in nucleonics in this country, I state that it would be a good idea to establish, with government aid, about four large research laboratories at four of the major universities. These laboratories should form a sort of a foundation for the country's research program and should include men who are able and willing to advise outlying laboratories as to the research program. The outlying laboratories might consist of government laboratories working on the more practical aspects of the field, and also regular university and industrial laboratories supported by government contracts or grants-in-aid. [10, pg.62]

On June 16, another committee on which Seaborg served, devoted to a consideration of future research, reported out to program management, and they struck a more hopeful tone about the future. They notably describe how the current approach to plutonium production was highly wasteful of uranium resources and was unsustainable in the future. They also pointed out the importance of moving to "breeder" reactors to consume most of the uranium-238 and thorium in order to bolster the long-term argument for nuclear energy.

The field of nucleonics has reached the stage where it is fairly clear as to what should be the next immediate steps. The section of this report dealing with the social and political problems indicates several choices which face the nation concerning the future of this field. It is believed that irrespective of the choice made and irrespective of whether or not the point of view is one which envisages an armament race or is one which happily contemplates a peaceful development, the immediate problem is unaltered. The time scale, however, on which the work is carried out will be very much affected by the realities of international policies.

The quantity production of fissionable isotope is now realized only at the expense of a raw material which is not at all abundant in our country and exists in rather limited quantities in the world as a whole. At the present rate of destruction of

this material it cannot be imagined that a wide-spread use of fissionable isotope will be permitted. The wasteful utilization of this raw material is forced upon us by the necessities of war, but the advisability of continuing present methods of production far into the future even for military needs can be doubted. It follows that research directed towards a more efficient utilization of the primary material is the main problem of the nucleonics program.

If a sizeable percentage of the isotope uranium-238 could be burned and if, in addition, useful fissionable isotope could be realized from thorium, then the whole future of the development would be on much more secure grounds. It is of paramount interest, therefore, that the fundamental research necessary to the design and construction of "breeder" piles be undertaken vigorously. It is only by demonstrating the practicability of the "breeder" principle that a sufficiently ample supply of fissionable material can be produced to permit the nucleonics program to proceed on the scale indicated by the benefits to be derived. It is also necessary that research and development leading to the useful utilization of the power from nuclear burning be vigorously supported. The ultimate goal here would be to operate machines which produce useful power and which at the same time produce as much or more fissionable isotope than is consumed. Under these circumstances no real limit could exist to the application of nucleonics to all phases of our scientific and industrial life.

The accomplishment of the goal set in the previous paragraph is not possible without an extensive program for basic research in physics, chemistry, and metallurgy. The present state of the art is such that new designs of reactor units invariably suggest situations about which no information is available. For the long term development of the field the research in the basic sciences will be of more importance than any immediate pile design or engineering. It is possible that the real future of atomic power does not involve the burning of uranium but rather other elements at the light end of the periodic table. Progress in this direction can only come if all possible freedom and support is given for investigations concerning the nucleus." [10, pg.65-66]

5.3 The World's Terrifying Introduction to Nuclear Energy

President Truman and the wartime allies, including Josef Stalin and Winston Churchill, were attending a conference in Potsdam, Germany, when Truman's War Secretary, Henry Stimson, received the message that the Trinity test had been successful on July 18. [1, pg.386] Elated by the news, Stimson informed the President, who later that afternoon passed the information on to Churchill. Both men felt confident that nuclear weapons—now proven to work—would force Japan into unconditional surrender. Churchill considered the weapon "a miracle of deliverance," which was fitting considering that both men were contemplating hundreds of thousands of Allied deaths, along with millions of Japanese soldiers and civilians, that would accompany an invasion of Japanese home islands, which at that time was contemplated for November 1945.

Truman and Churchill hesitated to divulge to Stalin the success at Trinity, but they realized that some attempt must be made to appraise Stalin of the nature of what was soon to come. On July

Figure 27: Winston Churchill, Harry Truman, and Josef Stalin at the Potsdam Conference in late July 1945.

24, after a plenary meeting had adjourned, Truman strolled over casually to Stalin and without his interpreter present informed Stalin that the United States had a new weapon of unusual destructive force. Stalin showed no special interest, but said only that he was glad that the US was in possession of such a device and that he hoped America would make good use of it against the Japanese. Truman felt relieved that he had taken the minimum step necessary to inform Stalin of what was to come. [1, pg.394]

The notion of using one of the weapons in a public demonstration intended to frighten the Japanese to surrender had been considered and rejected. Several scientists thought that no demonstration would be sufficiently dramatic to convince the Japanese. Further objections emerged. The weapon might fail to detonate, allowing an enemy to collect the fissile material, produced at such staggering expense, and fabricate a weapon of their own. Another possibility would be that the Japanese would place American prisoners-of-war in the area where such a public demonstration had been announced. Yet another possibility was that the Japanese would do everything possible to shoot down the plane carrying the demonstration weapon. Thus the war leaders concluded there would be no warning beyond the ultimatum they planned to issue. Its use in combat would be the world's introduction to the nuclear weapon. [1, pg.358]

The same day that Truman inferred the existence of nuclear weapons to Stalin, War Secretary

Figure 28: The B-29 bomber "Enola Gay" on Tinian island.

Stimson authorized the 509th Composite Group to prepare for attack on Japan as soon as August 3rd. The 509th Composite Group had been specially created to train for and execute nuclear bombing on Japan. Several cities had been held back from attack by conventional bombing campaigns so that they might be attacked by nuclear weapons. These were Hiroshima, Kokura, Niigata, and Nagasaki. Each was now cleared for attack by the planes of the 509th, and the powerful weapons they would carry. [1, pg.394]

On July 27, the Allied leaders in Potsdam issued their ultimatum to Japan for its unconditional surrender, known as the Potsdam Proclamation. [10, pg.128] It demanded an immediate cessation of hostilities by the Japanese, the disarmament of their military, and a restriction of their sovereignty to the Japanese home islands. It made clear that the Allies did not intend the destruction of Japan as a race or a nation. Three days later, Japanese Premier Suzuki rejected the ultimatum, deeming it unworthy of official notice. [10, pg.131] Japan had exhausted the patience of the Allies, who now would end the war with the "terrible swift sword" of nuclear weapons.

On the island of Tinian in the Pacific, Colonel Paul Tibbets of the 509th Composite Group supervised the loading of the first nuclear weapon, nicknamed "Little Boy", into the weapons bay of his B-29 bomber, which he had named after his mother: "Enola Gay." "Little Boy" was a gun-type weapon using highly-enriched uranium. This type had never been tested before—the Trinity test had been a test of the plutonium-implosion-type weapon. Nevertheless there was high confidence that "Little Boy" would work successfully. It was 120 inches long and weighed nearly 10,000 pounds.

On the morning of August 6, at 8:15am local time over Hiroshima, Japan, Tibbets' B-29 dropped "Little Boy" from an altitude of 31,000 feet onto the central region of the city of Hiroshima. After falling for 45 seconds and at an altitude of about 2000 feet, the weapon detonated with an explosive force estimated at 13,000 tons of TNT equivalent. The force of the blast killed 66,000 people in Hiroshima and injured 69,000 more. Almost everything within a one mile radius of the blast was completely destroyed, except for some highly reinforced concrete structures. The blast ignited a firestorm roughly two miles in diameter, which continued burning outward so long as fuel in the form of wood and flammable materials were available. Accurate casualty figures were

Figure 29: The Potsdam Conference in late July 1945.

Figure 30: The "Little Boy" highly-enriched-uranium "gun" type bomb.

impossible to determine, since many of the victims of the blast were cremated by the firestorm. The altitude of the detonation limited the radioactive fallout that was generated and no bomb crater was formed. Many were likely close enough to the blast to be harmed by neutron and gamma radiation, but they were killed in the firestorm before radiation effects would have been apparent.

Secretary of War Stimson was awakened at 7:45 Eastern time to a report that the Hiroshima bombing mission had been successful. It was now time to tell the world about nuclear weapons. Stimson authorized the notification of President Truman and the release of prepared remarks that would accompany the successful use of nuclear weapons in combat. [1, pg.402]

5.4 "A Rain of Ruin from the Air"

At eleven a.m. on August 6, 1945, radio stations around the United States began broadcasting a message from President Truman: [10, pg.143-145]

> Sixteen hours ago an American airplane dropped one bomb on Hiroshima, an important Japanese army base. That bomb had more power than 20,000 tons of T.N.T. It had more than two thousand times the blast power of the British "Grand Slam" which is the largest bomb ever yet used in the history of warfare.
>
> The Japanese began the war from the air at Pearl Harbor. They have been repaid many fold. And the end is not yet. With this bomb we have now added a new and revolutionary increase in destruction to supplement the growing of our armed forces. In their present form these bombs are now in production and even more powerful forms are in development.
>
> It is an atomic bomb. It is a harnessing of the basic power of the universe. The force from which the sun draws its power has been loosed against those who brought war to the Far East.
>
> Before 1939, it was the accepted belief of scientists that it was theoretically possible to release atomic energy. But no one knew any practical method of doing it. By 1942, however, we knew that the Germans were working feverishly to find a new way to add atomic energy to the other engines of war with which they hoped to enslave the world. But they failed. We may be grateful to Providence that the Germans got the V-1's and the V-2's late and in limited quantities and even more grateful that they did not get the atomic bomb at all.
>
> The battle of the laboratories held fateful risks for us as well as the battles of the air, land, and sea, and we have now won the battle of the laboratories as we have won the other battles.
>
> Beginning in 1940, before Pearl Harbor, scientific knowledge useful in war was pooled between the United States and Great Britain, and many priceless helps to our victories have come from that arrangement. Under that general policy the research on the atomic bomb was begun. With American and British scientists working together we entered the race of discovery against the Germans.
>
> The United States had available the large number of scientists of distinction in the many needed areas of knowledge. It has the tremendous industrial and financial resources necessary for the project and they could be devoted to it without undue impairment of other vital war work. In the United States the laboratory work and the production plants, on which a substantial start had already been made, would be out of reach of enemy bombing, while at that time Britain was exposed to constant air attack and was still threatened with the possibility of invasion. For these reasons Prime Minister Churchill and President Roosevelt agreed that it was wise to carry on the project here. We now have two great plants and many lesser works devoted to the production

of atomic power. Employment during peak construction numbered 125,000 and over 65,000 individuals are even now engaged in operating the plants. Many have worked there for two and a half years. Few know what they have been producing. They see great quantities of material going in and they see nothing coming out of these plants, for the physical size of the explosive charge is exceedingly small. We have spent two billion dollars on the greatest scientific gamble in history—and won.

But the greatest marvel is not the size of the enterprise, its secrecy, nor its cost, but the achievement of scientific brains in putting together infinitely complex pieces of knowledge held by many men in different fields of science into a workable plan. And hardly less marvelous has been the capacity of industry to design, and of labor to operate, the machines and methods to do things never done before so that the brain child of many minds came forth in physical shape and performed as it was supposed to do. Both science and industry worked under the direction of the United States Army, which achieved a unique success in managing so diverse a problem in the advancement of knowledge in an amazingly short time. It is doubtful if such another combination could be got together in the world. What has been done is the greatest achievement of organized science in history. It was done under high pressure and without failure.

We are now prepared to obliterate more rapidly and completely every productive enterprise the Japanese have above ground in any city. We shall destroy their docks, their factories, and their communications. Let there be no mistake; we shall completely destroy Japan's power to make war.

It was to spare the Japanese people from utter destruction that the ultimatum of July 26 was issued at Potsdam. Their leaders promptly rejected that ultimatum. **If they do not now accept our terms they may expect a rain of ruin from the air, the like of which has never been seen on this earth.** Behind this air attack will follow sea and land forces in such numbers and power as they have not yet seen and with the fighting skill of which they are already well aware.

The Secretary of War, who has kept in personal touch with all phases of the project, will immediately make public a statement giving further details.

His statement will give facts concerning the sites at Oak Ridge near Knoxville, Tennessee, and at Richland near Pasco, Washington, and an installation near Santa Fe, New Mexico, Although the workers at the sites have been making materials to be used in producing the greatest destructive force in history they have not themselves been in danger beyond that of many other occupations, for the utmost care has been taken of their safety.

The fact that we can release atomic energy ushers in a new era in man's understanding of nature's forces. Atomic energy may in the future supplement the power that now comes from coal, oil, and falling water, but at present it cannot be produced on a basis to compete with them commercially. Before that comes there must be a long period of intensive research.

> It has never been the habit of the scientists of this country or the policy of this Government to withhold from the world scientific knowledge. Normally, therefore, everything about the work with atomic energy would be made public.
>
> But under present circumstances it is not intended to divulge the technical processes of production or all the military applications, pending further examination of possible methods of protecting us and the rest of the world from the danger of sudden destruction.
>
> I shall recommend that the Congress of the United States consider promptly the establishment of an appropriate commission to control the production and use of atomic power within the United States. I shall give further consideration and make further recommendations to the Congress as to how atomic power can become a powerful and forceful influence towards the maintenance of world peace.

Truman's terrifying message to the world had surprisingly little effect on the Japanese deliberations about surrender. The attack on Hiroshima severed communication with Tokyo, and it was not until the evening of August 6th when the Japanese High Command received word that a small number of planes had wrought appalling damage on the city. It was not until the next day when they learned that a single plane had destroyed the entire city. Military planners could scarcely believe what they had heard, and many did not. Several high ranking officials sought an audience with the Emperor that they might advise him to accept the Potsdam Proclamation. Truman's radio broadcasts were dismissed as scare propaganda. [1, pg.403]

Japanese Ambassador Sato to the Soviet Union sought their intervention to mediate a ceasefire between the Allies and Japan. His request was rejected by the Soviet diplomat Molotov, who tersely informed Sato that by rejecting the Potsdam Proclamation the USSR would declare war on Japan, and that a state of war would exist between the two nations by August 9th. [1, pg.403]

5.5 Seaborg's Plutonium Ends the War

The declaration of war by the Soviets was a staggering blow to the Japanese, and strengthened the position of those leaders who were pushing for the unconditional surrender demanded by the Allies. As they met on the morning of August 9th, they argued about specific points of the surrender, and the meeting ended in deadlock even as news came of the destruction of another city, Nagasaki, by nuclear weapons.

Another B-29 from the 509th Composite Group, this one called "Bockscar" and commanded by Major Charles Sweeney, had taken off from Tinian for an attack on the Japanese city of Kokura. Clouds obscured the city, and Sweeney turned to his alternate target, the city of Nagasaki. On his B-29 was a plutonium-implosion weapon called "Fat Man", very similar to the weapon that had been tested in the Trinity Project. "Fat Man" was dropped and fell for 43 seconds before detonating at an altitude of 1650 feet above a military factory in Nagasaki. Forty thousand people were killed and 25,000 more were injured, which was the third highest fatality rate of any military attack in World War II, after the nuclear strike in Hiroshima and the firebombing of Tokyo on March 9th and 10th, which killed over 100,000 people.

Figure 31: The "Fat Man" plutonium implosion bomb.

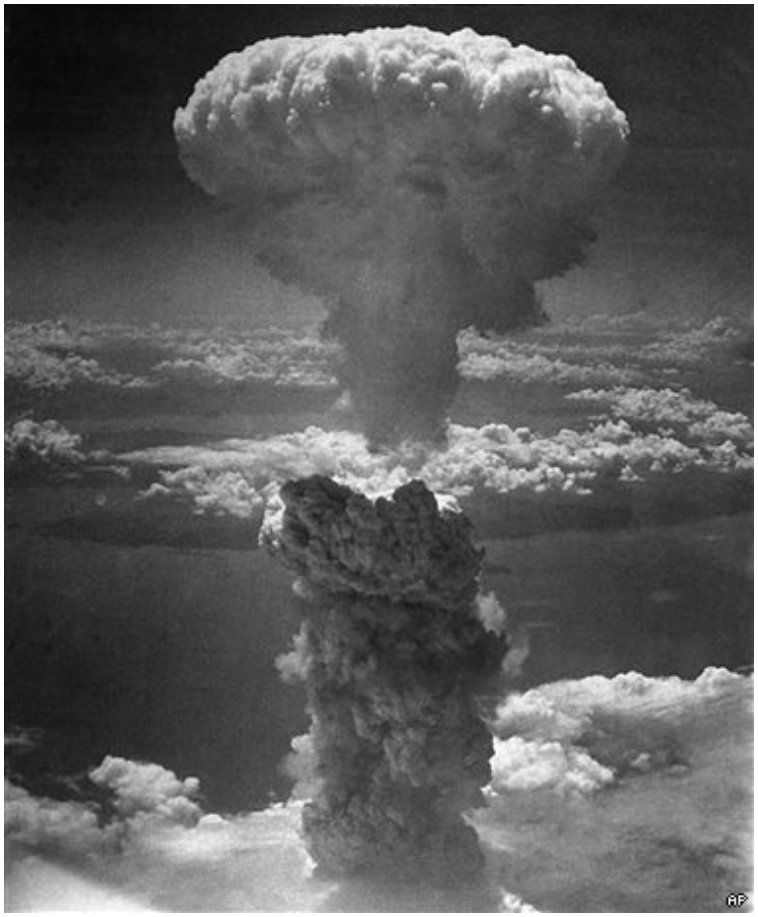


Figure 32: The explosive plume formed by the detonation of the "Fat Man" bomb over Nagasaki.

The Japanese High Command reconvened shortly after midnight and deliberated for two hours. Views were stated, and Premier Suzuki rose and asked Emperor Hirohito to indicate his wishes. Hirohito made a short speech and declared that he agreed that the war should end. The imperial decree was accepted by all present and the Cabinet met at once and ratified the decision. By the morning of August 10th, Japanese radio was broadcasting the acceptance of the Potsdam declaration and Japan's intention to unconditionally surrender. Truman swiftly notified the 509th to suspend any efforts to drop a third nuclear weapon on the Japanese unless expressly ordered to do so. The war was over. [1, pg.404-405]

An element that did not even exist on Earth five years earlier had been fashioned into a weapon that had ended the greatest military conflict in human history. Glenn Seaborg's penetrating curiosity, coupled with Ernest Lawrence's mighty cyclotron, had led to its discovery. An industrial effort of staggering complexity and cost had brought it to fruition. And its awesome power had finally convinced the Japanese warlords to abandon their belligerence.

Uncharacteristically, Seaborg said almost nothing in his journal about his thoughts or feelings on the day that plutonium was used to destroy Nagasaki and end the war. Did he feel pride that his hard work had brought the war to a close and saved millions of lives? Or did he think of the tens of thousands who were incinerated that day in "the rain of ruin from the air"?

Much has been said over the years about the guilt that nuclear scientists felt about the creation of nuclear weapons, but in the final analysis, are they really any different than the hundreds of thousands who labored to produce conventional chemical munitions? Were not more killed in the firebombings of Tokyo or Dresden by chemical weapons than were killed at Hiroshima and Nagasaki by nuclear weapons? Is the morality of war in any way related to the rate or ease at which adversaries destroy one another? The distinguishing factor of the nuclear weapon was that it represented a quantum leap in man's ability to inflict death and destruction against an enemy.

War is cruel, hateful, bitter, and irredeemable. And sometimes it is also unavoidable. But history has shown that since that fateful day of August 9, 1945, the mighty nations of the world have never lifted up arms against one another in the style of world war that had twice ravaged the planet in the first half of the twentieth century. Perhaps Seaborg's plutonium wasn't just a weapon; perhaps it was an antidote, forcing the leaders of the earth to confront the inevitable results of their actions before hostilities began. If such a thing is true than few things have saved so many lives as the invention of nuclear weapons.[7]

[7] Author's note: I once spoke with Dr. Kazuo Furukawa, who has since passed away, about the events of Hiroshima and Nagasaki. He described to me how he felt, as a sickly youth of seventeen, when he was told that he would be defending the Japanese home islands against an American invasion. He was expected to give his life repelling the invaders, and he was thoroughly resigned to the idea that he would soon die. When he learned of the nuclear bombings, he realized his government would capitulate and that he would live. He told me that he decided that he would learn all he could about nuclear technology and how to turn it into a source of peace and prosperity. His career as a nuclear technologist and advocate is an example of one of the lives saved by the events of early August 1945. Without the swift surrender of the Japanese government, millions of Japanese and hundreds of thousands of Americans would have certainly died.

6 The Postwar Environment

6.1 Exploring New Possibilities

The exigencies of war had consumed the intellectual capabilities of some of the most brilliant scientists of the world since the discovery of nuclear fission in 1938. With the news of the Japanese surrender, an opportunity finally existed to investigate so many other branches of the tree of possibilities that had been planted by the tremendous wartime investment in reactors, chemical processing, and neutronic applications. Several of these would have direct bearing on future efforts to utilize thorium as a nuclear fuel.

1. There was great anticipation that new political and scientific organizations would be formed to deal with matters of nuclear research and implementation. In years to come, these were formed across the world, and many of them began investigations into thorium as a source of energy. The most extensive was the Atomic Energy Commission in the United States, which funded several efforts into the development of thorium-based nuclear reactors.

2. Naval officers had been thinking about the application of a nuclear reactor to power a submarine that would never need to surface for air and would have unique military capabilities. They would succeed in 1954, and many years later, one of the reactors that descended from this effort would become the first to be completely powered by thorium and uranium-233, conclusively demonstrating that thermal breeding was possible.

3. Air Force officers, having heard of the triumphs of the Manhattan Project, began to think about using nuclear reactors to power aircraft. This would later lead to a program that would develop a high-temperature nuclear reactor using a fluid fuel composed of fluoride salts. Later this technology would be combined with the thorium fuel cycle to form what many believe to be the most compelling implementation of a thorium-fueled reactor.

4. The scarcity of fissile material and the proven value of plutonium for nuclear weapons would lead efforts on a breeder reactor away from the fluid-fueled thorium breeder that Wigner and his team envisioned to a solid-fueled, liquid-metal-cooled, fast-spectrum reactor that would be designed to produce much more plutonium than it consumed. The appeal of plutonium for the weapons program would lead to further emphasis on fast-breeder reactors, which would lead them to be chosen over thorium reactors as the focus of breeder reactor research.

5. Nuclear testing, which was truncated to a single event (Trinity) to maximize options against Japan, would resume and continue for many decades. Other nations would join the race to develop larger and more terrifying nuclear weapons. Above-ground detonation of nuclear explosives would release fission products that would make their way into the bodies of all organisms on Earth. Although largely harmless, this potent fact would lead to public revulsion against nuclear weapons testing which would catalyse general anti-nuclear sentiment. Even after testing was ended, anti-nuclear organizations would regroup around the opposition to civilian nuclear power stations, which would lead to a tremendous reduction in the

growth of nuclear energy. This in turn would reduce the compelling need to realize thorium as a new form of energy.

6. Glenn Seaborg originally created uranium-233 in order to explore a nuclear decay chain that no longer existed on Earth. With larger amounts of uranium-233 created in reactors, this decay chain was fully explored and found to possess unique attributes that differentiated it from the other three decay chains. It also contained radioactive isotopes of compelling value for science and medicine.

But one branch of the tree of possibilities that might have seemed so obvious in August 1945 never came to pass: **uranium-233 did not enter into the nuclear stockpiles of the world.** Although improvements in the production rate of uranium-233 and a reduction of the contamination from uranium-232 might have been realized through concerted effort, further research on uranium-233 as a material for weapons did not take place, and the tens of thousands of nuclear weapons that were built in the decades to come were based exclusively on highly-enriched uranium and plutonium. There was only one known test of an explosive containing uranium-233 in 1954, and its result was dubious[8].

6.2 Understanding and Recognition at Last

Shortly after word of the Japanese surrender reached the personnel of the Met Lab, director Farrington Daniels had to warn the employees of the Met Lab that they were still working on nuclear research and that they could say no more than the information that had been publicly released. [10, pg.157-158]

But there was no doubt that a great burden had been lifted from the whole nation. The secret work in Chicago, which had brought together some of the brightest minds in the world to design nuclear reactors, separate new elements using chemistry, and develop material for weapons that would end a world war, had been a resounding success. Nevertheless, tight wartime secrecy prevented Seaborg and others from saying very much about what they had done. The extent of the information they could reveal was bounded by a report issued by the Manhattan Project, written by Henry Smyth, called *General Account of the Development of Methods of Using Atomic Energy for Military Purposes*, now generally called the "Smyth Report". [1, pg.407]

This caused some discontent among Met Lab personnel. Some thought the Smyth report said too much about their work, others thought it said far too little. Speculation was flying throughout the media about the nature of nuclear energy and its implications, and those who knew most could say least. Walter Zinn grumbled, "It's more important than who gets the credit and who does not. If people are going to get the correct information they have to get it from the right people. Washington is really cutting off the right source of information which would educate the public." [10, pg.166-167]

[8] As a part of Operation Teapot on April 15, 1955, a composite plutonium-239/uranium-233 bomb core was tested in a Mark 7 HE assembly. The shot was called the "Military Effects Test" and its purpose was to evaluate the destructive effects of nuclear explosions for military purposes. Specifications called for a device whose yield was calibrated to $\pm 10\%$, but the composite core used in the MET had only 2/3 the yield predicted, a tremendous disappointment.

Work continued on a variety of esoteric subjects related to the chemistry of plutonium and other actinides, but there was a definite sense that the wartime intensity had dropped off. In September 1945, Seaborg pursued a lead from a company in Monterey, California, that said the black sand beaches of Monterey contained up to 60% thorium dioxide in some areas. [10, pg.186] Seaborg felt this was important to relay back to the military leadership of the Manhattan Project. [10, pg.206] He also prepared for his return to Berkeley after over three years in Chicago.

Seaborg was now in demand as a public figure. His youth, intelligence, and photogenic appeal led to requests for him to appear on national radio and television programs. [10, pg.331-334] As the discoverer of plutonium, there was intense public interest in this material and the man who had found it and helped to end the war. He was also asked to contribute articles to the Encyclopedia Brittanica on the subjects of uranium, thorium, artificial radioactivity, protactinium, plutonium, neptunium, and actinium for their 1946 edition. [10, pg.352]

6.3 Legislation for a New Organization

Public realization of the power and import of nuclear weapons led to pronounced debate on who should control the new weapons, and under whose authority they should be developed. On one side were those who felt that the US Army had shown their capability in administering the Manhattan Project, and that they should continue to lead any effort to develop weapons. On the other side were many of the scientists of the Manhattan Project who felt that a power so vast should come under civilian control. This dispute materialized into legislation in the fall of 1945.

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Military or Civilian Control Of Atomic Energy?

The controversy "should atomic energy in America be under military or civilian control" has been brought to a showdown in the Senate Special Committee on Atomic Energy.

On the extreme militaristic side, there is Senator Eugene D. Millikin of Colorado who believes that even General Groves favors too much civilian influence. But in justice to General Groves, his scheme of an Atomic Energy Commission containing four military among its nine members with an active officer as administrator, can be relied upon to bring about complete military control.

The argument in favor of leaving research and development of atomic energy in the hands of the military goes as follows: "The atomic bomb is the most powerful weapon in existence. The applications of atomic energy to peacetime industrial processes can be postponed without endangering our national economy. With the world political situation as it is, we must give first priority to the military problem, and this can best be achieved by leaving the control of atomic energy in the hands of the military.

Secrecy regulations, which military mentality is likely to force upon fundamental scientific research, will cause a paralysis of scientific progress. This paralysis will spread from governmental atomic bomb laboratories to all laboratories working in the nuclear field. The necessity may arise of establishing secret courses in our universities, leaving the majority of students in ignorance of basic facts of their science. With nuclear physics as an opening wedge, the same disintegration may permeate the fields of bacteriology, medicine, and other sciences all of which may be used in the next total war.

Figure 33: There was great public controversy over whether the proposed Atomic Energy Commission would be under military or civilian control.

The first piece of legislation put forward came from the US House of Representatives. Andrew Jackson May, chairman of the House committee, presented a bill that had been drafted by two experienced lawyers that worked for the War Department, Kenneth Royall and William Marbury. [1, pg.428] The bill called for a new organization to oversee nuclear development. It would consist of a nine-man commission, consisting of five civilians, two representatives from the Army, and two

from the Navy. [1, pg.412] These men would serve indefinite terms and would be insulated from political pressures and the President's powers of removal. The commission they envisioned would have custody of all raw materials and deposits, all plants, facilities, equipment, and materials, all technical information and patents, and all contracts and agreements related to the production of fissionable materials. To accomplish this, the commission would have virtually unlimited powers of condemnation and eminent domain. These sweeping powers would give the commission the power to undertake nearly any effort related to the development of nuclear weapons.

The scientists of the Metallurgical Laboratory and other Manhattan Project sites were horrified with the proposed legislation, and felt highly restricted in their ability to speak out by wartime secrecy orders.

Freshman senator Brien McMahon of Connecticut put forward a very different alternative in the US Senate: an Atomic Energy Commission that consisted entirely of civilians, from which military personnel were specifically excluded. McMahon's bill won the support of the scientists and the public at large. It was unanimously passed by the US Senate on a voice vote on Saturday, June 1, 1946. [1, pg.516]

In conference with the House of Representatives, and Congressman May, it was clear that amendments would be added to the bill. These were appended to the bill before it passed the House on Saturday, July 20, with a vote of 265 to 79. [1, pg.529] In conference, the differences between the Senate and House versions of the bill were worked out, usually to the favor of the Senate version, before the bill cleared both chambers and was signed into law by President Truman on Thursday, August 1, 1946. [1, pg.530] It was known as "The Atomic Energy Act of 1946", and it created the Atomic Energy Commission, which would relieve the Manhattan Project of their responsibilities to develop nuclear weapons and other activities.

6.4 Dissolution of the Manhattan Project

In July 1946, the Manhattan Project conducted the public tests of nuclear weapons that had only been contemplated before the end of the war. The location of the tests was a remote island called Bikini Atoll in the Marshall Islands. There an armada of captured Japanese and German warships had been assembled in the harbor. The testing sequence was called "Operation Crossroads" and it would include two weapons tests. The first would be done by bombing the fleet anchored in the center of the atoll by a single bomb dropped from an aircraft. The other test would involve a weapon detonated underwater.

The first test detonation, code-named "Able", took place on July 1, 1946, when a B-29 dropped an implosion bomb on the ghost fleet at Bikini. [1, pg.580] On dozens of Navy ships about thirty miles away, military experts, congressmen, scientists, foreign observers, and an entire shipload of journalists were witnessing the test. The contrast between this test and the handful of of men who saw Project Trinity could not have been more pronounced. For the first time the public was "present" at the detonation of a nuclear device, as the test itself was reported live on the radio to listeners in the United States on CBS radio. On July 24, the underwater detonation, code-named "Baker", took place.

Controversy surrounded the testing at Bikini. Many argued that the tests had little military

Figure 34: President Truman signs the Atomic Energy Act of 1946 on August 1, 1946.

value, and that they were nothing more than a stunt intended to impress or frighten the world. For the Manhattan Project, Operation Crossroads represented nearly the end of the line. With the signing of the Atomic Energy Act of 1946 and the creation of the Atomic Energy Commission, civilian control of nuclear weapons and development would soon be a reality. The Manhattan Project worked to transfer facilities, personnel, and information to the new civilian agency before dissolving as an organization at the end of calendar year 1946.

Figure 35: "Baker", the second detonation at Bikini Atoll as part of "Operation Crossroads", on July 24, 1946.

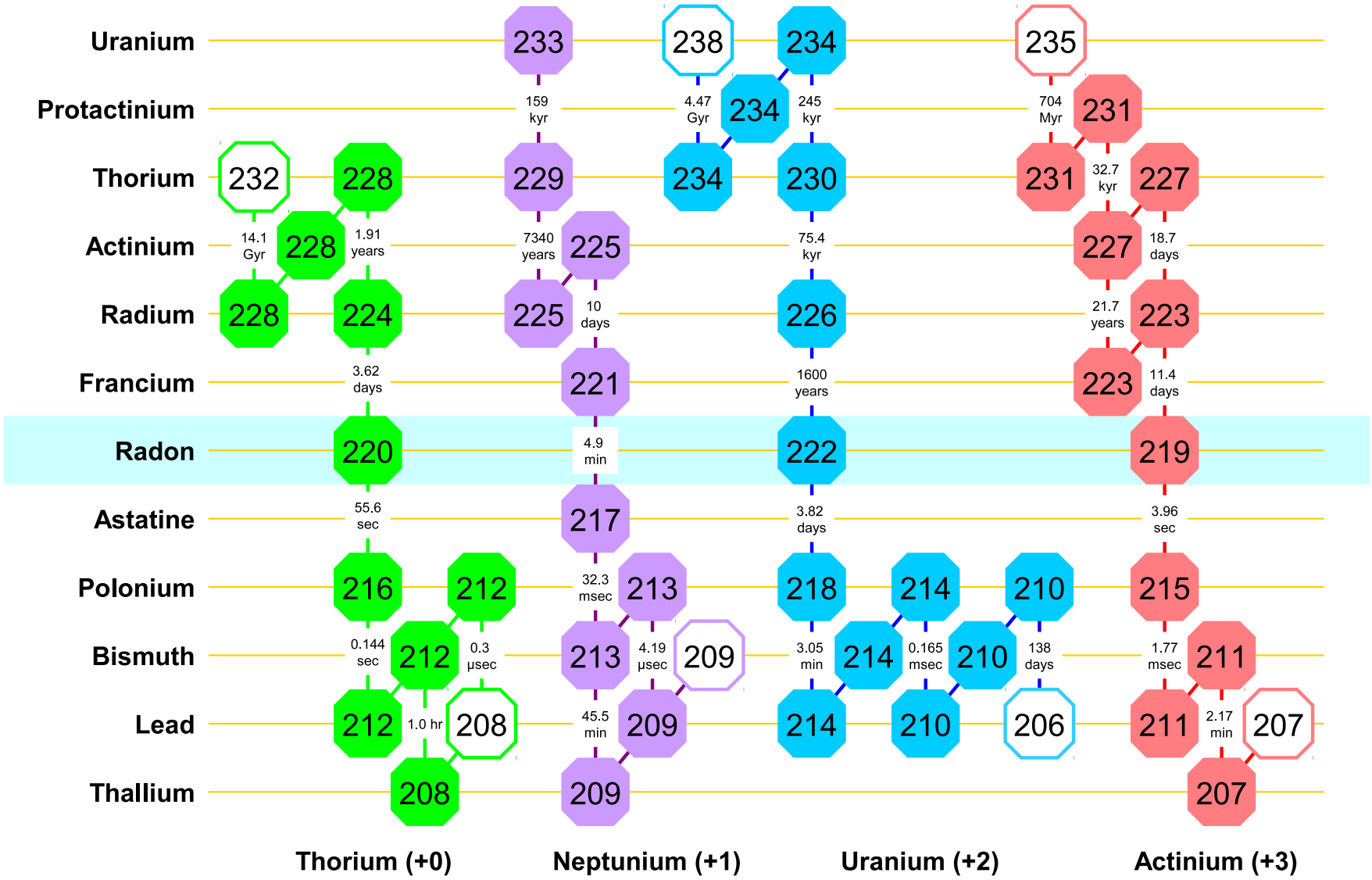

Figure 36: The four decay chains, including the neptunium (+1) chain that was realized through the discovery of uranium-233. This new decay chain was unique in several respects. 1) It did not have an isotope of radon in its decay chain, and 2) it ended on bismuth rather than an isotope of lead.

7 Conclusion

By the end of the Manhattan Project, uranium-233 derived from thorium had been produced, evaluated, and rejected as a nuclear weapons material. Plutonium had also been produced, in far greater quantities, and despite difficulties with production and isotopic purity, it had been made to operate in nuclear weapons. In large part, success with plutonium was due to the development of the implosion method of nuclear detonation. This method also reduced the amount of fissile material needed for a given yield, and was thereafter applied to both plutonium and highly-enriched uranium weapons.

Uranium-233 had superior properties as a nuclear fuel over uranium-235 and plutonium-239 in a thermal spectrum reactor. A fluid-fueled reactor, based on uranium salts dissolved in heavy water, was seriously considered as a thermal breeder reactor. But this concept never got beyond the conceptual stage during the Manhattan Project and it did not offer a significant breeding gain. It was quickly overtaken by an effort to build a fast-spectrum breeder reactor using plutonium and uranium-238 as the fertile material. The fast-breeder held the promise of a significant breeding gain, which was an important consideration in an era when every gram of fissile material was of tremendous worth, and when any diversion of fissile material away from weapons had to be justified as having eventual military value.

The thermal-spectrum, thorium-uranium-233 breeder reactor concept could not promise high breeding gains like a fast-spectrum plutonium reactor could, but it had the great merit of being able to operate in the thermal spectrum and with moderated neutrons. Interest in this reactor later continued at Oak Ridge National Laboratory, where two experimental aqueous homogeneous reactors were built to test the principles that had been sketched out in the final days of the Manhattan Project.

An effort to develop a converter reactor, that would consume plutonium and generate uranium-233, later led to ideas for a pressurized light-water reactor. This reactor type, proposed by Alvin Weinberg, would later power submarines and a decade later would be adapted for civilian use in the United States. It would go on to become the predominant reactor type in the world as of 2013.

Uranium-233 was never developed into an operational nuclear weapon material. One test took place in 1955 of an experimental composite plutonium-uranium-233 core as part of the Teapot series of nuclear tests, but the test had a substandard yield and no further experiments with uranium-233 were continued. The details of the test, along with the relative amounts and arrangements of the plutonium and uranium-233 in the device, remain classified to this day.

Glenn Seaborg, leader of the technical group that discovered and characterized uranium-233, went on to serve as the chairman of the Atomic Energy Commission from 1961 to 1971, serving under Presidents Kennedy, Johnson, and Nixon. He was also at the controls of a molten-salt reactor experiment in 1967 where uranium-233 was first used to power a nuclear reactor, which went on to operate for thousands of hours successfully on this fuel.

The development of thorium as an energy source remains in its infancy in 2013, having progressed relatively little since the era of the Manhattan Project. In large part this is due to an early emphasis on nuclear weapons in the United States, Soviet Union, Great Britain, and France that led to early characterization of uranium and plutonium in reactors and chemical processing plants.

Plutonium was a suitable nuclear weapons material and could be produced in reactors that used natural (unenriched) uranium, and it could be separated chemically. Uranium-233 could not be produced in reactors based only on natural thorium, since thorium did not multiply neutrons. Any effort to produce uranium-233 in reactors using uranium simply diminished the potential production rate of plutonium, and was undesirable.

The origins of uranium-233 in the days before the Manhattan Project, and its investigation and consideration during the wartime era, laid the foundations for how thorium and uranium-233 would be considered today, and have lasting relevance.

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A Extracts from *Early History of Heavy Isotope Research at Berkeley*

1940-09-23 John W. Gofman and Spofford G. English, two graduate students who are starting at Berkeley this fall, have elected to carry out their graduate work under my direction. English has decided that he will not be very active in research during the first semester, but Gofman has elected to begin his research program immediately. Gofman will work in room 307. (In addition to supervision of graduate student research, my teaching assignment consists of teaching two freshman chemistry laboratory sections.)

Gofman has accepted my suggestion that for his thesis problem he undertake the search for the missing 4n+1 heavy nuclei and the measurement of their radioactive properties. He will start with an investigation of Pa233, the protoactinium daughter of the 26-min. Th233 which is produced by neutron capture in thorium.

Hahn and Strassmann (O. Hahn and F. Strassmann, Naturwiss 28, 543 [1940]) have recently pointed out that the radioactivity of 25 days half-life assigned to Pa233, originally reported by Meitner, Strassmann and Hahn (L. Meitner, F. Strassmann and O. Hahn, Zeits f. Physik 109, 538 [1938]) to be the daughter of the 26-minute Th233 produced by neutron capture in thorium, might actually be due to an isotope of zirconium. There is a 25-day zirconium formed in the fission of uranium by neutrons, and hence probably also in the fission of thorium by neutrons. Since Meitner, Strassmann and Hahn used zirconium as the carrier material for their radioactive substance, the question arises as to whether their 25-day radioactivity from thorium plus neutrons might be due to an isotope of zirconium rather than protoactinium.

Gofman will begin by attempting to demonstrate whether this 25-day activity is indeed due to Pa233. Kennedy is collaborating with us in this research, making his main contribution in developing the counting equipment that will be required. [6, pg.2-3]

1940-11-04 After becoming familiar with the chemistry of thorium, protoactinium and uranium, gathering his required chemical equipment and learning about the various relevant counting apparatus, Gofman is getting underway with his Pa233 problem. He is working in room 307, Gilman Hall, the same room in which Wahl and Friedlander are situated. [6, pg.3]

1940-11-10 Gofman made his first bombardment of thorium nitrate today using neutrons produced by the bombardment of phosphorus with deuterons at the 60-inch cyclotron. (The phosphorus target was for another experiment and advantage was taken of the by-product neutrons.) The thorium nitrate was surrounded by paraffin and the bombardment took place from 10:45 a.m. to noon.

Gofman brought the thorium to room 307, dissolved it in water, then added H_2O_2 to precipitate insoluble thorium peroxyhydroxide which he washed with water and alcohol several times. He followed the decay of this using electroscope no. 3 with my help, and we found

that the Th233 in this sample decays with a half-life of approximately 26 minutes as expected. [6, pg.3-4]

1940-11-24 Gofman bombarded about 15 gm of thorium nitrate encased in a large paraffin block for about an hour with the neutrons produced during the bombardment of phosphorus with 100 microamperes of deuterons in the 60-inch cyclotron. The thorium was dissolved and precipitated from acid solution as the iodate after the addition of appropriate carriers for the fission products. After redissolving the thorium iodate in concentrated hydrochloric acid, successive precipitations of zirconium phosphate were performed. If the 25-day activity formed by the slow-neutron bombardment of thorium is due to Pa233 resulting from the beta decay of Th233, then the successive zirconium phosphate precipitates (which coprecipitate the protoactinium) should have decreasing amounts of Pa233 beta radiation. Measurements using electroscope no. 3 do show that the intensities are less for the successive precipitates, consistent with the Pa233 growing from a parent of approximately 26 minutes halflife, presumably Th233. It remains to be demonstrated that beta radioactivity in the zirconium phosphate precipitates will decay with the expected 25-day half-life. [6, pg.4]

1940-12-08 Gofman bombarded 50 gm of thorium nitrate for several hours with the neutrons produced from the bombardment of phosphorus with deuterons at the 60-inch cyclotron in order to produce a stronger sample of the 25-day activity to be used to establish by chemical experiments that it is due to protoactinium. [6, pg.5]

1940-12-13 Gofman began his chemical identification of the 25-day beta activity using material produced in last Sunday's bombardment. He plans to dissolve the thorium nitrate, precipitate zirconium phosphate to carry the 25-day activity, convert this to zirconium oxychloride, and then subject this to fractional crystallization from hydrochloric acid solution. Experiments by Grosse and Booth (A. V. Grosse and E. T. Booth, Phys. Rev. 57, 664 [1940]) have shown that in such a fractional crystallization the protoactinium concentrates in the mother liquor. [6, pg.6]

1940-12-17 Gofman has completed his zirconium oxychloride experiments started last Friday and finds that the specific activities (activity per decigram ZrO_2) of the first fractions are of the order of 5% of the specific activity of the overall sample, showing an accumulation of the activity in the mother liquor as should be the case for protoactinium. This is further evidence that the 25-day activity is due to Pa233. [6, pg.7-8]

1940-12-19 Today Gofman extracted a purified uranium fraction through the precipitation and reprecipitation of sodium uranyl acetate from a solution of thorium nitrate made up of neutron-bombarded thorium. The purpose is to see whether there is any beta-emitting U233 daughter in the Th233 -> Pa233 -> U233 decay sequence. He found no beta particles in the uranium fraction that could be attributed to U233. It is, of course, to be expected that U233 will decay by the emission of alpha particles and we intend to conduct experiments to look for these. [6, pg.8]

1941-01-14 Gofman has followed the decay of the radioactivity in the zirconium phosphate precipitates extracted from the solution containing Th233 on November 24, 1940 and has found in each case a half-life of about 25 days, indicating that Th233 decays to a beta emitter of 25-days half-life, which therefore must be Pa233.

We prepared a letter to the editor describing our work on the identification of the 25-day entitled "Radioactive Isotopes of Protoactinium" under the authorship of G. T. Seaborg, J. W. Gofman and J. W. Kennedy. We are today sending this letter for publication in The Physical Review. In this letter we describe our experiments concerning the genetic relationship of the 25-day activity to its parent Th233, its chemical identification as protoactinium by the fractional crystallization of zirconium oxychloride, the absence of daughter U233 beta activity, and the measurement of the upper energy limit of beta particles as 0.4 MeV. [6, pg.15-16]

1941-02-01 Our letter to the editor entitled "Radioactive Isotopes of Protoactinium" under the authorship of Seaborg, Gofman and Kennedy, appeared in today's issue of The Physical Review (Phys. Rev. 59, 321 [1941]). As a neighboring article in the same issue there is an article by Grosse, Booth and Dunning announcing similar results (Phys. Rev. 59, 322 [1941]). [6, pg.21]

1941-02-21 Today we began the bombardment of 1 kg of thorium nitrate, imbedded in a box containing paraffin, with neutrons at the 60-inch cyclotron with the aim of preparing out largest sample of Pa-233 through an intensive bombardment. [6, pg.23]

1941-03-04 We concluded our bombardment of 1 kg of thorium nitrate, started on February 21, with neutrons at the 60-inch cyclotron. The material in a box filled with paraffin has been in various positions near the target (usually beryllium) bombarded with deuterons, sometimes in the best position and sometimes in a less advantageous position, depending on the competition for neutrons. Exposure corresponds to 9700 microampere-hours of deuterons. [6, pg.27]

1941-03-28 (date of Pu-239 fissionability discovery) During the last month Gofman has carried out a number of experiments to develop his method for the extraction of from the bombardment of our 1 kg of thorium nitrate with neutrons at the 60-inch cyclotron between February 21 and March 4. Starting today he will make his initial extraction by precipitating zirconium phosphate to carry the and then will separate the from the zirconium by precipitating it with lanthanum oxalate under conditions which leave more than 90% of the zirconium in solution, presumably as an oxalate complex. [6, pg.35]

1941-04-10 Gofman has completed the extraction, which he started on March 28, of Pa233 from the 1 kg of neutron-bombarded thorium nitrate. He dissolved the thorium nitrate in water, precipitated zirconium phosphate to carry the Pa233 dissolved this in HF, converted to the hydroxide, dissolved this in HCl, and precipitated lanthanum oxalate which carried the Pa233 while leaving the zirconium in solution. He had to handle numerous fractions and combine precipitates in order to carry out in practice this separation and concentration procedure, which is so simple in principle but difficult in execution; a number of cycles were

carried out. He wound up with the final Pa233 in several fractions which he mounted in order to test for the growth of daughter U233 alpha particles. Two of these fractions have intensities of Pa233 beta radiation of 27 and 40 microcuries, as measured with our FP-54 ionization chamber and using Wahl's calibration of January 11, 1941. Wahl's calibration, 3.1 x 10^4 mm galvanometer deflection per microcurie, with the standard resistance and shunt setting R2S1, is for 93-239 beta radiation and hence its application to beta radiation can give only approximate results, but these are sufficient for our purposes. Gofman will attempt to measure the growth of alpha particles in these Pa233 samples using our screen-windowed ionization chamber setup. [6, pg.37-38]

1941-04-23 A small but discernible quantity of alpha activity has grown into the beta-emitting Pa233 samples isolated by Gofman on April 10. Measurements with our screen-windowed ionization chamber indicate that the 40 microcurie sample shows an alpha counting rate of 2 per minute, and our 27 microcurie sample shows an alpha counting rate of 1.2 per minute. Assuming that the alpha particles are due to daughter U233 (which needs to be proved), taking into account that the 25-day Pa233 has decayed for 13 days, and correcting for the geometrical efficiency of the alpha counting ionization chamber, we can calculate the half-life for the daughter U233, as an average of the two determinations, to be of the order of 100,000 years.

This, of course, is a very rough determination, and should perhaps be regarded as a lower limit. However, it may indicate that we should be able to produce, with more intense neutron bombardments of larger amounts of thorium, sufficient U233 to not only determine its alpha half-life, but also its cross section for fission with slow neutrons. **Of special importance is our demonstration through these results that U233 is sufficiently long-lived to be a practical source of nuclear energy should it be found to be fissionable with slow neutrons and should methods for its large scale production be developed.** [6, pg.38-39]

1941-06-25 During the last couple of months Gofman has been experimenting with methods to prepare by electrolysis thin samples of Pa233 suitable for measuring the growth of daughter U233 alpha activity. He has prepared two samples of strength 6 microcuries and 10 microcuries as measured with our calibrated FP-54 ionization chamber. It is necessary for Gofman to return to his home in Cleveland, Ohio because of our inability to find funds to support him for the summer. He has been supported during the school year on his small stipend as a teaching assistant which is insufficient to take care of his subsistence needs for the remainder of the summer. He is asking Wahl and English to make alpha particle measurements on his electrolyzed 6 microcurie and 10 microcurie samples to look for the growth of daughter U233 during his absence. [6, pg.47]

1941-06-29 Gofman left today to return to his home in Cleveland, Ohio to spend the remainder of the summer before the fall semester opens in August. [6, pg.47]

1941-07-10 Today I wrote to Dr. Lyman J. Briggs at Professor Lawrence's suggestion to propose some additional work that might be done on the measurement of fission cross sections in the

uranium and transuranium regions under a research contract for which I might act as Official Investigator. I suggested that the proposed work might be divided into three groups: (1) the measurement of the slow neutron fission cross section of 93-239; (2) the measurement of the slow neutron fission cross section and the fast neutron fission cross section of 93-237 (on the assumption that we can isolate this as the daughter of 7-day beta-decaying U237); (3) the measurement of the slow neutron fission cross section and the fast neutron fission cross section of U233 (the expected daughter, formed by the beta decay of Pa233 and Th233 which results from the capture of slow neutrons by ordinary stable Th232). I indicated that the first two of these projects could probably be handled without any additional paid help, but the U233 project could best be done with the additional help of a paid chemist Ph.D. assistant. All the projects would need some funds for equipment, materials and supplies, and to pay for the time of use of the 60-inch cyclotron. I suggested a total budget of $21,000. [6, pg.49]

1941-07-16 Measurements by Wahl and English on Gofman's electrolyzed 6 and 10 microcurie Pa233 samples have not been able to establish the growth of any daughter U233 alpha particles, undoubtedly due to the small intensity of the samples and the long half-life of U233. This is consistent with the measurements of April 23 which suggest that the half-life of U233 is 100,000 years or longer. [6, pg.50]

1941-08-22 Gofman returned from his home in Cleveland, Ohio to resume his graduate research program, courses and teaching assistantship at Berkeley. [6, pg.54]

1941-09-02 Dr. Raymond W. Stoughton, a Berkeley Ph.D. of last year, started to work with me today in a postdoctoral capacity supported by the funds from my new contract no. OEMsr-206 which includes a provision of $2,500 to pay for a postdoctoral chemist for one year; he will work in room 311, Gilman Hall, which has been made available to my group. [6, pg.56]

1941-09-03 Stoughton began work today to try to improve the methods for the extraction of Pa233 from neutron bombarded thorium nitrate. He and Gofman plan to establish the best conditions for the carrying of Pa233 by zirconium phosphate and other carriers and will also attempt to perfect the conditions for electroplating the final samples of Pa233 which we plan to isolate in order to allow its growth to daughter U233. The aim is to develop an overall procedure that can be used to isolate as large as possible samples of U233 from large quantities of thorium nitrate subjected to long neutron bombardments at the 60-inch cyclotron. [6, pg.56]

1941-09-09 Stoughton today tested the carrying of Pa233 by MnO_2, formed by the oxidation of Mn++ by ClO_3-, and found good results. This method for coprecipitating protoactinium was developed by A. V. Grosse and B. Agruss (J. An. Chem. Soc. 57, 438 [1935]). [6, pg.57]

1941-09-17 Gofman has resumed his electrolysis experiments in order to develop methods for preparing thin samples of Pa233. This is preparatory to the production of a large sample

of Pa233 through the extensive neutron bombardment of a large quantity of thorium nitrate, which we plan to carry out as soon as the chemical extraction methods have been perfected. [6, pg.59]

1941-09-26 We ordered 25 pounds of thorium nitrate from the Fairmont Chemical Co. for use in our planned neutron bombardments to produce a large sample of U233. [6, pg.61]

1941-10-04 Stoughton has performed numerous experiments during the last month to establish the best conditions for the carrying of Pa233 by zirconium phosphate out of thorium nitrate solutions. [6, pg.61]

1941-10-09 Today Stoughton verified that a precipitate of zirconium phosphate incorporating tracer Pa233 can be dissolved in HF with good recovery of the Pa233 in the solution and negligible loss on a concurrent precipitate of thorium fluoride. [6, pg.62]

1941-10-20 Stoughton is commencing experiments to develop an improved procedure for the removal of Pa233 from large quantities of neutron-bombarded thorium nitrate through the initial use as carrier of manganese dioxide, produced by the reaction of Mn++ with permanganate ions. [6, pg.64]

1941-10-30 Today Gofman and Stoughton began the bombardment of 5 kg of dissolved thorium nitrate with neutrons from the deuteron bombardment of beryllium at the 60-inch cyclotron. The thorium nitrate is surrounded by 3-5 inches of paraffin (except between the sample and the beryllium target). It has been completely purified from any content of natural Pa231 by three separate manganese dioxide precipitations. It is planned to measure the growth of U233 alpha particles from the chemically isolated Pa233 parent, which is the reason for the removal of the natural 30,000-year alpha emitting Pa231 from the thorium nitrate. The isolation procedure also contemplates the removal of natural uranium as well as thorium from the Pa233 so that it may decay to pure daughter U233. [6, pg.65]

1941-11-07 Gofman and Stoughton have developed a method for extracting Pa233 from a large volume of thorium nitrate solution through coprecipitation on manganese dioxide formed by the reaction of manganous chloride with potassium permanganate in hot solutions. This can then be followed by dissolving the manganese dioxide and further concentrating the Pa233 by coprecipitation with zirconium phosphate and dissolving the zirconium phosphate in HF. They developed their procedure for reducing the volume of carrier by repetitions of this cycle and then finally isolating the Pa233 by electrolysis. Thus, they are ready to perform the chemical extraction of the Pa233 from the 5 kg of thorium nitrate whose bombardment with neutrons at the 60-inch cyclotron started on October 30. [6, pg.66]

1941-11-10 The neutron bombardment of 5 kg of thorium nitrate (in saturated aqueous solution) which commenced on October 30 was terminated today after a total of 14,250 microampere-hours of deuterons on beryllium at the 60-inch cyclotron. Gofman and Stoughton immediately started their extraction procedure. [6, pg.66]

1941-11-17 Gofman and Stoughton completed today their extraction of Pa233 from the 5 kg of neutron-bombarded thorium nitrate. This procedure, begun last Monday, started by dissolving thorium nitrate in 26 liters of water which was adjusted to a nitric acid concentration of 0.5 M. To this solution about 400 gm of manganous chloride was added. This was precipitated from the hot solution in three separate portions by the addition of potassium permanganate to the solution. Each precipitate of manganese dioxide was centrifuged out separately. The protactinium is carried down with the manganese dioxide, more or less quantitatively, under these conditions.

The combined manganese dioxide precipitates were dissolved in a mixture of hydrogen peroxide and hydrochloric acid. After decomposing the hydrogen peroxide by boiling, about 200 mg of zirconium oxychloride was added to this solution, the zirconium was precipitated as zirconium phosphate by the addition of phosphoric acid, and the precipitate was centrifuged out. Several further precipitations of zirconium phosphate were performed by adding furthur 100-mg portions of zirconium oxychloride, each precipitate being centrifuged out separately. Zirconium phosphate carries doen the protactinium essentially quantitatively.

It was then necessary to remove the Pa233 from the rather large amount of zirconium with which it was present and to purify it from uranium and thorium. The zirconium and protactinium phosphates were brought into solution by treatment with dilute hydrofluoric acid. This step also gave a further separation from thorium which was precipitated as the insoluble fluoride at this point. This solution was cooled with ice water, and zirconium and protactinium hydroxides were precipitated by adding dilute sodium hydroxide solution; the solution must be kept cold in this precipitation or difficulty will be experienced in redissolving the precipitated hydroxides. The hydroxide precipitate was then dissolved in nitric acid, and the protactinium was carried away from most of the zirconium by another series of manganese dioxide precipitations. Some of the zirconium came along with the manganese dioxide precipitate in this procedure so that a further removal of the zirconium, as well as the manganese, from the protactinium was necessary. This involved going through the above described cycle two more times, finally ending up with a small zirconium phosphate precipitate containing the protactinium.

This zirconium phosphate precipitate, which now contained only about 10 mg of zirconium, was dissolved in hydrofluoric acid and, after precipitation of the hydroxide as described above, was converted to the nitrate by dissolving the hydroxide in nitric acid. Another precipitation of the zirconium phosphate was then made in order to be sure that all the uranium and thorium in amounts less than a microgram were removed. The final zirconium phosphate precipitate was made at 11:59 p.m. [6, pg.69]

1941-11-18 Continuing their work, Gofman and Stoughton converted last night's final zirconium phosphate precipitate (containing all of the Pa233 recovered from their large bombardment) to zirconium sulfate and dissolved this in 0.33 M ammonium fluoride solution preparatory to isolation of the Pa233 by electrolysis onto copper plates. The first electrolysis began at 11:00 am and continued until 8:15 pm. This sample, which is labeled Th+n-I-Pa-I, should contain the majority of the Pa233. They then started their second electrolysis at 8:30 pm and

continued until 9:45 pm, labeling this sample Th+n-I-Pa-II; this should give a small sample convenient for some of the measurements.

They started their third electrolysis at 10:00 pm with the intention of running it overnight.

Measurements of the alpha activity of sample Th+n-I-Pa-II, using the ionization chamber-linear amplifier-magnetic field setup (W-1), commenced at 11:00 pm and will continue over the next several months to observe the growth of daughter U233. It is planned to measure concurrently the alpha counting rate of a standard sample in order to monitor the sensitivity of the apparatus. The measurement showed a counting rate of 22 alphas per minute, even though the sample has just been purified, indicating the presence of a Pa alpha emitter; this is undoubtedly due to the 30,000-year Pa231 produced during the bombardment via the reactions:

$$\mathrm{Th}^{232}(n,2n)\mathrm{Th}^{231} \xrightarrow[\beta]{25\mathrm{hr}} \mathrm{Pa}^{231}$$

[6, pg.70]

1941-11-19 Gofman and Stoughton completed a third electrolysis of Pa233 as the end product from their large neutron bombardment of thorium nitrate at 11:00 am and labeled this sample Th+n-I-Pa-III; this should yield a sample of intermediate intensity. They then started a fourth electrolysis at 12 noon which ran until 9:00 pm; they labeled this sample Th+n-I-Pa-IV. Very rough order of magnitude measurements of the intensities of the beta emission from these Pa233 samples using our ionization chamber FP-54 electrometer setup with absorbers (to help deal with the high intensities of radioactivity) give the following results: for sample Th+n-I-Pa-I, 300 millicuries; for Th+n-I-Pa-II, 6 millicuries; for Th+n-I-Pa-III, 30 millicuries; for Th+n-I-Pa-IV, 1/2 millicurie.

The sample Th+n-I-Pa-I will be allowed to decay until the major portion has been converted into daughter U233, after which the U233 will be separated from the undecayed Pa233. The plan is to use samples Th+n-I-Pa-II and -III to measure the decay and make absorption measurements on the Pa233 radiations using the ionization chamber FP-54 setup, and to determine the half-life of daughter U233 by measuring the growth of its alpha particles using the ionization chamber-linear amplifier-magnetic field setup (W-1). The beta radiation from the Pa233 will be measured with Lauritsen electroscopes and with our ionization chamber connected to a FP-54 electrometer tube, depending upon the intensity of the samples. Our Lauritsen electroscopes have aluminum windows of thickness about 3 mg/cm2 and the FP-54 ionization chamber has an aluminum window of thickness about 0.1 mg/cm2. In order to determine the half-life of daughter U233, these instruments must be calibrated in an absolute manner for the radiations of Pa233 and Th233 which will be done with the help of our Geiger-Mueller counters whose geometrical efficiency is being established. Since the radiation contains a large number of conversion electrons, it will be necessary to determine the percentage of these; this will be done through Geiger-Mueller counter measurements of the beta particles of the 25-minute Th233 and its daughter Pa233. In addition, since the Pa233

samples are mounted on copper, it will also be necessary to correct for backscattering of the beta particles.

The measurement of the beta intensity of samples Th+n-I-Pa-III and -II on the ionization chamber FP-54 electrometer setup will be carried out on step 2 under "standard conditions" by observing the deflection, using or converting the reading to standard resistance and shunt settings R5S5. The plan also is to make observations on the deflections produced by our beta standards RaE-St.-I and UX2-St.-4 at the time of each measurement in order to check the sensitivity.

Measurements of the alpha activity of sample Th+n-I-Pa-III using the ionization chamber-linear amplifier-magnetic field setup (W-1) commenced at 12:30 pm (also to be monitored with concurrent measurements on our standard Pa231 sample). The measurement showed a counting rate of 116 alpha counts per minute, even though the sample has just been purified, indicating the presence of a Pa alpha emitter; as with sample Th+n-I-Pa-II, this is undoubtedly due to the 30,000-year Pa231 produced during the bombardment via the reactions

$$\mathrm{Th}^{232}(n,2n)\mathrm{Th}^{231} \xrightarrow[\beta]{25\mathrm{hr}} \mathrm{Pa}^{231}$$

[6, pg.70-71]

1941-11-25 Gofman conducted absorption measurements in aluminum, usinq the ionization chamber FP-54 outfit, on the Pa233 radiations in sanple Th+n-I-Pa-II. He found an end point fior the beta radiation of 165 mg Al/cm2, corresponding to an energy of 0.5 MeV using the Varder & Eddy relationship. There is evidence of soft radiation (the initial portion of the absorption curve shows a half thickness of 11.5 mg/cm2) suggesting the presence of an appreciable number of conversion electrons; the observed value of 700 for the ratio of beta-particle to gamma-ray ionization is consistent with this conclusion. [6, pg.71-72]

1941-12-04 Using our air-filled ionization chamber FP-54 electrometer setup, Gofman is making beta decay measurements on samples Th+n-I-Pa-III and Th+n-I-Pa-II. Corrected back to the time of last separation from daughter U233 the initial Pa233 beta intensities (for the resistance and shunt setting R5S5 and with the samples on step 2) correspond to a galvanometer reading of 1795 mm for sample Th+n-I-Pa-III and 340 mm for sample Th+n-I-Pa-II. The measurements on sample Th+n-I-Pa-III are being made through 30 mg/cm2 of Al absorber in order to be manageable for so intense a source and the 1795 mm reading is a corrected one that allows for the attenuating effect of the absorber. [6, pg.74]

1941-12-07 The Japanese bombed Pearl Harbor today. I heard the news on my radio while I was listening to the Chicago Bears-Chicago Cards football game in my room at the Faculty Club. The Bears had to win to tie Green Bay for the Division title; they won 34-24. The news of the Japanese attack reached the press box at halftime. There were only 18,000 fans at Comiskey Park in Chicago. [6, pg.75]

1941-12-10 Stoughton began to work on the problem of separating uranium from protoactinium in order to develop a procedure for isolating the daughter U233 from the undecayed Pa233 in sample Th+n-I-Pa-I. [6, pg.75]

1941-12-14 Today Gofman began a new series of experiments in connection with the development of an efficient process for the separation of uranium from protoactinium to be used for the separation of U233 from undecayed Pa233 in sample Th+n-I-Pa-I. [6, pg.77]

1941-12-19 Gofman and Stoughton completed today a bombardment of 5 kg of thorium nitrate (in concentrated aqueous solution) with the neutrons produced from the irradiation of beryllium with 3,000 microampere-hours of deuterons in the 60-inch cyclotron (bombardment no. Th+n-II). They commenced their procedure for the extraction of Pa233 at 8:00 pm. [6, pg.78]

1941-12-22 Gofman took one of the later electrolytically deposited plates of Pa233 (sample Th+n-I-Pa-IV from bombardment Th+n-I which was prepared last month) in order to practice the recovery of daughter U233. By comparison of the intensity of Pa233 beta radiation in this sample with that of sample Th+n-I-Pa-III and the growth of daughter U233 in the latter, we can estimate the number of U233 alphas that should be present in this sample as corresponding to a decay rate of 100 per minute. He scraped the sample from the copper backing plate, dissolved it in acid, removed the dissolved copper by precipitation of CuS, and then removed the undecayed Pa233 by numerous precipitations with zirconium phosphate. The final filtrate from these was then evaporated to a small volume and subjected to electrolysis beginning at 6:15 pm. [6, pg.79]

1941-12-23 Gofman and Stoughton completed the isolation of the Pa233 from bombardment Th+n-II by the chemical procedure started last Friday. They used a procedure like that used on sample Th+n-I-Pa-I. The Pa233 was extracted from the dissolved 5 kg of thorium nitrate by coprecipitation with manganese dioxide, followed by the reduction of carrier by several alternate precipitations of zirconium phosphate and manganese dioxide. The last small zirconium phosphate precipitate containing the Pa233 was dissolved and the Pa233 separated by electrolysis onto copper, leading to a Pa233 sample with beta radiation of about 40 millicuries as determined very roughly by measurement with the ionization chamber FP-54 setup. This Pa233 sample will be divided into a number of fractions and used for the calibration of our ionization chamber FP-54 setup and Lauritsen electroscopes for the absolute measurement of the beta radiation from Pa233.

Gofman completed the electrolysis started yesterday and measured the resultant alpha activity on our alpha ionization chamber G-2. He found an intensity of about 15 alphas per minute and although the yield is lower than expected, this sample is notable in that it represents an actual isolation of the long-sought daughter U233 from its parent Pa233. [6, pg.80]

1942-01-02 A greatly expanded effort is getting underway at many places on the overall project of the preparation of fissionable material for use in a nuclear weapon. This now has a high national priority and a large contract ($405,000 for six months) in the name of

Lawrence is going into effect here at the University of California, Berkeley. This covers all of the work that I am doing, other than a couple of my specific contracts, as well as Lawrence's ambitious program for the separation of U235 by the electromagnetic process, to which Kennedy and Segre are shifting their efforts. Similarly, a large program is getting underway at the University of Chicago under Arthur H. Compton concerned with the production of the isotope 94-239 from the nuclear chain reaction with natural uranium; this will be the center for this research program which has been given the code name Metallurgical Project, with the main component of this, at the University of Chicago, to be known as the Metallurgical Laboratory. The overall organization of Section S-1 has been changed. In Section S-1, still under the chairmanship of Dr. Lyman J. Briggs and remaining in the Office of Scientific Research and Development, the responsibility for the scientific aspect is divided between the Program Chiefs, E. O. Lawrence, A. H. Compton and H. C. Urey, working with Dr. J. B. Conant, Chairman of the National Defense Research Committee. In addition, also with OSRD, there will be a Planning Board of distinguished chemical engineers with Mr. E. V. Murphree of the Standard Oil Development Company of New York as its chief.

I am assuming leave status from my position as Assistant Professor of Chemistry in order to devote full time to this high priority research program. My group is being substantially expanded in order to broaden the scope of my work. My graduate students, Arthur Wahl (who has essentially completed his graduate work), Jack Gofman, Spofford English and John Hamaker, are continuing on their programs: Wahl on the investigation of the chemical properties of elements 93 and 94, the identification of 93-237 and the measurement of the radioactive and fission properties of 94-239 and 93-237 and the radioactive properties of the 50-year 94 and its 2.0-day 93 parent; English is collaborating with Wahl on the measurement of the radioactive and fission properties of the two isotopes of element 94; Gofman is continuing his work on the search for U233 and the measurement of its radioactive and fission properties in collaboration with Dr. Raymond W. Stoughton (who is working as a postdoctorate under my contract OEMsr-206 supporting this work); Hamaker is working to set up the apparatus for eventual ultra microchemical investigations of the chemical properties of 94-239. Morris Perlman is continuing his work on the search for elements 93 and 94 in uranium ores. In addition, Robert B. Duffield, a graduate student working with Professor Melvin Calvin, is shifting to participation in our program in view of its urgency. [6, pg.82-83]

1942-01-08 Gofman dissolved the Pa233 of sample Th+n-I-Pa-IV (prepared on November 19) in acid in order to make a practice run to recover the daughter U233. He removed the dissolved copper by precipitation of copper sulfide, then precipitated a number of zirconium phosphate precipitates to remove the undecayed Pa233, finally added acetic acid and electrolyzed the solution to recover the U233. He calculates from the estimated amount of U233 in the sample at this date that he achieved a recovery of 75%. This is a pretty good test of the procedure to separate uranium from protoactinium which is under development in order to recover our large sample of U233 from sample Th+n-I-Pa-I. The Pa233 content could be estimated by comparison with the growth of U233 alpha activity into sample Th+n-I-Pa-II as of this date

using the ratio of beta intensities of samples Th+n-I-Pa-IV and -II.

This is also notable in that it represents an actual isolation of the daughter U233 from its parent Pa233.

Today Stoughton completed his measurements on the backscattering of Pa233 beta particles from copper using part of the Pa233 sample isolated on December 23 from bombardment Th+n-II. The purpose is to calibrate our ionization chamber FP-54 setup so that we can convert our readings on samples Th+n-I-Pa-II and -III to absolute intensities of beta radiation. His measurements show that a sample of mounted on copper leads to 49% greater deflection on the recording system than does an identical sample mounted on cellophane; this corresponds to a correction factor of 1/1.49 = 0.67. The small effect of backscattering from cellophane can also be corrected for by comparing the Geiger-Mueller counting rate of a sample mounted on cellophane in the upright position but covered by an identical sheet of cellophane with that of the sample in the inverted position. The average of several such determinations of this comparison for Pa233 radiation has shown that the counting rate for the covered sample measured upright is 1.03 times that of the counting rate for the uncovered sample measured inverted. Thus, there is a 3% increase due to backscattering due to cellophane, and the correction factor is 1/1.03 = 0.97. [6, pg.86-87]

1942-01-12 Gofman took a sample of U233 and added a couple of micrograms of natural uranium to test further the separation of uranium from protoactinium. From acid solution he precipitated the dissolved copper as copper sulfide, made a number of precipitations of zirconium phosphate to remove the Pa233, added acetic acid to the remaining solution, and electrolyzed to obtain a yield of 75%. This indicates that there is no great difference in yield between microgram and submicrogram amounts of uranium in their evolving procedure. [6, pg.89]

1942-01-17 Gofman took some of the Pa233 isolated on December 23 from bombardment Th+n-II in order to prepare a U233 stock solution for use in subsequent experiments. He removed the Pa233 very completely by repeated precipitations with zirconium phosphate. This stock solution now contains a concentration of U233 corresponding to about 143 alpha counts per minute (on ionization chamber W-3) per cc, or about 300 alpha disintegrations per minute per cc. [6, pg.90]

1942-01-19 Gofman and Stoughton have completed their experiments for separating uranium from protoactinium and have established a procedure for separating the daughter U233 from undecayed Pa233 in sample no. Th+n-I-Pa-I. The thin film containing the Pa233 and U233 will be dissolved in acid, the small amount of copper dissolved from the backing plate will be removed by H2S precipitation from acid solution. From this solution zirconium phosphate will be precipitated; a large number of zirconium phosphate precipitations will be performed in order to remove more than 99.9% of the undecayed Pa233. The volume of this solution will then be reduced to about 5 cc and after the adjustment of the acidity, including the addition of acetic acid, the electrolysis onto a platinum plate will be carried out. By

this procedure Gofman and Stoughton have demonstrated that more than 90% of the uranium is plated out in a well-adhering film, probably platinum from the electrodes, of entirely negligible weight (about 0.2 mg/cm2).

Today Gofman used the U233 tracer prepared last Saturday to make a careful study of the electrolytic deposition of U. He used what he now thinks is the optimum concentration of acetic acid and the best conditions for electrolysis, and succeeded in obtaining a recovery of about 97%.

He then went on to test his procedure in practice by beginning with a portion of the Pa233-U233 plate on copper from bombardment Th+n-II. He dissolved this in acid, removed the copper by precipitation of copper sulfide, removed the Pa233 by two precipitations of zirconium phosphate, and then set up the conditions for electrolysis. He succeeded in producing a good electrolytic deposition of U233 on platinum with a yield of about 93%. [6, pg.90-91]

1942-01-28 Preparatory toward taking the big step of dissolving sample Th+n-I-Pa-I (prepared on November 18) in order to produce our large sample of U233, Gofman today carefully compared its beta radioactivity with that of Th+n-I-Pa-III which has been carefully measured with our calibrated ionization chamber FP-54 setup. In order to measure samples of such a high intensity, he used our thick-windowed ionization chamber filled with freon (window thickness 130 mg Al/cm2) and filtered the radiation through 225 mg Al/cm2. He measured a deflection of 216 mm compared to 29 mm for sample Th+n-I-Pa-III, indicating that the former has 7.5 times as much Pa233 (plus U233) as the latter.

Gofman then took the plunge. Under the assumption that he and Stoughton have now developed an adequate procedure for the separation of the daughter U233 from the undecayed Pa233, he scraped the material of sample Th+n-I-Pa-I off the copper plate, then washed the plate with 12 M HCl until it showed less than 0.1% of the original gamma activity. He added a few drops of HNO_3, then evaporated to a small volume. [6, pg.96]

1942-01-29 Proceeding with his work started yesterday, Gofman continued evaporation of his volume until 2 cc were left, passed in H2S to precipitate copper sulfide. The filtrate was then made 8 M in HCl of total volume 10 cc, zirconium oxychloride added and the zirconium precipitated as zirconium phosphate by the addition of phosphoric acid. He then precipitated a second zirconium phosphate to further remove the Pa233 after which he reduced the volume by evaporation to 2 cc and went home for the night. [6, pg.97]

1942-01-30 Gofman spent the entire day carefully carrying out eight successive zirconium phosphate precipitations, to continue to remove Pa233 (in continuance of his work of yesterday) from the U233 in solution. [6, pg.97]

1942-01-31 Working all day in continuance of his isolation of U233 from sample Th+n-I-Pa-I, Gofman removed successively eight more zirconium phosphate precipitates in his effort to eliminate undecayed Pa233 to as great an extent as possible.

This afternoon Stoughton made another bombardment (no. Th+n-IV) of thorium with neutrons with the aim of measuring the Th233 -> Pa233 decay to determine the conversion

electrons of Pa233. The thorium nitrate was cleansed of natural decay products through the precipitation of lead and bismuth sulfide and lead and barium sulfate immediately before the bombardment. He bombarded one gram of thorium nitrate with neutrons produced by the irradiation of a beryllium target with 50 microamperes of 16 MeV deuterons from 5:00 to 5:55 pm. He has modified the chemistry in order to produce a cleaner sample of Th233. His procedure in this case was to first precipitate the dissolved thorium as the iodate, then dissolve this in concentrated HCl with the addition of a number of fission product carriers. From this solution zirconium phosphate was precipitated twice and from the filtrate the thorium was precipitated as the fluoride at 6:43 pm. A portion of this was then mounted on cellophane and the radiations of Th233 measured with electroscope no. 9 with the sample placed on step 2, the measurements beginning at 7:20 pm.

He used another portion of the sample to determine the efficiency of Lauritsen electroscope no. 9 for the beta radiation of Th233 by making measurements with the electroscope and effecting his absolute calibration through the use of Geiger-Mueller counter no. Z-32 (counting efficiency, 29%). He finds a sensitivity for Th233 beta radiation of 2.03 divisions per second per microcurie on step 2 and, from the known relationship of step 1 to step 2, a sensitivity of 5.3 divisions per second per microcurie for step 1.

By 8:30 pm the main sample had decayed sufficiently so that he could continue the measurements on Geiger-Mueller counter no. Z-32. He continued to follow the decay until a little past midnight. The plan is to continue to follow the decay on Geiger-Mueller counter no. Z-32 until all of the Th231 has decayed and the intensity of the Pa233 daughter of Th233 can be established. [6, pg.98-99]

1942-02-01 Gofman took out his 17th zirconium phosphate precipitate, then evaporated the remaining solution, containing the daughter U233, to a volume of approximately 0.15 cc. He then added 5 cc of water, 0.02 cc of glacial acetic acid, warmed the solution to insure that everything was dissolved, poured it into his electrolytic cell, and began electrolysis onto a platinum plate at 4:15 pm.

Today I sat down and wrote a summary of the status of the work of my group, for use in a report to be sent by Lawrence to J. B. Conant, Chairman NDRC, as follows:

"Chemical procedures are being developed for the isolation of U233, which is the end product formed after a chain beta-decay when thorium is bombarded with slow neutrons. About 12 pounds of thorium nitrate have already been subjected to an intense neutron bombardment, and it is planned soon to isolate several micrograms of U233 to be used for the measurement of its neutron fission cross sections and spontaneous fission rate." [6, pg.99]

1942-02-02 Working past midnight, Gofman terminated his electrolysis at 1:15 am this morning, removed the sample and gave it the formal sample no. Th+n-I-Pa-I-U-I, which we decided to name more colloquially Sample J. This avoids the use of the designation Sample I, the next letter in the line of designation of our major samples, to eliminate confusion with the Roman numeral I.

Gofman is beginning to use room 234 in the Old Chemistry Building, which has been assigned to us to help us meet our expanding space requirements; this has served until this semester as a laboratory for the undergraduate course in quantitative chemical analysis (Chemistry 5). He will use this room for the measurement of the slow and fast neutron fission cross sections of U233 with neutrons from our 300 mg Ra-Be source, as well as the spontaneous fission rate of U233.

After going home for some sleep Gofman came back this evening to make the initial test on whether U233 undergoes fission with slow neutrons using Sample J. He used our 300 mg Ra-Be neutron source and placed Sample J on one electrode of an ionization chamber connected to a linear amplifier adjusted to discriminate against the high alpha particle emission rate in passing spontaneous fission signals to the recording system. Our Uranium Standard No. 6 (U-St-6), containing 200 micrograms of natural uranium and hence 1.4 micrograms of U235, will be used as a comparison standard. Measurements on Sample J were made with and without a cadmium shield in order to determine the effect of slow neutrons.

Beginning his counting at 9:44 pm, Gofman found 39 counts per hour which could be attributed to the slow neutron fission of U233 in our Sample J. **This, then, represents the first demonstration of the slow neutron fissionability of U233**, but quantitative results will await the determination of the weight of the U233 by measurements of its alpha counting rate and the use of the half-life of U233 as determined from our U233 alpha growth experiments from the decay of Pa233 in samples Th+n-I-Pa-II and -III. [6, pg.100]

1942-02-03 Continuing his work of yesterday, Gofman determined in the early morning hours that the slow neutron fission counting rate of U-St-6 is about 17 counts per hour. This, then, gives us a very preliminary measure of the slow neutron fission ratio

$$\frac{\text{Sample J}}{\text{U-St-6}} = \frac{39}{17} = 2.3$$

Our determination of the relative cross sections of U233 and U235 will await determination of the weight of U233 in Sample J.

It is clear that the use of our 300 mg Ra-Be source makes the determination of the slow neutron fissionability of U233 a much simpler task than our determination of the slow neutron fissionability of 94-239 last May using neutrons from the 37-inch cyclotron.

After going home for some sleep, Gofman returned this afternoon to continue his measurement of the comparative slow neutron fission in Sample J and U-St-6 (containing 1.4 microgram of U235), using our 300 mg Ra-Be source. He found a ratio for slow neutron fission of

$$\frac{\text{Sample J}}{\text{U-St-6}} = \frac{48}{12} = 4$$

Gofman then decided to make a preliminary look for spontaneous fission in Sample J starting the counting at 11:08 pm.

Using a number of Pa233 samples (from bombardment Th+n-II of last December) yesterday and today Stoughton calibrated our Lauritsen electroscopes no. 5 and no. 9 for the detection of the beta radiation from Pa233. He counted the samples on our Geiger-Mueller counter no. Z-32 (of known counting efficiency, 29%) and on the electroscopes. Correcting for the absorption of the radiation in the thin windows of the counter and electroscopes (using the known half thickness of 11.5 mg/cm2), he finds for electroscope no. 5 a sensitivity of 6.25 divisions per second per microcurie when the sample is placed on step 1, 2.53 divisions per second per microcurie (step 2); and for electroscope no. 9, a sensitivity of 7.85 divisions per second per microcurie (step 1), and 3.00 divisions per second per microcurie (step 2). (The unit "microcurie" here refers to the total radiation consisting of conversion electrons as well as nuclear beta particles.)

Late this evening Stoughton made an hour's bombardment of 1 gm of thorium nitrate with neutrons at the 60-inch cyclotron (bombardment no. Th+n-V), then isolated chemically a purified thorium fraction using the same procedure he used on Saturday; he will use this fraction to make another determination of the efficiency of electroscope no. 9 for Th233 beta radiation using Geiger-Mueller counter no. Z-32 for absolute counting. [6, pg.101]

1942-02-04 Continuing his work on yesterday's bombardment (Th+n-V) early this morning, Stoughton made measurements on the Th233 beta radiation in his thorium fraction with Lauritsen electroscope no. 9 and Geiger-Mueller counter no. Z-32 and found a value for the sensitivity of electroscope no. 9 for the beta radiation of Th233 in good agreement with the value he found in his similar experiment of January 31.

Returning this morning, Gofman found that at 11:25 am Sample J has not produced any spontaneous fission counts during the interval of 12 hours and 17 minutes since last night. He changed the "A" battery of the amplifier and then started another spontaneous fission count experiment at 8:40 pm.

Stoughton completed his measurements on the Pa233 formed as the daughter of Th233 in the continuance of the experiment that began on January 21 (bombardment no. Th+n-III) to study the Th233 -> Pa233 decay sequence. Using his calibration of Lauritsen electroscope no. 9, he finds the disintegration rate of the parent Th233 to be 205,000 per minute when extrapolated back to the time of its separation from daughter Pa233. The disintegration rate of the Pa233 extrapolated back to the time of the beginning of its decay (which requires a correction for the contribution of the 24.5-hour Th231) is 180 per minute. Using a correction factor of 1.26 for the absorption of the soft Pa233 electrons (absorption half thickness of 11.5 mg/cm2) in the window (3.9 mg mica/cm2) of our Geiger-Mueller counter no. Z-32, we can calculate the ratio of daughter Pa233 electrons to parent Th233 electrons as

$$\frac{180 \times 27 \times 24 \times 60 \times 1.26}{205000 \times 25.5} = 1.69$$

using 25.5 minutes as the half-life of Th233 and 27 days as the half-life of Pa233. Analysis of the Th233 -> Th231 -> Pa233 decay complex indicates a half-life of 25.5 minutes for

Th233. [6, pg.102]

1942-02-05 Gofman returned and found no spontaneous fission from Sample J at 12:35 pm, representing an interval of 15 hours and 55 minutes. [6, pg.103]

1942-02-06 Continuing his measurements on the slow neutron fission of the U233 in Sample J with the 300 mg Ra-Be neutron source, Gofman found a counting rate of 40 fissions per hour with Sample J and 14 fissions per hour for comparison sample U-St.-6, corresponding to a ratio between the two of 2.9. [6, pg.105]

1942-02-07 As the result of a telegram from Arthur Compton, I met in Ryerson Hall at the University of Chicago yesterday and today with a group to discuss the general problem of the production of 94-239 using the chain reaction in natural uranium and its subsequent separation from the uranium and fission products. Present during the meetings at various times were Arthur H. Compton, Enrico Fermi, Franklin A. Long, Norman Hilberry, Gregory Breit, John A. Wheeler, Herbert N. McCoy, Herman I. Schlesinger, Morris S. Kharasch, Herbert C. Brown, R. S. Archer, and B. L. Benbow.

In the course of the meetings I gave rather complete reports on our work at Berkeley on the identification of 94-239, including the measurements of its radioactive properties and slow and fast neutron fission cross sections, our work on the radioactive properties of 93-239, U237 , the 50-year 94, its parent 2.0-day 93, and our predictions for 93-237 and U233 which I indicated we are in the process of identifying.

I described our work on the chemical properties of 94 and our procedures for isolating 94 and 93 from large amounts of UNH and fission products using our oxidation-reduction procedure with rare earth fluoride as carrier.

As a result of this meeting I now fully realize the magnitude of the project being planned around our new element 94 and the enormity of the chemical separation problem of isolating 94-239 from large amounts of uranium and almost fantastic intensities of fission products. When Compton asked me if I thought I could devise very soon a chemical process for separating 94 from such uranium and fission products—a process that could be successfully scaled up for actual use in a chemical extraction plant—I indicated that I thought I could, but I must confess to some misgivings.

One of the questions that we discussed was the matter of who might head up this chemical extraction work on 94 at the Metallurgical Laboratory here in Chicago. Compton is trying to convince Frank Long to undertake this responsibility, but Frank is doubtful that he should leave the other war work he is doing, which he regards as very important. [6, pg.104]

1942-02-10 Stoughton completed today his measurements on the intensity of electron radiation from Pa233, which is the follow-up of the experiment that began on January 31 (bombardment no. Th+n-IV) to study the Th233 -> Pa233 decay sequence. He finds the disintegration rate of Pa233 extrapolated back to the time of the beginning of its decay to be 227 per minute. The intensity of the parent Th233 beta radiation extrapolated back to the time of separation

from daughter Pa233 is 270,000 per minute, based on the calibration of electroscope no. 9. Hence, when a correction factor of 1.26 is applied for the absorption of beta particles in the Geiger-Mueller counter no. Z-32 window, we can calculate the ratio of daughter Pa233 electrons to parent Th233 electrons as

$$\frac{227 \times 27 \times 24 \times 60 \times 1.26}{270000 \times 25.5} = 1.62$$

using 25.5 minutes as the half-life of Th233 and 27 days as the half-life of Pa233. [6, pg.106]

1942-02-11 From continued measurements on the slow neutron fission of the U233 in Sample J with the 300 mg Ra-Be neutron source, Gofman has found a counting rate of 45 fissions per hour with Sample J and 14 fissions per hour for comparison sample U-St-6, corresponding to a ratio between the two of 3.2. [6, pg.106]

1942-02-16 This evening Stoughton bombarded 1.6 gm of thorium nitrate, from which natural decay products had been previously removed, with neutrons from the 60-inch cyclotron from 8:15 pm to 10:00 pm (bombardment no. Th+n-VI). He immediately isolated a pure thorium fraction using a procedure essentially like that of January 31, and began measurements of the Th233 decay using electroscope no. 9 at 10:29 pm. He changed over to Geiger-Mueller counter no. Z-32 at 11:25 pm. [6, pg.108]

1942-02-18 Intermittently since his first measurements on the slow neutron fission rate of U233 started on February 2, Gofman has continued his measurements. He finds that the ratio of induced fissions, using the slow neutrons from the 300 mg Ra-Be neutron source, with the neutron source and the ionization chamber surrounded with paraffin, on the basis of measurements made without and with the cadmium shield, corresponds to a rate for Sample J about three times that of the standard uranium sample (U-St-6). [6, pg.111]

1942-02-19 Yesterday and today Gofman has made a preliminary measurement of the range of the alpha particles of U233 (Sample J) using our low geometry ionization chamber-linear amplifier setup no. W-2. Making comparative measurements of the absorption in aluminum foil of U233 and Po210 (sample Po-St-6) alpha articles, he finds the range of Po210 alphas to be longer than those of U233 by 1.34 mg/cm2 of Al. Converting to range in air (conversion factor of 1.52) this difference corresponds to 0.88 cm of air, indicating the range of U233 is 3.92 (the air range for Po210) minus 0.88 or 3.04 cm of air at standard conditions (760 mm and 15 deg C). This should be considered to be a preliminary, rather rough, value.

Today Gofman is beginning his longer series of measurements to determine the spontaneous fission rate of U233 using Sample J with his apparatus set up in room 234 in Old Chemistry Building. [6, pg.112]

1942-02-23 Stoughton bombarded a 1 gm sample of thorium nitrate, previously purified from its natural disintegration products, with neutrons at the 60-inch cyclotron from 11:00 am to 12 noon (bombardment no. Th+n-VII). He then isolated a purified thorium fraction by a

procedure essentially the same as that used on January 31 and began measurements on the decay of Th233 using electroscope no. 9 at 12:35 pm. He divided the sample into four portions, which he labeled samples no. 1, 2, 3 and 4. He then followed the decay of all four samples on electroscope no. 9. He shifted samples no. 3 and 4 to Geiger-Mueller counter no. Z-32 at 2:23 pm. The plan is to follow the decay of these two samples until the intensity of the the electron radiation from daughter Pa233 can be established in each case. He also intends to measure the absorption in Al of the low energy part of the Pa233 beta spectrum in order to obtain better information to correct for the absorption of the Pa233 beta particles in the Geiger- Mueller counter's mica window (approximately 3 mg mica/cm2 thickness). The purpose is to improve the calibration of our Lauritsen electroscope no. 9 (and hence our ionization chamber FP-54 setup) for absolute measurement of the intensities of Pa233 beta radiation, information that is needed in connection with our experiment to determine the half-life of U233. [6, pg.113]

1942-02-24 Stoughton completed his measurements on the decay of daughter Pa233 produced in the decay of parent Th233 isolated from bombardment no. Th+n-VI on February 16, 1942. Using his absolute calibrations of electroscope no. 9 for Th233 beta radiation and Geiger-Mueller counter Z-32 results for daughter Pa233 beta radiation, and correcting for absorption in the Geiger-Mueller counter window, he finds a value of 1.65 for the ratio of Pa233 electrons to Th233 electrons. [6, pg.113]

1942-02-25 Gofman's attempts to measure the spontaneous fission rates of U233 commenced on February 19 with the apparatus set up in room 234, Old Chemistry Building, has been plagued by spurious counts due to electrical interference. He is therefore moving his ionization chamber, amplifier and recorder for the spontaneous fission work to his home (apartment number 106, 1890 Arch Street, Berkeley). [6, pg.113]

1942-03-02 Stoughton bombarded 1 gm of thorium nitrate, previously purified of its natural decay products, with the neutrons from the 60-inch cyclotron from 7:10 pm to 8:15 pm (bombardment no. Th+n-VIII). After the bombardment he purified the thorium fraction by the usual procedure, finishing his separation procedure at 8:53 pm. The purpose of this bombardment is to determine the shape of the absorption curve and the energy of the Th233 beta particles by absorption with aluminum using electroscope no. 9 with the sample on the second step. He completed the absorption measurements by 10:23 pm with the result that he found an end point of about 700 mg/cm2, corresponding to an upper energy limit of about 1.6 MeV. The initial part of the absorption curve shows a half thickness of 60 mg per cm2 of Al. The ratio of the ionization due to the beta particles to that due to the gamma rays is approximately 1,000, indicating that there are essentially no nuclear gamma rays in the Th233 radiation; the small amount of apparent electromagnetic radiation is probably to be attributed to bremsstrahlung. This apparently lends weight to the assumption that there are no conversion electrons present in the radiation of Th233. The shape of the absorption curve also indicates the absence of conversion electrons. [6, pg.115-116]

1942-03-05 Today Stoughton completed another calibration of our ionization chamber FP-54 electrometer setup for the beta radiation of Pa233. Using a sample of Pa233 isolated from bombardment Th+n-II on December 23, he made measurements using our calibrated electroscope no. 9 and the FP-54 ionization chamber (step 2). He finds a sensitivity of 0.058 mm of galvanometer deflection per microcurie of Pa233 total electron radiation (conversion electrons plus nuclear beta particles) at our standard resistance and shunt setting R5S5. This is in good agreement with the value of 0.061 mm which he found on January 9. [6, pg.117]

1942-03-09 Stoughton completed the measurements on sample nos. 3 and 4 containing daughter Pa233 resulting from the decay of Th233 isolated on February 23 from bombardment no. Th+n-VII. He calculates that the ratio of electrons from Pa233 to those of Th233 is 1.82 for sample no. 3, and 1.73 for sample no. 4. These are probably the best among the five determinations that have been made of this ratio.

He also measured the absorption of the Pa233 beta radiation in 3 mg/cm2 aluminum using Geiger-Mueller counter no. Z-32 in order to determine the equivalent absorption of this radiation in the window of the counter (approximately 3 mg/cm2 mica thickness); he found that the ratio of the counting rate with 3 mg/cm2 Al to that without is 1.3, indicating that this is the correction factor to allow for the absorption of Pa233 beta radiation in the window of Geiger-Mueller counter no. Z-32. [6, pg.118]

1942-03-10 Stoughton calculated a weighted average of the ratio of Pa233 electrons to Th233 electrons determined in experiments from bombardments Th+n-III, -IV, -VI and -VII as 1.75. In the course of these determinations he has concluded that the best value of the half-life for Th233 is 23.5 minutes, rather than the 25.5 minutes that we have been using. Recalculated on this basis, the ratio becomes 1.66. Also from his value of the absorption half thickness (60 mg per cm2 in Al), he has calculated the small correction for the absorption of the Th233 beta radiation in the Al window of Geiger-Mueller counter no. Z-32 (a factor of 0.96), leading to a final corrected value of 1.59 for the ratio. [6, pg.119]

1942-03-16 The beta decay of sample Th+n-I-Pa-III as followed on our FP-54 ionization chamber indicates a half-life of 27.4 days for Pa233. The growth of U233 alpha particles in this sample as measured with the ionization chamber-linear amplifier-magnetic field outfit (W-1) is consistent with a half-life of 27.4 days for the parent beta emitting Pa233 and extrapolates to a total at infinite time of 428 alpha counts per minute after the constant contribution of Pa231 alpha particles is subtracted. From this disintegration rate of daughter U233 alpha particles and the disintegration rate of parent Pa233 beta particles measured on December 4, 1941, corrected to zero time, it is now possible to calculate the half-life of U233. Correcting for the geometry factor of the ionization chamber-linear amplifier-magnetic field setup (W-1), which has been determined to be 0.019 (by counting a small sample of U233 both in this ionization chamber and our calibrated "inside" ionization chamber, W-3), we can calculate that the total U233 disintegration rate is 375 alpha particles per second. The Pa233 the galvanometer deflection measured with our ionization chamber FP-54 electrometer on December 4, 1941, corrected for absorption and decay, is 1795 mm (standard conditions of

R5S5) corresponding to the time of last separation of Pa233 from daughter U233 (11:59 pm, November 17, 1941). Using this value, the calibration for radiation of March 5, 1942 (0.058 mm of galvanometer deflection per microcurie of Pa233), and correcting for the backscattering of Pa233 beta particles from copper relative to cellophane (factor of 0.67) and from cellophane (factor of 0.97), for the absorption of Pa233 radiation in the mica window of the Geiger-Mueller counter used in the calibration of the FP-54 ionization chamber (factor of 1.3), and for the conversion electrons in the radiation (factor of 1.6), we have

$$\frac{1795 \times 0.67 \times 0.97 \times 1.3}{0.058 \times 1.6} \cong 16300 \text{ microcuries}$$

or 16.3 millicuries in sample no Th+n-I-Pa-III. Since one microgram of 27.4-day Pa233 corresponds to 20.5 millicuries, we can calculate the weight of U233 in sample Th+n-I-Pa-III after all of the parent Pa233 has decayed as

$$\frac{16.3}{20.5} = 0.80 \text{ micrograms}$$

From these data we can now calculate the alpha half-life of U233 as

$$\frac{27.4 \times 16.3 \times 3.7 \times 10^7}{365 \times 375} = 120,000 \text{ years}$$

The beta decay of sample Th+n-I-Pa-II as followed on our FP-54 ionization chamber also indicates a half-life for Pa233 of 27.4 days. The growth of U233 alpha particles in this sample as measured with the ionization chamber-linear amplifier-magnetic field outfit (W-1) also corresponds to a half-life of 27.4 days for the parent beta-emitting Pa233; the alpha intensity extrapolates to about 75 per second at infinite time. Considerations similar to those for sample Th+n-I-Pa-III indicate an initial intensity of Pa233 beta particles of 3.0 millicuries which, together with the intensity of alphas from daughter U233 (75 per second), also leads to an alpha half-life of close to 120,000 years for U233. The beta decay of Pa233 samples isolated on December 23, 1941 from bombardment Th+n-II, measured with our electroscope no. 9, also show a half-life of 27.4 days; the error corresponding to an average of all these determinations is $\pm$0.2 days—*i.e.*, the half-life of is 27.4$\pm$0.2 days. [6, pg.121-123]

1942-03-18 This evening I completed my dictation to Helen of the report "Chemical Properties of Elements 94 and 93." Besides the discussion of the tracer chemical properties, a section on yields of 93-235 or 238 (and thus of 94-235 or 238) from deuteron bombardments and of 93-239 (and thus of 94-239) from neutron and deuteron bombardments is included. **This paper, under the authorship of Wahl and me, also includes for the first time the suggestion that element 94 be given the name plutonium (symbol Pu) after Pluto, the second planet beyond Uranus, in line with McMillan's suggestion that element 93 be given the name neptunium (symbol Np) after Neptune, the first planet beyond Uranus, from which uranium derives its name.** We also point out that there is practically no resemblance in

chemical properties between neptunium and plutonium on the one hand and rhenium and osmium on the other. We indicate that the chemical properties of neptunium and plutonium are those of a rare-earth-type group of elements starting at the upper end of the Periodic Table and go on to suggest that the group can be just as well considered to be starting with actinium or thorium as with uranium, which has been previously suggested as the starting point. [6, pg.124]

1942-03-24 Now that the weight of U233 in sample no. Th+n-I-Pa-III (corrected for incomplete decay of Pa233) is known, it is possible to determine the weight of U233 in Sample J by determining the ratio of the alpha activity in the two samples. Using the ionization chamber-linear amplifier-magnetic field setup (W-1) and a screen to cut down the intensity of alphas in order to avoid coincidence corrections, Gofman and Stoughton measured the alpha count of Sample J as 640 per minute and of sample Th+n-I-Pa-III (corrected to infinite time) as 131 per minute (after subtracting the contribution of Pa231). Thus, Sample J contains 640/131 = 4.9 as much U233 as does Th+n-I-Pa-III. The weight of U233 in Sample J is therefore (4.9)(0.8), or actually (4.9)(16)/20.5 = 3.8 micrograms. [6, pg.128-129]

1942-03-30 Today Gofman completed his measurements on the spontaneous fission of U233 using Sample J with the apparatus set up in his apartment on February 25.

After overcoming a number of difficulties with the stability of his amplifier circuits, he has managed to conduct 243 hours of meaningful operation during which he has observed no spontaneous fission counts with the 3.8 micrograms of U233 in Sample J. In order to determine the efficiency of his ionization chamber and amplifier outfit for detecting spontaneous fission events, he placed a sample of natural uranium on the electrode of his ionization chamber and then measured the variation of the neutron-induced fission rate with amplifier gain. Also taking into account the geometry of the ionization chamber, he finds that the overall efficiency of the apparatus for recording spontaneous fissions amounts to about 72%. Hence, his 243 hours of counting corresponds to 243 x 0.72 = 175 hours of effective counting during which no spontaneous fission events were recorded. Since the expected average interval between fission counts for a half-life of 10^{14} years corresponds to 135 hours, it seems probable that the half-life for the spontaneous fission of U233 is of the order of, or greater than, 10^{14} (100 trillion) years. [6, pg.130]

1942-03-31 I enclosed in my letter to Compton a summary entitled "Additional Investigations Underway by My Group at Berkeley" and a summary of "Investigations Which Might Be Considered in Case A Very Large Chemical Program Were Undertaken" as follows:

ADDITIONAL INVESTIGATIONS UNDERWAY BY MY GROUP AT BERKELEY

"1. Investigation of radioactive and fission properties of U233 (nearing completion)."

INVESTIGATIONS WHICH MIGHT BE CONSIDERED IN CASE A VERY LARGE CHEMICAL PROGRAM WERE UNDERTAKEN

"2. The development of chemical methods for separating kilograms of U233 from large amounts of thorium and fission products in case U233 should prove to be a useful isotope."

"7. A search for U232 formed in the deuteron bombardment of uranium from the reactions Th232(d,2n)Pa232 and Pa232 -> U232, in the hope that U232 might be a better tracer for microgram amounts of uranium than are U233 or U237." [6, pg.133]

1942-04-01 Yesterday and today English and Wahl attempted to develop a fluoride volatility method for separating 94 from uranium and fission products. They prepared a solution of uranyl nitrate containing 94 and precipitated the uranium and 94 from the solution with hydrogen fluoride to obtain uranous tetrafluoride incorporating 94 fluoride or 94 oxyfluoride. Fluorine was then passed over the mixture of uranium and 94 in their lower fluoride states in an attempt to produce the higher fluorides and for the purpose of determining whether the uranium hexafluoride or the 94 higher fluoride is more volatile. Unfortunately, the experiment was a failure because uranium hexafluoride was not produced—perhaps because of an insufficient supply of fluorine or because the apparatus was not dry. [6, pg.136]

1942-04-02 During the last several days Gofman has continued his measurements on the slow neutron fission of U233 in Sample J and has now accumulated, including his experiments in February, a total of 453 fission counts in 8.2 hours of counting time, corresponding to 55.2 counts per hour. When the ionization chamber was surrounded with the cadmium shield (0.35 gm/cm2 thickness), 161 counts were obtained in 17.6 hours of counting time, corresponding to 9.1 counts per hour. Therefore, the U233 fission counting rate due to slow (cadmium-absorbable) neutrons amounts to about 46.1 counts per hour. When Sample J was replaced by standard sample U-St-6 (200 micrograms of natural uranium containing 1.4 micrograms of U235), 330 fissions were obtained in 15.5 hours of counting, amounting to 21.3 counts per hour under the same conditions. When the ionization chamber was surrounded with the cadmium shield, 168 fission counts were obtained in 20.7 hours of counting time, which amounts to 8.1 counts per hour. Thus, the slow neutron fission counting rate of the U235 is 13.2 counts per hour. The ratio of the slow neutron fission counting rate of Sample J to standard sample U-St-6 is therefore 46.1/13.2 = 3.5 from which the slow neutron fission cross section of U233 relative to that of U235 can be calculated, using the weight of U233 in Sample J (3.8 micrograms), as (3.5)(1.4)/3.8 = 1.3. [6, pg.137]

1942-04-05 Yesterday and today Gofman made measurements of the slow neutron fission rate due to the 0.8 microgram of U233 in sample Th+n-I-Pa-III. Again, he used the 300 mg Ra-Be source and placed the sample on one electrode of an ionization chamber imbedded in paraffin and made measurements with and without a cadmium shield. His counting rate of 11.2 counts per hour (21.8 hours of counting time) without the cadmium shield and 1.9 counts per hour (11.5 hours of counting time) with the cadmium shield indicates 9.3 counts per hour due to slow neutron fission. This counting rate of about one-fifth of that obtained with Sample J, with this sample which has about one-fifth as much U233, again leads to the result that the slow neutron fission cross section of U233 is about 1.3 times that of U235. [6, pg.139]

1942-04-07 Today Gofman measured the range of the alpha particles from U233 by comparison of the absorption of these in aluminum foils with the absorption of Po210 alpha particles

(sample Po-St-6) measured under the same conditions. He used our low geometry ionization chamber-linear amplifier-magnetic field setup (W-1), with the magnetic field turned off. He finds the range of Po210 to be longer than that of U233 by 1.1 mg per cm2 of Al. Differences in range between the alpha particles can be converted to differences in air range at 760 mm and 15 deg C using the Livingston and Bethe (Livingston and Bethe, Rev. Mod. Phys. 9, 276 [1937]) conversion factor of 1.52 mg per cm2 Al per cm of air. Thus the range in air of U233 alpha particles is 0.72 cm less than that of Po210 (whose range is 3.92 cm), or 3.2 cm of air at standard conditions (760 mm Hg and 15 deg C. Taking into account his results of February 18-19 (a range of 3.0 cm), the best value for the range is 3.1 ± 0.2 cm, corresponding to an energy of 4.8 MeV. These data for U233 are the first obtained for an alpha emitter of the missing (4n+1) radioactive series. It is interesting to note that this point falls near the Geiger-Nuttal curve for the uranium (4n+2) series. [6, pg.140]

1942-04-08 Working in room 234, Old Chemistry Building, Gofman is beginning his measurements on the fast neutron fission cross section of U233 using our new 1 gm radium-beryllium source acquired from the Canadian Radium and Uranium Company in New York. We now have available a sample prepared by Kennedy from the output of the uranium isotope separation using the 37-inch cyclotron magnet. This sample contains 1.3 micrograms of U235 and 5.2 micrograms of U238 (and 0.0014 microgram of U234); thus, it is possible by also using a sample of natural uranium to make comparative measurements of the fast neutron cross section of U233 in comparison with those of U235, U238, as well as 94-239 (using our Sample B) and Pa231 (being isolated by Stoughton). Gofman has built a fission-detecting ionization chamber, by the use of cadmium plate and boron carbide, with an optimal arrangement for measuring the cross sections due to fast neutrons. [6, pg.141]

1942-04-09 Gofman has run into trouble with his experiments on measuring fast neutron cross sections using our new 1 gm Ra-Be neutron source because this source has sprung a leak resulting in radon contamination of the ionization chamber. [6, pg.142]

1942-04-12 I wrote an abstract of our report "Properties of U233" authored by Gofman, Stoughton and me. The exigencies of my schedule in getting ready for my move to Chicago prevents me from preparing the whole report at this time and so I will get to this soon after my arrival at the Metallurgical Laboratory. We report in the abstract that we have prepared by neutron bombardment of Th232 a sample of U233 weighing 3.8 micrograms and with this have demonstrated that this isotope undergoes fission with slow neutrons with a cross section about 1.25 times that of U235, has a spontaneous fission "half-life" greater than 10^{14} years, and decays by the emission of alpha particles of range 3.1 ± 0.2 cm, with a half-life of 120,000 years. [6, pg.144]

1942-04-13 Today there was a briefing by members from the Army Intelligence Division concerning the importance of secrecy on our project. It took place at 2:00 pm in room 213 LeConte Hall and all members of my group, including myself, and all members of the project were required to attend. [6, pg.144]

1942-04-14 I wrote to Professor Arthur H. Compton enclosing copies of our reports "Properties of U233" (an abstract) by Gofman and me, "Properties Of 93-237 by Wahl and me (both dated April 13, 1942), and "Production and Properties of 50-Year Element 94" dated March 20, 1942. I also indicated that Dr. Alexander Langsdorf has bombarded uranium with deuterons for us at St. Louis so that we can compare the production of 50-year 94 with that produced in Berkeley; we have found that the yield at St. Louis is of the order of 1/10 as much as that at Berkeley.

I also wrote to Dr. Briggs in washington enclosing these three reports and indicating that this represents the work done under my contract OEMsr-206. I also informed Dr. Briggs that I am transferring a portion of my men and work to Chicago.

Later Note: The paper "Properties of U233" (abstract) was issued immediately by the S-1 Committee in Washington as report number A-153 dated April 13, 1942 with authors G. T. Seaborg, J. W. Gofman and R. W. Stoughton. After the war it was published under the changed title "Nuclear Propertics of U233: A New Fissionable Isotope of Uranium" by G. T. Seaborq, J. W. Gofman and R. W. Stoughton in Phys. Rev. 71, 378 (1947). [6, pg.146]

1942-04-17 The leak of the 1 gm Ra-Be neutron source has been temporarily fixed and Gofman and Friedlander are continuing with the measurement of the comparative fast neutron fission cross sections of U233, U235, U238, 94-239, and now Pa231.

Today is the day that I am scheduled to leave with Isadore Perlman to take up my position at the Metallurgical Laboratory at the University of Chicago.

Perlman will accompany me so as to be at the Met Lab from the very beginning, and English will join us there within a week or two. With that start we will try to build up our group by hiring new people.

The remainder of my group will stay at Berkeley to carry on the research program and I hope to interest Dean Wendell M. Latimer to take an interest in, and serve as a leader for, this continued program.

Gofman, working with Friedlander, will finish the measurements on the relative fast neutron fission cross sections of U233, U235, U238, 94-239 and Pa231. He will also work on other aspects of induced radioactivity in thorium, such as the products of deuteron bombardment. He will, however, put an increasing amount of his time on the development of chemical procedures for the isolation of 94-239 from uranium and fission products.

Stoughton will continue his work on U233 and the development of methods for its separation from thorium and fission products.

In the late afternoon Isadore Perlman and I boarded the streamliner "City of San Francisco" for our move to Chicago.

This, then, terminates my account of the early Berkeley work. It will be continued as part of my description of the continuing work at the Metallurgical Laboratory at Chicago. [6, pg.148-149]

B Extracts from *History of Met Lab Section C-I, Volume 1*

1942-04-19 This morning at 9:30 a.m. Isadore Perlman and I arrived in Chicago aboard the City of San Francisco. Although our trip from Berkeley took almost two full days, we feel that the time has not been wasted. Many lively discussions ensued in the privacy of our bedroom and, with appropriate care, in the club car regarding ways to separate element 94 chemically from uranium (that will be neutron-irradiated in chain-reacting piles) and from the fission by-products that will be produced concurrently in the neutron-irradiation process. This overall problem of element 94 isolation will occupy most of our attention for some time to come.

This day marks my 30th birthday and a transition point in my life, for tomorrow I will take on the added responsibility of the 94 chemistry group at the Metallurgical Laboratory on the University of Chicago campus, the central component of the Metallurgical Project. [7, pg.1]

1942-04-20 Iz Perlman and I are now official members of the Metallurgical Laboratory, University of Chicago campus, and my salary for the time being has been set at $360 per month by Dick Doan, who has the title of Laboratory Director. [7, pg.2]

1942-04-21 I also spent a good deal of time continuing my conversations with von Halban. Halban and I had a talk with Compton about the importance of U233 as an alternate of 94-239.

In my continuing conversation with Compton I raised the question of Ray Stoughton's continuing our neon work (code name for thorium-U233 project) in Berkeley. Compton is very eager that he do so, and with all possible speed, as it may play an important part in the general program of the future. As part of our research program at Berkeley, Gofman, Stoughton and I showed that U233 has a slow-neutron fission cross section about 25% greater than that of U235. (Gofman is doing his Ph.D. research work with me at Berkeley, continuing there after my departure; he, together with Stoughton, working with me in a postdoctoral capacity, participated in the discovery and demonstration of the slow neutron fissionability of U233.) Also, it may turn out that U233 is superior to 94-239 because more secondary neutrons may be emitted during fission and it is easier to isolate chemically. I wrote to Ray informing him of this decision, which I am sure will please him. [7, pg.8]

1942-04-28 I dictated to Mrs. Sullivan some of the first draft of the complete report "Properties of U233" by G. T. Seaborg, J. W. Gofman and R. W. Stoughton. This will describe in detail our work at Berkeley on the discovery and determination of the slow neutron fissionability of U233. [7, pg.19]

1942-05-05 I prepared a letter to Gofman to be used in transmitting the rough draft of the write-up on U233 as soon as Mrs. Sullivan types it. I asked that Stoughton as the other co-author have a look at it too. In my talks with Halban he convinced me that we should be bombarding solid thorium nitrate rather than the saturated solution that we used last time. I asked Gofman to convey this information to Stoughton, who should prepare a solid thorium nitrate sample of about 75 pounds and use the same geometrical arrangement employed by Kamen, Wahl

and Kennedy in the big neutron bombardment of uranium that is under way to produce experiinental quantities of 94-239. [7, pg.34]

1942-06-02 Ray Stoughton, who is working in Room 311, and I went over the work that he has been doing on the Berkeley project. We also had a chance to talk about our joint paper, co-authored by Gofman, on the properties of U233. We expect its publication as a Laboratory Report in a week or so. It will be rather long; it sums up all our investigations on U233, including the various experiments performed to measure the radioactive and fission properties of U233 and the chemical procedures followed.

We also discussed future experiments on the separation of thorium-232, protactinium-233 and uranium-233 by the fluorination method. I proposed that he prepare some thorium fluoride incorporating 7-day U237 tracer and then send it to us at Chicago for fluorination. We will then return it so that he can identify the U237 to see whether it will be found in the volatile uranium hexafluoride fraction as expected. In addition, we discussed the optimum ratio of hydrogen to thorium for his forthcoming large bombardment of thorium with neutrons at the 60-inch cyclotron to produce more U233, and parent Pa233, for experimentation purposes. Ray is a good Ph.D. chemist and enthusiastic about his participation in the program. [7, pg.66]

1942-06-11 Our Report "Properties of U233" under the authorship of Gofman, Stoughton and me was issued today as Report CC-126. This covers our work at Berkeley and was written (dictated to Mrs. Sullivan) soon after my arrival in Chicago with the help of Gofman and Stoughton through correspondence. I had previously written an abstract of this report (Report A153) while still in Berkeley. In this more complete report we describe the processes for the production and chemical isolation of our 3.8 microgram sample of U233 (Sample J) and our measurements of the growth of U233 in chemically isolated samples of the parent Pa233, one of which led to an additional sample of U233 of 0.8 microgram. We describe our measurements on these two samples with our 300 mg radium-beryllium neutron source which have established the slow neutron fission cross section of U233 as about 1.3 times that of U235. We also report that the growth experiments have established a half-life for U233 as 120,000 years and that our measurements on its alpha particles indicate a range of 3.1 ± 0.2 cm of air (at 15°C, 760 mm). We also report that our search for spontaneous fission in U233 using the 3.8-microgram sample indicates that the "half-life" for this process may be greater than 10^{14} years. [7, pg.76]

1942-06-29 Stoughton wrote from Berkeley notifying me that he is sending two samples of thorium fluoride (containing 7-day U237 as tracer) for use in testing the U233-thorium fluoride volatility separation process which we discussed when I was in Berkeley the first of this month. Shortly after receiving the letter, the samples themselves were delivered and Brown and Hill set to work fluorinating them. They finished tonight and we airmailed the fractions back to Stoughton. [7, pg.95]

1942-06-30 I replied to Stoughton, informing him that his fluorinated samples are now on the way back and giving him suggestions on the procedures to follow in measuring them. I asked that

he order another fifty pounds of thorium nitrate in view of the slow delivery. Also informed him that I am distressed about the interruption in the bombardment of his thorium sample. [7, pg.97]

1942-07-09 I received a letter from Gofman saying he and Duffield are ready to start the fast neutron fission measurements as soon as the one-gram Ra-Be arrives; he has worked up the deuteron-bombarded thorium and is now following the decay of the samples. He reports that there is no hope of producing U233 by this method as the yield is too low. Also there appears to be a short-lived Pa232 decaying by beta emission into an alpha-emitting U232 of reasonably short half-life, which he will verify in about a week. He reassures me that the electroplated samples of U234 that Fontana gave him are truly U234. He has completed a long spontaneous fission count on the 0.4-microgram sample and stands ready to write a report on this and on the new Pa232-U232 isotopes. [7, pg.109-110]

1942-07-17 Ray Stoughton's letter was the bearer of good news. Just as Kamen has promised, the 60-inch cyclotron at Berkeley is now back in business and our thorium nitrate sample is receiving a good neutron bombardment. The sample went in Thursday of last week and soon will be removed for chemistry.

Ray went on to say that he has ordered the additional fifty pounds of thorium nitrate according to my request of June 30. He described the results of working up the two neutron-irradiated samples of thorium fluoride he sent us June 29 for fluorination for his thorium-uranium separation and which we returned the same day. The distillate from the fluorination yielded 25-35% of the uranium, and that which remained in the thorium fluoride residue was successfully extracted by digestion with fuming sulfuric acid. He also reported that 23 micrograms of Pa231 were obtained from the carnotite ore residue obtained from Morris Perlman and Bonner. He said that he, Wahl and Duffield would like me to send them past and future copies of the notes from my course (Tuesday evening lectures) in nuclear chemistry. These are the notes prepared by Coryell which are already available individually for a number of the lectures and which will soon be available as a total package. [7, pg.125]

1942-07-18 I also sent a telegram to Stoughton, referring to the thorium nitrate, "How many microampere hours do you plan to give it?" [7, pg.128]

1942-07-20 Stoughton sent a telegram in response to my inquiry about the thorium neutron bombardment. He said that it was completed last Wednesday midnight with 15,000 microampere-hours. [7, pg.133]

1942-08-06 Latimer arrived in Chicago this morning. He came to my office and we had an interesting discussion regarding the work in general at Berkeley and here. Later my secretary gave me a letter that just arrived from Stoughton in Berkeley. He was perturbed that their project has no A-1-A priority yet, although their secretary, Mrs. Moquin, expects the priority in a week or so, and asked for my help in pushing it through. They are finding it difficult to order equipment and electronic parts. I responded that I would see that something is done about it immediately. My letter also stated that I was pleased his U233 separation has gone

so well and noted how the U233 from this run will be used. (1) A sample of approximately five micrograms will go to Manley here so that he and others can determine the fast-neutron fission cross section of U233, and (2) as large a sample as possible will go to Gofman so that he can measure the spontaneous fission rate of U233. In regard to his problem of separating small amounts of uranium from large amounts of thorium, I suggested that U232 might serve as the best tracer in practice experiments with unirradiated thorium. I asked him to tell Gofman that Fermi extends him his most cordial thanks for the fission measurements made on UX1 and UY. [7, pg.168-169]

1942-09-13 Working in our apartment this afternoon, I dictated to Helen a memorandum on the position of U233 in our project, something that has been on my mind for some time. I have the feeling that insufficient attention is being given to the potential of this isotope as a back-up product should the nuclear properties of 94-239 and U235 be deficient in some respect. I feel that if I prepare and circulate a memorandum pointing out this possibility it will attract the required attention. In my memorandum I describe the production of U233 by the slow neutron bombardment of Th232 and also review its radioactive and fission properties. I point out that the U233 will have to be produced in conjunction with a chain-reacting pile operating on uranium and that the intensity of fission products can be kept relatively low because Th232 is not fissionable with slow neutrons. I also describe a number of possible methods for the chemical separation of U233 from the large masses of thorium, including the volatility process in which the volatile uranium hexafluoride is separated, the peroxide process in which the thorium and uranium are precipitated as peroxides with the subsequent separation of uranium by dissolving its peroxide in sodium hydroxide solution, and the fluoride method in which the thorium is precipitated as the fluoride leaving the soluble uranyl in solution. I emphasize that we have worked on all of these methods and that our research program on extraction processes for the separation of 94-239 from neutron-bombarded uranium also gives us a background of relevant experience. [13, pg.185]

1942-09-18 My other report, "Proposal for the Production and Use of U233," which I dictated to Helen last Sunday, is being issued as Report CF-268. [7, pg.251]

1942-09-19 I was glad to receive a letter from Stoughton today, but the contents were somewhat disturbing. His draft board in Bryan, Texas, referred him to Berkeley Local Board #70 for a physical examination, which he took last Wednesday night. It was announced during the exam that all who passed would be reclassified into 1A within ten days and then be inducted thereafter. Hamaker, he said, who had a similar experience a week earlier, received a notice on Thursday, the day the letter was mailed, that his draft board is going to leave him in 2A status until November 21, at which time it will consider his case for another six month's deferment.

Helen and I left today for my visit to Site X and a week-long vacation for us in the Smoky Mountains of Tennessee... Later, Helen and I took a train to Cincinnati, where in the evening we changed to another train for Knoxville, Tennessee. Gatlinburg (and the Smokies) is only a short but scenic bus ride from there. [7, pg.257]

1942-09-25 We took an early bus for Knoxville, dropped our bags at the Andrew Johnson Hotel and boarded another bus for Clinton, a small town that lies within a few miles of the farmlands that will be converted into Site X. Here we got off for a visit to Norris Dam and sightseeing until another bus came along to take us back to Knoxville where we spent the night in the Andrew Johnson Hotel. [7, pg.260]

1942-09-30 Stoughton wrote a four-page letter. He is sending me 250 microcuries of Pa233, and later some U232, that Gofman said I requested. He reports on his difficulty in working up the target from a 500 microampere-hour run of deuterons on thorium (to produce Pa232 and daughter U232). The big run on thorium plus neutrons (to produce Pa233 and daughter U233) apparently has a lower yield than expected; but the chemistry, which he describes in detail, went satisfactorily; he expects to find about 8 to 10 micrograms of U233.

"As I told you," he wrote, "Fontana is now working on the uranium-thorium separation with me. Since he wants to get as close to organic chemistry as possible, he is now working on the ether extraction and will later work on organic compounds of uranium and thorium as a means of separation. I am putting all available time on inorganic methods and any other methods which can be found. I believe several methods will work all right, but they all so far involve removing the thorium from the uranium, except for the ether extraction which does look somewhat promising. If any work has been done that you know of on the effect of radiation on organic material (especially ether), I would like to know about it, since the Pa233 will make the thorium very active." [7, pg.272]

1942-10-26 "(1) Space should be left adjoining Pile II in which to place thorium for the production of U233. An amount of U233 of the order of milligrams to grams will be useful for the further study of the neutron and spontaneous fission properties of U233. Likewise it is desirable to practice on the extraction of U233 from larger amounts of thorium than have been used so far. About one or two tons of thorium compound should be sufficient for this purpose, and therefore a sufficient amount of space to accommodate this amount of material will be needed. It does not seem advisable at this time to try to devise arrangements for placing the thorium within the pile; the placement of the thorium near the edge of the pile should be sufficient to meet the requirements of these experiments." [7, pg.314]

1942-10-31 Our report, "Production and Properties of U232" by Gofman and me, was issued today as Report CN-332. This covers work that has been done at Berkeley by Gofman during the last few months at my suggestion. He has found that the bombardment of thorium with the 16 MeV deuterons in the Berkeley 60-inch cyclotron produces, in addition to the 27.4-day Pa233, the new isotope Pa232. The Pa232 is produced by a (d,2n) reaction and decays with a half-life of 1.6 days by the emission of beta particles and gamma rays. The Pa232 decays to the alpha-emitting daughter U232 whose half-life of 30 years is estimated by measuring its yield of alpha particles produced from the complete decay of a sample of Pa232 whose absolute beta intensity was determined with the use of a calibrated Lauritsen electroscope. [7, pg.322]

1942-11-03 During the last few weeks I have been worried about an aspect of a nuclear weapon made of element 94 that might make it inoperable and have discussed this thoroughly with Perlman. Suppose we purify 94-239 for a bomb, but there is a minute trace of boron or other light element in it. Won't the alpha particles emitted by the 94 in its radioactive decay react with such light elements to produce neutrons and thus trigger the bomb prematurely? If so, what about the great concern that Oppenheimer and the other theoreticians have about the possibility that 94-239 has an unacceptable rate of spontaneous fission? The more I have pondered the matter the more I have become convinced of my conclusion. So I started making rough calculations. My calculations are inescapable: if I am right, then 49 for a working bomb would have to be purified from light elements beyond anyone's imagination. I have checked my concern with Teller and Manley, and they agree that the fate of the whole 49 project hangs in the balance.

By the time I reached my office this morning I already had composed a letter in my head to Oppenheimer which I dictated to Miss Smith (with a copy to Arthur Compton):

> "Dear Robert: "There is one point that has been worrying me quite a good deal since the discussion that we had in Berkeley the early part of last nonth. This relates to the number of neutrons that it is permissible to have present in the final 49 product in order not to cause complications in its control. You have stated that it would be ideal if the spontaneous fission rate were as long as 10^{19} years. If this figure is a desirable limit, and if you cannot get around it any other way, this creates a formidable chemical problem because of the possibility of forming neutrons from the (α,n) reaction on light element impurities. Since the alpha-emitting life of 49 is about 10^4 years, 10^{15} alphas must not produce an undesirable neutron. Assuming that an element like boron, for example, has a cross section for the (α,n) reaction of about 1 in 10^4, this would mean that boron must be absent in the final product to the extent of 1 part in 10^{11}.
>
> "You can see why I am disturbed if this calculation is correct and if you are actually limited by this requirement. Even if you can stand 10^5 times as many neutrons as this, making the limits of light impurities which are permissible one part in about a million, the final chemical purification is still a formidable problem. In fact, if these requirements are going to be placed upon our chemical procedures, I should like to know definitely about it as soon as possible since this will require a great deal of development work along lines which so far have never been attempted.
>
> "I have discussed this matter somewhat with Dr. Manley and Dr. Teller and I believe that they also consider it a rather serious problem." [7, pg.325-326]

1942-11-09 All last week I was in suspense wondering what Oppenheimer's response to my letter would be. Today I received his reply. He also sent copies to Manley, Teller and Compton. Because of its significance, I will quote in full:

> "Dear Glenn: "Thanks for your fine letter. The problem you raise is not a new

one, nor is it limited to the 49. I've thought about it a little in connection with the 25-24, and it is probably time to get possibilities clear.

"With the 25 alone matters would not be so bad: a concentration of light impurities (B, Be, Al, C, N) of 10^{-4} would give us the more than ideal 10^{19} years equivalent. If, as in our present set up, almost all the 24 goes along with the 25, then things are about 25 times worse. I believe that we can probably get along with 10^{17} years equivalent if we have to. Therefore I should say that the chemical problem with the 24-25 mixture was hard but probably soluble. Do you agree in this?

"With the 49, even allowing a factor 10 in your estimate of yield and a factor 100 in our requirements, it still comes to a purity of about 10^{-8}. Is this hopeless?

"It would be a help to know what you think on these points fairly soon, since if your answers are in the negative it will mean not only rather radical and I am afraid rather inefficient redesign, but also the immediate prospect of some new production problems for other materials.

"How bad is 23?

"Thanks again for your letter, and let me know what your ideas are this chemistry are as soon as you can.

"With all good greetings,"

It is clear that we have a very serious problem in the purification of 49 and that this is going to receive intensive top-level attention in the days immediately ahead. [7, pg.337-338]

1942-11-24 "New Isotopes.—Work on methods of separation of 94 is being continued. Development of design information for three of the most immediately promising types of extraction process is being continued by the chemical engineers, for handling one ton of metal per 24 hours. By bombardment of thorium with deuterons at Berkeley, the new isotope Pa232 has been produced. This decays with a half-life 1.6 days into U232, an alpha-emitter with half-life about 30 years. This U232 will be extremely useful for tracing very small amounts of uranium, *e.g.*, in the work of the health group. [7, pg.371]

1942-11-25 The meeting turned to a discussion of the relative merits of heavy water versus helium-cooled piles for the production plant. Later on I suggested that it would be desirable to push production of U233 inasmuch as uranium metallurgy is already understood and U233 could stand ten times as many impurities as 94-239; moreover, its nuclear properties are as well known. Its production rate is less than that of 94-239, but a heavy water pile would help decrease this disadvantage. [7, pg.376]

1942-12-02 The Lewis investigative committee is back in Chicago following their visit to Berkeley, presumably to inform Compton about the fate of the Metallurgical Laboratory. The first committee member I saw today was Greenewalt. We met late this afternoon in the corridor of Eckhart Hall. As he approached me, I could see from his demeanor he was bursting with good news. The aura of cheerfulness and excitement that he carried with him and the way he held out his hand in greeting told me that this signified more than just taking pleasure

in seeing me again. Then when I heard him say he had just come from the West Stands, I understood the reason for his jubilation. Fermi has produced a chain reaction—the pile is a success! Greenewalt said that Fermi, Whitaker, Zinn and their crew started the experiment this morning. As the control rods were cautiously withdrawn throughout the day a few inches at a tine, Fermi would take the new meter readings, and using his slide rule, would calculate the multiplication factor. Greenewalt said that he and Compton stood on the balcony, alongside Fermi and most of his crew, watching the proceedings. Then at 3:20 p.m., Fermi called for a few more centimeters. The pile became self-sustaining; in a few minutes the output of the chain reaction rose to one watt. Fermi ordered the reaction stopped, and everyone was tremendously relieved to see that the activity could be extinguished by shoving the rods back into place. The reaction took place without requiring as much material as Fermi earlier anticipated (and thus he was two weeks ahead of schedule), and it was not necessary to use the balloon to exclude air from the pile. However, he did use almost six tons of uranium metal, 50 tons of uranium oxide, and 400 tons of graphite, nearly as much as predicted. (I learned later that Zinn, Anderson and their pile builders reached a point where they knew the pile would be self-sustaining very early this morning but did not proceed to this historic point, leaving this dramatic act for a more convenient time later in the day.)

Of course we have no way of knowing if this is the first time a sustained chain reaction has been achieved. The Germans may have beaten us to it. I wonder, are they aware that U233 can be made from Th232 and 94-239 from U238 in a chain-reacting pile and that either of these isotopes can be used in a fission bomb? And if they have a pile that chain-reacts, would they use it to generate power or to produce vast amounts of radioactivity as a military weapon? One thing is certain; although Fermi has demonstrated that we now have a means of manufacturing 94-239 in copious amounts, it is the responsibility of chemists to show that the 94 can be extracted and purified to the degree required for a working bomb. [7, pg.390-391]

1942-12-09 I wrote to Stoughton complimenting him and Fontana on the excellent work with U233, as evidenced by a report they recently sent me. I said that the information office may decide to issue all U233 reports in a special category. It was Stoughton, along with Gofman and me, who demonstrated the fissionability of U233 last February 2, almost a year ago. I informed Stoughton, "I have made considerable progress within the last few weeks in selling the authorities here on the idea of producing and using U233 as an alternative to the use of 94-239 and U235. The advantages are becoming more and more apparent to those who make the decisions, and I think that there is no doubt that thorium will be placed around some of the operating piles. For example, there will probably be placed about a ton or two of thorium near pile number two (*i.e.*, the approximately 10 MW pile) in order to further test the usefulness of U233."

In my letter I said that there are a number of factors in the production and use of U233 that I did not bring out in my report CF-268, "Proposal for the Production and Use of U233," issued last September 18, and that I intend to write a supplementary report bringing out these facts as soon as I find time. I told him that Brown had done some more work on the

extraction of U233 using hydrogen fluoride and fluorine, and perhaps we can soon issue all our reports together. "I should like to keep in close touch with your work on U233," I continued, "and I will try from now on to keep you more closely informed on the progress which I make in actually arranging for the manufacture and use of this material. As you know, the use of U233 is a rather new idea (although it shouldn't be) to a number of the people who are running things, who are all wound up in their considerations about the use of 94-239 and U235." [7, pg.405]

1942-12-16 A report entitled "U233 Production and Extraction" (No. CC-384) is being issued covering the period to December 15, 1942, inaugurating a new monthly series of reports summarizing progress of work on U233. This report opens with a section "General Considerations on the Production of U233" written by me. Here I amplify the general considerations that I covered in report CF-268 issued on September 18 of this year. I describe the methods of production of U233 through the bombardment of Th232 by the neutrons produced in a chain-reacting pile, its possible use as an alternate source of energy for a nuclear weapon should the use of 94-239 or U235 prove to be not feasible, and various methods of chemical separation of the U233 and its immediate parent Pa233 from the neutron-irradiated Th232 and fission products. Also included are a report by R. W. Stoughton of the Berkeley Laboratory on "Production of U233 by Use of Thorium in Conjunction with the Uranium Pile," a report by B. J. Fontana of the Berkeley Laboratory on "Separation of Minute Amounts of Uranium from Large Quantities of Thorium by Ether Extraction," a report by Brown and Hill on "Volatility Methods for Isolating U233," and a report by A. C. G. Mitchell of Indiana University on plans to make measurements on the beta particles of Pa233. Stoughton at Berkeley is now working full time on the U233 problem while Fontana is devoting some of his time to this problem but is phasing into other work on plutonium chemistry. [7, pg.418-419]

1943-01-06 I had a long meeting with Stoughton to discuss his progress on the U233 program. I invited him to join my group at the Met Lab, on a permanent basis, to lead the U233 work there; this work will be phased out at Berkeley, continued and expanded at the Met Lab. Stoughton accepted my offer and will move to Chicago within about a month. [7, pg.447-448]

1943-01-20 The report "U233 Production and Extraction" for the month ending January 15, 1943, has been issued with the report number CC-426. In this report I have written a section entitled "Production of Experimental Amounts of U in Conjunction with Intermediate Power Chain-Reacting Structures" in which I describe procedures for the production of U233 by irradiating thorium in conjunction with a chain-reacting pile. I describe methods of extracting the intermediate Pa233, separating it from uranium, and then allowing it to decay in order to form pure U233 daughter; this method of isolation of U233 gets around the problem of its dilution with any initial uranium impurity that is present in the neutron-irradiated thorium. I describe volatility methods using the volatile fluoride of Pa233 and also aqueous chemical methods for isolating the Pa233. The method I describe is for the first production of pure U233 in small amounts in conjunction with the piles that the Metallurgical Project has un-

der consideration. In the ultimate production units where large amounts of U233 would be produced, the procedure may be to use uranium-free thorium and to extract the U233 itself rather than the Pa233 intermediate from the thorium. [7, pg.472]

1943-01-21 They asked me if I am still interested in irradiating some thorium in the pile [the second pile at Argonne] to produce U233, and I replied there isn't sufficient time to prepare the extraction equipment. I suggested that the chemical extraction plant for 94-239 at Argonne might occupy a building 20 feet x 80 feet in area including a chemistry laboratory and a counter room each 20 feet x 20 feet. [7, pg.473]

1943-02-07 Ray Stoughton, John Gofman and Cliff Garner arrived in Chicago from Berkeley; Ray is joining my section and the other two are visiting our laboratory. [7, pg.512]

1943-02-08 Raymond W. Stoughton started work in Section C-I as a Research Associate, coming from the University of California, Berkeley, where he worked with me on the discovery of U233, the demonstration of its slow neutron fissionability and the general problem of its production and chemical separation. **It is my intention that he carry on the U233 research here—in fact, be in charge of it—but in view of the urgency of work on the adsorption method for the separation of plutonium from uranium and fission products, I am having him start on this problem.** He will eventually make his headquarters in Room 2 but will be helping Willard in Room 11 in the interim. [7, pg.512]

1943-02-15 "Report for Period Ending February 13, 1943. U233 Production and Extraction" (Report No. CN-454) is being issued. In this Stoughton presents information on the two different methods under which a U233-thorium separation may be made: (1) directly, after a few months of Pa233 decay and (2) in two steps, the first separating the intermediate Pa233 from the thorium immediately after bombardment, and the second separating U233 from the remaining Pa233 after sufficient decay of the latter. He reports on experiments to attempt to increase the efficiency of some of the methods already under consideration for U233-Th separation and to seek a satisfactory organic precipitating agent. [7, pg.524-525]

1943-02-16 The first memorandum to Whitaker proposed that space be left with the pile at Site X for the irradiation of 50-100 pounds of thorium salt, which would produce enough U233 to make possible further studies of its nuclear properties. I suggested that slots be left in the center of the pile so as to allow for the placing of four 1"-diameter tubes about 20 feet long to contain the thorium salt. [7, pg.527]

1943-03-24 Stoughton submitted a four-page memorandum to me proposing experiments to be carried out for the purpose of testing various methods for separations of Pa233 from thorium; U233 from thorium; and U233 from Pa233. It was divided into five sections: I. Volatility Methods, II. Wet Methods, III. Ether Extraction for U233, Th and U233, Pa233 Separations, IV. Ultramicrochemical Experiments, and V. Preparations of thorium compounds for use in conjunction with a uranium pile. A final section was devoted to a suggested order of conducting the experiments. He noted: "The ether extraction method is considered very

important and it will undoubtedly be used in some part of the extraction, decontamination, purification of U233; it was put in Group III merely because it has already been pretty well investigated. In order to accomplish the planned work in a few months time, at least three additional men will be required." [7, pg.599-600]

1943-03-30 I submitted a memorandum to Compton outlining the duties of the 20 men from the "Chemistry of Final Products" section who will be at the Site X laboratory. Besides assisting the semiworks people with development work and trouble-shooting in the extraction plant, they will work on decontamination studies since there will be sufficient fission products present to give final answers on the extent of decontamination possible. There will also be some microchemists to prepare samples of the progressively increasing amounts of plutonium for shipment to Chicago and elsewhere. other problems to be handled will be (1) development of methods for extracting protoactinium and uranium-233 from the thorium salt to be irradiated in the pile; (2) to attempt the extraction of 93-237 as a means of determining the yield of the (n,2n) reaction in the operating pile and to obtain samples for chemical studies; (3) the development of methods for separating plutonium from the UF_6 tubes to be operated in the pile. [7, pg.616]

1943-04-12 Stoughton told me that last Wednesday he made lead absorption measurements on the gamma rays from the 27.4-day beta-emitting Pa233 using different Geiger counters and slightly differing set-ups, each designed to avoid the contributions of bremsstrahlung. He found by analysis of the absorption curves the presence of two gamma rays of 0.4 and 1.5 MeV, respectively. Judging from their intensity and the known large number of conversion electrons, he estimates that in Pa233 there are probably two gamma ray transitions per beta particle and about one-half gamma ray per beta particle emerges without internal conversion. [7, pg.647]

1943-04-13 At the meeeting of Section C-I Extraction Group Research Associates held in my office this evening, I reviewed the status of bombardments at St. Louis, the status of plans for Sites X and W as they affect our Section, and the properties and potential for use of U233. [7, pg.651]

1943-04-14 Paul A. Schulze started to work in Section C-I today as a Research Assistant, coming from the University of Chicago where he has been working on the OSRD uranium project directed by Professor Schlesinger. I plan to have him work on the U233 problem in Room 2 as soon as I can get a group under Stoughton started on this. [7, pg.651]

1943-04-19 Separation methods for extraction of U223 from thorium—Summaries are given of methods considered for separation of U233 from thorium (wet methods, volatility methods, ether extraction, etc.), separation of Pa233 from thorium (MnO_2 extraction, iodate concentration, wet fluoride) and separation of U233 from Pa233 (ether extraction, wet methods, volatility). [7, pg.663]

1943-04-23 Leonard I. Katzin, who arrived yesterday, started work as a Research Associate in Section C-I today, coming from the University of Rochester School of Medicine, where

he has been a research fellow in the Radiology Department. He graduated from UCLA in chemistry, where I knew him as a fellow chemistry student at the time of my attendance there. I am going to have him work with Stoughton in Room 2 on the U233 problem. [7, pg.674-675]

1943-04-26 Gofman and Sheline had dinner with us this evening. At the Chemistry Division seminar afterwards, Sheline described the present status of the Berkeley research on the Sodium Uranyl Acetate Process, Gofman summarized their investigations of 93 chemistry and fission products and Stoughton gave a status report on our U233 program. Waldo Cohn played an amusing record which is a sort of parody on the cyclotron. After the meeting I had a beer with Ghiorso, Gofman, Sheline, Boyd and Coryell at Hanley's, a beer joint on 55th Street. [7, pg.678]

C Extracts from *History of Met Lab Section C-I, Volume 2*

1943-05-26 Wigner called attention to some articles in the German scientific literature on the diffusion length of neutrons and their adsorption indicating that they were running neck and neck with us in 1941 and 1942 in their investigation of these subjects. There was also some discussion of using thorium in the pile shield in order to produce some uranium-233 as a second line of defense should the use of plutonium-239 prove not to be feasible. [8, pg.32]

1943-05-31 The U233 production and extraction group now consists of Katzin, Stoughton, Schulze, and Steahly. They have been working the last month or so on the development of methods for separating the U233 that will be produced by neutron bombardment of thorium in the pile at Site X. Since the concentration of U233 in the neutron-bombarded thorium will not exceed about one part in 10^7, it is necessary to isolate the U233 in the form of its parent, the Pa233 intermediate. They have developed methods for precipitating Pa233 from the dissolved neutron-bombarded thorium on manganese dioxide and also on thorium iodate in a partial precipitation of thorium. The decision has been made to bombard the thorium in the pile at Site X in the form of thorium carbonate because this compound dissolves quite readily in nitric acid and should contain no elements other than thorium which have high neutron-capture cross sections. [8, pg.38-39]

1943-06-01 I attended the regular Tuesday evening meeting of the extraction group of our section. Others present were Apple, Beaton, Brown, V. R. Cooper Cunningham, Dreher, English, Fries, Ghiorso, Hindman, Jaffey, Katzin, Orlemann, Peery, Perlman, Clifford Smith, Steahly, Thompson, Watters, and Willard. Katzin and Steahly discussed the choice of a thorium compound for bombardment in the pile at Site X. Present plans call for the use of thorium carbonate which will be compressed into cans, with about 80 pounds to be irradiated at one time. As the amount of U233 to be produced is small, it is planned to separate the parent Pa233 from the thorium in order to obtain the daughter u233 free from traces of ordinary uranium that may have been present in the original thorium. Experiments now in progress show that MnO_2 carries 90% of the Pa233 with only 1% of the thorium. The Pa233 concentrates in the precipitate when thorium is partially precipitated as the iodate. This method of concentration might work better at higher levels of Pa233 concentration than does the MnO_2 method. It is estimated that 4 or 5 mg of U233 may be formed in 80 pounds of thorium carbonate by 10 days' bombardment in the pile at Site X. Brown discussed current experiments he and Bohlmann are performing at Argonne to determine the feasibility of pumping UF_6. It is planned eventually to circulate UF_6 through a pipe inserted in the pile. [8, pg.44]

1943-06-08 Perlman sent a memo to Dr. Stone indicating additional possible hazards to personnel in the experimental work to be carried out by our section at Clinton Laboratories. The major new hazards anticipated are radiation from neptunium-239 during the isolation of the plutonium and from protactinium-233 during its separation from pile-irradiated thorium and the possibility of burns from fluorine gas and fluorides. [8, pg.51]

1943-06-15 Compton also reported his suggestion to Conant that within six months we should be giving emphasis to other aspects such as power production. Conant agreed in principle but questioned the date. In a discussion with Compton and Allison, Fermi indicated that probabilities are that U233 will be less useful than Pu239. Five hundred micrograms of U233 would be enough for neutron multiplication tests. [8, pg.56]

1943-06-19 The U233 production and extraction group, consisting of Katzin, Stoughton, Schulze, and Steahly, has found that Pa233 can be volatilized by hydrofluorination of neutron-bombarded thorium. [8, pg.59]

1943-06-26 "Chemical Research—U233 Production and Extraction. Report for Month Ending June 21, 1943" (CC-739) is being issued and includes the following report by Katzin, Schulze, Steahly, and Stoughton. The precipitation of Pa233 with MnO_2, from a solution of thorium nitrate has been studied in some detail and found satisfactory up to 36 gm $Th(NO_3)4$-$4H_2O$ per 100 cc solution. The concentration of Pa233 with respect to the manganese dioxide carrier and the thorium precipitated with the carrier appears to be accomplished quite well by either a series of MnO_2 cycles or thorium iodate cycles. Volatility methods for both the separation of Pa233 from the original thorium and for its separation from MnO_2 are now under investigation. After a study of its physical characteristics, it has been decided that thorium carbonate will be the thorium compound used in the pile at Site X. [8, pg.69]

1943-06-29 Tonight there was the regular Tuesday evening meeting of the Extraction Group of my section attended by Apple, Brown, V. R. Cooper, Cunningham, Davidson, Dean, Dreher, English, Ghiorso, Hindman, Katz, Morris, Peery, Potratz, Clifford Smith, Steahly, Stoughton, and Thompson. The meeting consisted of a review of current work on extraction methods. Stoughton outlined the work now being done on the effect of thorium to iodate ratio and total iodate concentration on the carrying of protactinium by the partial precipitation of thorium as the iodate. [8, pg.73]

1943-07-07 Whitaker mentioned it would probably be September 1 before the Clinton pile starts up and that he would like to have some definite plans and schedules on the materials that are to go into the pile. This brought up the subject of U233 production. Allison said that Newson is planning to put in enough thorium to make 1 milligram per day at 1,000 kW, and Compton remarked that Site Y has requested 2 milligrams in addition to what is needed here at Chicago. Compton then requested Allison to prepare a memo in collaboration with Fermi on the amounts of U233 required. [8, pg.84]

1943-07-17 Perlman sent a memorandum to Donald Hughes, Jr., Special Assistant to Compton, commenting on a letter from Conant to Compton regarding the relative amounts of gamma activity associated with the "23" system as compared with the "49" system. Perlman gave the results of computations on gamma activity associated with 250 grams of product (Pu239 or U233); these show that, at moderate intervals of time after pile shutdown (6 days to 60 days), the gamma activity of the two systems is comparable; at a shutdown time of one year, the U233 system is considerably more favorable with a fission product activity of 200 curies

for U233 versus 10,000 curies for Pu239. At the earlier time periods there is a considerable contribution from the 27-day Pa233 in the "23" system. [8, pg.93]

1943-07-26 There was a Laboratory Council Information Meeting on chemistry at 9:30 am, attended by Allison, Boyd, Brewer, Burton, Chipman, Compton, Conant, C. M. Cooper, Coryell, Davies, Doan, Fermi, Garrison, Gofman, Hilberry, Hughes, W. Johnson, Miles, Mulliken, Perlman, Potratz, Rubinson, Stearns, Stone, Sugarman, Wigner, Willard, Smyth later, Spedding later, and Johns even later. The presentation of the work of our Chemistry Section C-I was as follows. Willard discussed reduction methods for plutonium, waste treatment studies, the work of English, *et al.*, on control analysis procedures and the new alpha counter with magnetic deflector. Also cited was Cefola's work indicating possibility of a double salt of lanthanum and plutonium fluorides and the work on adsorption processes using Catex-284. **Perlman discussed the plans to obtain U233 from the Clinton pile for the study of physical properties of its production process.** [8, pg.106]

1943-08-07 "Chemical Research—U233 Production and Extraction, Report for Month Ending August 7, 1943" (CC-887) was issued. **Stoughton reported it is now planned that the thorium will be irradiated in the Site X pile in the form of the carbonate, with the extraction to be accomplished by dissolving the thorium carbonate in nitric acid and removing the intermediate Pa233 from this solution with manganese dioxide precipitation. The Pa233 will then be allowed to stand in a suitable medium until a large portion of it has decayed to U233 that will then be extracted and purified.** Experiments demonstrate that MnO_2 carries about 90% of the protactinium and less than one percent of tracer quantities of uranium. The latter finding is important from the standpoint of separation of the Pa233 from any natural uranium impurity in the thorium. [8, pg.122-123]

1943-08-12 I held the regular evening meeting of the Council of Section C-I in my office attended by Perlman, Brown, Cunningham, Willard, and Thompson. I pointed out there will be three groups under Perlman at Site X—English, in charge of electronics and extraction; Brown, in charge of dry methods and also concentration; Stoughton, in charge of the U2 problem. There is now great pressure on the plutonium metal production work, and Manning will be transferred from the extraction group to work with Kirk. It is evident that all the groups of Section C-I must have more men. Cunningham will take over Stoughton's U233 sample. It is now planned that the pile at Site X will start up on September 25 and by the first of November we should begin to get some production of plutonium from Site X. [8, pg.127]

1943-08-18 Compton mentioned that the emphasis on U233 is reduced at present because it would be impossible to produce military quantities of it before 1947. According to the present research program on U233, we will investigate the nuclear characteristics of U233 and then stop, unless Pu239 proves unsatisfactory as bomb material. [8, pg.134]

1943-08-24 I sent a memo to [Richard] Apple in the Technical Division about setting up a semi-works group for U233 work. As background information I say that in 1934 Amaldi, D'Agostino, Rasetti, and Segre published the first report of production of a 24-minute radioactivity from

the bombardment of thorium by neutrons. This was later shown by Gofman, Stoughton, and me (Report A-192 and CC-126, published June 11, 1942) to be the first step of the chain:

$$\mathrm{Th}^{232}(n,\gamma)\mathrm{Th}^{233} \xrightarrow[\beta]{23.5\mathrm{min}} \mathrm{Pa}^{233} \xrightarrow[\beta]{27.4\mathrm{days}} \mathrm{U}^{233} \xrightarrow[\alpha]{120{,}000\mathrm{yr}}$$

Our report stated that since U233 has a slow neutron fission cross section at least as great as that of U235, it becomes important to consider it as a substitute for the fissionable material which forms the basis of the present program.

A chemical program on preparation of U233 is now under way in my section, the memo continues, with a staff of four under the leadership of Stoughton, which will be continued under Perlman at Site X. The work has progressed to the point where small-scale production at Site X is feasible. The process depends on isolation of Pa233 from bombarded thorium carbonate by successive concentrations on manganese dioxide carrier. After the last concentration step, the material will be allowed to decay for a period of one to several half-lives (one to three months). It is then planned to precipitate the remaining Pa233 away from the U233 and purify the latter by appropriate means.

It should be pointed out, I write, that the reason for this primary isolation of Pa233 is the natural uranium content of the thorium used. Especial care must be taken to purify the Pa233 from uranium. The uranium impurity in the thorium carbonate is equal to or larger than the U233 to be expected as a product under Site X conditions. If we assume full rated power of the Site X pile, a charge of 65 pounds of $ThO(CO_3)$, irradiated for ten days, should contain about 4.5 mg of Pa233. If 2 mg of this can be recovered in the final concentration step (very possibly an optimistic figure) and two half-lives allowed for decay, 1.5 mg of U233 is available. Assuming 1 mg of this can be isolated in pure form, we can see that 65 pounds of $ThO(CO_3)$ must be irradiated in the pile for 10 days, Pa233 isolated, and two half-lives (8 weeks) of decay allowed before it is possible to isolate 1 mg of U233. These are minimum conditions, as the yield is probably optimistic.

I go on to say that inasmuch as present demands of experimental groups seem to be at least one milligram U233 a month, a considerable amount of work is entailed. In addition to the isolation of the product from the irradiated material, there is the labor and time of preparing and treating approximately 800 1-inch pellets of carbonate and filling and sealing 200 aluminum cans for each charge. There is also an important research program on volatility methods, improvements on present methods, and development for operation under other conditions, as at Site W. The manipulations of isolation themselves are greatly complicated by the intense radioactivity of Pa233 itself (20 curies per milligram) and, therefore, the necessity for remote control operations. Because of these factors, I tell Apple, the chemical group as constituted at present cannot adequately handle the whole assignment unaided. We believe, however, that a group of four or five persons to handle the semiworks portion of the aluminum can preparation and the extraction procedure should be sufficient to do the job. [8, pg.140-141]

1943-08-27 I received from Howe a summary of the arrangements for canning the pressed thorium carbonate to be irradiated in the Clinton pile. After being filled by Stoughton, the cans are to be sealed by the Aluminum Company of America in New Kensington, Pennsylvania, by welding and spinning down of the ends to simulate the slugs designed for the Clinton pile. [8, pg.146]

1943-09-02 Last month on the 16th Groves, in a letter to Compton, asked how much of the isotope U233 is in existence. Today Compton replied, "I can say with certainty that until material is produced at X, there will be only several micrograms of 23 available; 23 does not occur naturally and to date the total amount, produced by cyclotron bombardment of thorium, is at most 15 micrograms. This 23 is not separated from the thorium as only tracer amounts (hundredths of micrograms) have been used in preliminary development of chemical methods of separation for use at X. A few micrograms could probably be separated but this would be an upper limit." [8, pg.154]

1943-09-08 A U233 Production Group, under Stoughton, to obtain milligram amounts of pure U233 for determining physical constants and to develop extraction methods for removing uranium from thorium and protactinium from thorium and uranium (staff: Katzin, Schulze, Steahly, plus one or two more). [8, pg.159]

1943-09-15 In Chicago it is the last day of work at the Met Lab for Ray Stoughton and Paul Schulze. They are transferring to Clinton Laboratories and will report to Riordan of the Clinton Laboratories in Knoxville tomorrow. [8, pg.165]

1943-09-20 Last Saturday Compton was at the Montreal Laboratory in Canada, together with General Groves, Tolman, Smyth, and Wigner, to attend a meeting for the "Exchange of Information with the British." Such an exchange was sanctioned last month on August 19 when Roosevelt and Churchill signed the so-called Quebec Agreement, "Articles of Agreement Governing Collaboration Between the Authorities of the U.S.A. and the U.K. in the Matter of Tube Alloys." The British code word "Tube Alloys," of course, stands for the uranium project. Those representing the British at the Montreal meeting were Chadwick, Halban, Placzek, and Newell; and there were informal discussions between the Americans and Paneth, Auger, and Sir Edward Appleton, as well as other research men.

At this meeting Halban pointed out that they have been concerned with the relative merits of U233, U235, and Pu239. Much of their work has been aimed toward using U235 for producing U233 or Pu239. Compton mentioned that Wigner has emphasized likewise the possible advantage of transforming U235 into U233 or Pu239 as Halban suggests. Chadwick commented that his "money is on the U235," giving as his chief reason the probable occurrence of spontaneous fission neutrons in the other isotopes. These he anticipated from his experience (with which we Americans are unfamiliar) that neutrons are present with radon in clean nickel tubes to the extent of about 0.001 the number that are present in radon-beryllium mixtures. Compton and the other Americans were skeptical regarding the applicability of this observation to the presence of spontaneous fission neutrons with Pu239 but did not discuss the point. [8, pg.171-172]

1943-09-23 A batch of thorium carbonate pellets was shipped to Stoughton at Clinton Laboratories from New Kensington, Pennsylvania, where it was sent for encasing in aluminum cans. The pellets will be used in the pile to generate U233. [8, pg.182]

1943-10-15 (b) Two hundred aluminum cans have been filled with compressed thorium carbonate, taken to New Kensington for sealing, and shipped to Site X to be placed in the pile for the purpose of U233 production. [8, pg.209]

1943-10-19 The U233 group is working on handling 10-20 pound lots of thorium, in preparation for processing the $Th(CO_3)2$ that is to be irradiated in the Site X pile. [8, pg.212]

1943-10-28 Katzin wrote to Ray Stoughton in Oak Ridge, detailing the problems being encountered in preparing 1,300 cans of pressed thorium carbonate—a job that will require six to seven weeks. [8, pg.224]

1943-11-04 Stan and I are now staying at the Guest House in Oak Ridge. We went out to the X-10 area this morning, where there was much excitement. The reactor went critical for the first time at 5:07 a.m. Loading of the 1,248 channels with aluminum-encased uranium slugs started yesterday. Only 31 tons of uranium were required, which filled about half the channels. Whitaker expects to use twice this much uranium in the next month or so to bring the pile up to 1,000 kW, its design capacity. This is the world's third pile to become chain-reacting that we know of, the other two being the Chicago and Argonne piles. It is planned that 5 tons of the irradiated uranium slugs will be removed from the pile November 26. After cooling until December 9, the uranium will be put through the Bismuth Phosphate Process in the Chemical Separations Plant (200 area) followed by the concentration and isolation procedure that hopefully will yield 10-40 milligrams of plutonium by January 15. A run-through of the Chemical Separations Plant will start for the first time around November 24 when one ton of uranium metal from the Argonne pile will be processed for approximately 10 milligrams of plutonium. (See Figure 17 and 18.) [8, pg.234]

1943-11-08 On Wednesday Katzin sent Stoughton the current status of the pressing and canning of the thorium carbonate. He mentioned that the operation has been interrupted by a couple of emergencies that came up in the lab, but that the work was finished yesterday.

The first meeting of the P-9 Information Committee took place last Wednesday at 2:00 pm in Room 209, Eckhart Hall, with Wigner presiding. Others present were Allison, Burton, Chapin, Creutz, Hiskey, Hogness, Huffman, Ohlinger, Sugarman, Szilard, Vernon, Willard, Young, and Zinn. Wigner, speaking for the P-9 Steering Committee, reviewed the general status of the P-9 program and outlined in brief detail the events and directives that have led to the present program. He read the directive from General Groves for the Military Policy Committee that states "that our efforts (should) not exceed the construction of a low-powered heavy water pile for use as a general experimental tool and the setting up of a group of scientists assisted by a small number of engineers to study the general problems and to prepare the way in the event that it should become necessary to carry on rapid development." Wigner said that the application of this directive is interpreted by the Steering Committee as

meaning a selection of suitable arrangements for a production pile should it be needed, with the principal effort being concerned with the development of the slurry pile. He mentioned that at the present time there are only two groups working full time on the slurry pile: these are Vernon's and Hiskey's, representing engineering and chemical research.

After Wigner spoke, each of the individuals on the Committee discussed his function as far as the P-9 program is concerned. Willard, speaking for Section C-I, said that our section is making an effort to adapt existing methods to the extraction of plutonium produced in a slurry pile; Hogness asked us to summarize this for him in a memorandum. Allison informed the Committee his function is to serve as interim director of the P-9 Steering Committee until Smyth arrives from Princeton November 9 to take up his duties as Associate Laboratory Director and as director of the P-9 Steering Committee; he plans to spend alternate weeks in Chicago. Zinn stated that he is principally concerned with the immediate construction of the heterogeneous P-9 experimental pile to operate at 250 kW. Concrete for the shield is being poured at the present time. It was decided that the next meeting of the P-9 Information Committee is to take place at Argonne, November 10, and that Zinn will arrange for a bus to leave that day at 11:30 am from Eckhart Hall for Information Committee members. [8, pg.239]

1943-11-16 [Warren Johnson] also said that by this week 80 cans of thorium carbonate will be in the Clinton pile and should yield about 1 mg of U233. [8, pg.254]

1943-11-17 Compton gave his "State of the Nation" summation. He stated that the successful operation (better than expected) of the Clinton pile represents another milestone. The improved international military situation emphasizes the need to concentrate all possible effort toward bringing Hanford into production at the earliest possible date. Because of the national scientific manpower shortage, we must re-evaluate our program and transfer all possible effort from alternatives which cannot be useful within a year to those which have a high probability of such usefulness. The effect of this new emphasis on current programs is: (1) The P-9 project. Build the heavy water heterogeneous pile as a radiation tool, but discontinue work toward P-9 production units as they cannot offer a real alternative to present Hanford plans. (2) Dry fluoride work. A process adaptable to Hanford needs cannot be developed within the year. Hence, the work will have to be dropped, although use for purification and other purposes will be considered. (3) Uranium-233 studies. Without a P-9 power plant it is not possible to make enough U233 to be useful; therefore, this work should be discontinued for the present. [8, pg.258]

1943-11-18 At the 8:30 a.m. Section Chiefs' meeting in Franck's office, Hughes, Potratz, Franck, Sugarman, Burton, Hiskey, Manning, Allen, and I were informed by Franck that at yesterday's policy meeting the Project Council decided to abandon U233 work and dry fluoride work for extraction and decontamination. The P-9 program and adsorption and scouting work are to come up for discussion later.

I discussed the forthcoming directive from Conant that, in view of manpower shortage, attention be concentrated on "mainline" work; I expressed the view that since plans for production

of U have gone this far, it would be very desirable to do sufficient additional work to obtain a small quantity of pure U233 for physical measurements. [8, pg.258, 261]

1943-11-23 I sent a memo to Whitaker at Clinton Laboratories about the proposed curtailment of the U233 program. I gave the following information which may warrant consideration in deciding upon the rate of deceleration of the work:

> (1) There is valid reason to speculate that U233 may be the product for which the Germans are striving in connection with their large scale production of heavy water. Thus, it is important that we know its fission cross section, spontaneous fission rate, and neutron yield per fission. (2) Teller, who feels strongly that enough U233 should be produced and separated to make the basic physical measurements at Site Y, has stated that uranium containing only 10% U233 would be adequate for these measurements. With this tolerance for natural uranium it should be possible to extract and separate uranium directly (rather than via its protactinium parent) from the thorium being irradiated in the Clinton pile, a much easier task which would involve only 12 man-months. (3) To stop the U233 work before attaining the goal stated above would have a very bad effect on the morale of many of the chemists on the project since it would appear to them as an unreasonable reversal of policy.

[8, pg.264]

1943-11-25 I ran into Leo Szilard today, and I mentioned to him my concern that the capture of neutrons by Pu239 to form Pu240 might lead to problems in the design of a plutonium weapon if the capture cross section is appreciable and if Pu240 decays by spontaneous fission at an appreciable rate. He expressed doubt that this constitutes a problem.

Katzin wrote to Stoughton telling him that 250 cans of thorium carbonate should go out for welding tomorrow. [8, pg.266-267]

1943-11-26 In a memo to Franck, Allison repeated the objectives of Compton's memorandum, but added a comment that for Section C-I, the U233 production and recovery should be severely curtailed and that the present charge of thorium carbonate in the Clinton pile should be left there until the U233 that forms can be taken out as element 92 without being merely a small fraction of the material already present as uranium. He questions my contention that as many as three men will be needed during the four months (while the U233 is growing) to devise methods for separating uranium from thorium.

Allison expressed the view in a letter to General Groves that much more rapid progress in investigating enemy installations in Europe could be made if a limited number of Met Lab personnel were allowed to have some inkling of what Army Intelligence has discovered there. He suggested that, if we were allowed to learn what was found by the party that raided the Rjukan Hydroelectric Plant (heavy water) in Norway, or what information Professor Bohr

has concerning enemy activities in this field, we could draw conclusions which would not be apparent except to experts in the field of our project.

I received a copy of Stoughton's November 24 memo to Warren Johnson at Clinton that gives a proposed program for finishing the U-233 work with a minimum number of man-days. Stoughton proposes a two-step procedure whereby (1) some of the thorium carbonate will be irradiated only long enough to produce 10-100 micrograms of Pa233 that will be isolated through Pa233 separation, thereby reducing the radiation hazard by a factor of 100. This will provide sufficient material for specific activity measurements. (2) Irradiate another quantity of carbonate to produce one or more mg Pa233 that will be allowed to decay for about 2 half-lives; the resulting U233 is then to be separated, together with the natural uranium impurities, for physical measurements. [8, pg.269]

1943-11-29 I sent Franck a description of the present status of the U233 program at Site X, pointing out that we have placed 120 cans of thorium carbonate in the pile, that 60 have been withdrawn for experimental purposes after 4 kW-days of pile operation per average ton of uranium, and that 60 cans still remain in the pile and have received about 120 kW-days of irradiation per ton of uranium. I proposed that these 60 cans, plus an additional 38 cans on hand at Site X, be left in the pile for a few months after it starts up again, thereby producing about 1 ppm of U233 to be recovered by the direct method and not via the Pa intermediate. I suggested that the three men (Stoughton, a research associate, and a research assistant) be asked to work on the separation procedure as their first objective during the time the carbonate is being irradiated and then if they have any time left over, they might direct their efforts to other, mainline, work. [8, pg.270]

1943-12-02 After Cunningham, Kirk, and others finished, I discussed the possible formation of heavy isotopes in pile operation. It is my contention that the search for U236 should be carried out again in view of the very recent news from Site Y that the reaction U235 + n shows three sharp resonances. Inasmuch as U236 is probably an alpha emitter, the detection procedure should be to get uranium from pile metal and to determine accurately the specific alpha activity. It is also possible that resonance levels similar to those of U235 exist in the case of 94-239; a rough calculation shows an estimated ratio of 10-2 for the ratio 94-240/94-239 after 100 days operation in a future Site W pile. If this calculation is correct and if the half-life of 94-240 were about 200 years, the alpha emission from 94-240 would equal that from 94-239. A shorter half-life would greatly complicate the purification problem, particularly if the half-life were as short as six months, resulting in alpha particles with an energy of 7 MeV or greater which would make alpha,n reactions possible on O16 and N14. Spontaneous fission decay of 94-240 could also cause problems. If 94-240 should prove to be a beta emitter, then 95-240 would be formed that in all probability would be an alpha emitter and hence complicate the purification problem. I suggested it would be a desirable precaution to study the chemistry of element 95 with a view to separating it from plutonium if the need arises. To obtain element 95 for chemical studies, cyclotron-produced deuteron bombardments of plutonium should be tried. Another possibility which must be considered is the occurrence of neutron reactions with 93-239 in the pile, which could also lead to the

production of 94-240. I feel that the foregoing discussion constitutes a strong argument for continuation of the U233 work as insurance against unfavorable reactions in the pile. [8, pg.276]

1943-12-03 Plans for December and January at Clinton Laboratories call for dropping all work on the dry fluoride process and on the isolation of U233, using the fifteen men thus released to step-up activities on the process flowsheet (Bismuth Phosphate), and also on adsorption as an alternative extraction method which may prove useful in some part of the mainline process. [8, pg.276]

1943-12-08 In a discussion of the Met Lab program, Allison said that my section has three men working on the U233 problem, and he proposed that the thorium in the Clinton pile be left until appreciable amounts of U233 can be extracted, as Oppenheimer and Teller desire. Compton agreed that this program is a Chicago responsibility, whereupon Johnson said the cans should then be left in the pile indefinitely. [8, pg.283]

1943-12-15 Compton informed Greenewalt and me of a directive he has just received from General Groves. Groves asks that the Metallurgical Laboratory halt until further notice all work directed toward the development of a uranium-233 process and that the Clinton Laboratories discontinue all work with the chemistry of U233 until April 1 next year, when the position of this problem may be reconsidered. Compton has now implemented this directive by sending a letter to Allison and a memo to Whitaker. In order to make use of the materials already prepared, he further instructs Whitaker to leave the capsules containing thorium in the pile to accumulate U233; also, such additional capsules as may be already prepared should be inserted in the pile if this procedure does not interfere with its operation. [8, pg.289]

1943-12-18 Allison sent a memo to Compton referring to General Grove's directive that the U233 program should be postponed completely until further notice. Allison admitted that he had been misinformed when he claimed at the Project Council Policy Meeting that three men are working on this problem at the Met Lab, that actually only one man (Katzin) was employed in canning thorium for insertion in the pile. This will now be discontinued. He went on to say that the other chemists who are working on this problem have been moved to the Clinton Laboratories and the decision on their activities should be made by Whitaker and Doan. [8, pg.296]

1943-12-20 Warren C. Johnson reported that at Site X the dry fluoride work was terminated during the month, as has been the U233 program. Cans of thorium will be left in the pile until April, when some 15 mg of U will have been produced. [8, pg.301]

1943-12-21 In a discussion of some of the chemical results obtained at Clinton, Johnson mentioned that 40 one-half pound cans of thorium carbonate are in the pile and that 50 more are to be added at the next shutdown. The yield will be 15 mg U233 by next April, assuming 500 kW average power. The sample will be 20% contaminated with natural uranium impurity. Johnson also mentioned that the dry fluoride work has been stopped.

Someone showed me a copy of the current "Newsweek." After remarking about the Nazi superiority to the Allies in certain kinds of technological development, *e.g.*, rocket-glider-bombs, the article states that "there is the so-called German secret weapon. The propaganda concerning it is not being discounted as usual in Allied capitals, and last week the Nazis unofficially announced that it would be used against Britain within fifteen days:" Some of us suspect this announcement might herald the explosion of an atomic bomb!

There was a meeting of the administrative staff at which Munnecke, having recently returned from Washington, reported that the draft deferment situation is becoming difficult and that shortly an order will be issued cancelling deferment for anyone not past his twenty-third birthday, except that such deferment may be granted by the State or National Director of Selective Service. The mood of the administration here is to stop hiring men who are under age 38. Stearns believes that we should do more toward the training of women to replace men. It was suggested that a survey be made of the male employees under age 23 because wholesale drafting of these employees could cause serious damage to our scientific program. [8, pg.302-304]

1943-12-28 The "Monthly Report on the Activities of the Chemical Division, December 20, 1943" (MUC-JF-114) is being issued by Franck. Activities of Section C-1 are reported as follows:

U-233 manpower (this month) 1 (next month) 0 (priority) B-3 (lowest on the list) [8, pg.322]

1944-01-08 Greagor, Kay, Warren Johnson, Hogness, and I met today as a Steering Group. We discussed the distribution of manpower required to effectively work on the problems assigned to Clinton and Chicago. A proposal was made concerning the transfer of personnel to meet manpower requirements. Seven men from my section will be transferred to Clinton as soon as feasible. Four of these men will go directly to the Separations Division at Clinton. The time for the transfers will depend in part upon the reassignment of men from Perlman's group to the Separations Division and upon the procurement of additional personnel elsewhere. **Stoughton, Frank Steahly, and Paul Schulze are slated for immediate transfer.** [8, pg.339]

1944-01-20 I also reported that all our work on uranium-233 is being discontinued except for that on the material now actually in the pile. [8, pg.379]

1944-01-27 I sent a rather long memorandum to Compton outlining the possible utilization of uranium-233 by the enemy, in response to Compton's request. I give reasons why I think the Germans may be able to produce at least one uranium-233 device in the near future. I suggest this possibility be considered and be brought to the attention of our High Command in order that steps may be taken to distribute governmental facilities of the Allied Powers in such a way that a single device alone could not deal a fatal blow. It seems highly desirable that we sustain enough effort on the uranium-233 problem in order that we will be able to evaluate any developments the enemy may claim or employ. I gave the following reasons why I think it probable that the Germans may be preparing to use a U-233 device:

(a) "We know that the Germans are making strenuous efforts to continue heavy water production in Norway, and there seems to be no other possible use for this material other than a nuclear power program. Likewise, we can infer that such efforts would not continue unless they had in mind an overall heavy water production program in other places as well as Norway, calculated to make or have made an amount sufficient for their final objective within a reasonable length of time.

(b) "U233 can be separated from its parent thorium by relatively well-known chemical processes. The isolation of U233 may be very simple compared to that of Pu239 or U235. This is in part due to the fact that essentially only one radioactive species (Pa233) is involved in the separation. I believe that the Germans could handle this separation problem in one of their large industrial chemical companies, who in turn would isolate the U with only slight modifications in their existing equipment.

(c) "The tolerance for light element impurities (which must be removed to prevent neutron production by the α, n reaction) is some tenfold greater than that of Pu239. Furthermore such purification as must be obtained can be tested with uranium, which, in contrast with Pu239, would have been available for experiments during production of the actual U233. Our own present knowledge of the purification and metal production of uranium leads us to the practically certain conclusion that this problem is rather easily solvable for U233. If we should run into serious difficulties in the final processing of Pu239, it is conceivable that the Germans might produce a final U233 device before we produce one from Pu239 even if their program were not very far along today.

(d) "Raw materials for U233 production are probably available to the Germans. Using a uranium-heavy water pile in conjunction with thorium, it is quite probable that they would not need to meet the high purity standards for these raw materials that we have been required to meet for use in the uranium-graphite pile. It may even be that they have not needed to make either uranium or thorium metal, but can employ compounds of these elements in the system. The construction of a heavy water pile and its operation and attendant chemical extraction work for the preparation of the U233 device might be carried out without requiring a serious drain on the manpower or natural resources of the enemy." [8, pg.398-399]

1944-02-07 Gofman and Sheline sent me a joint letter which arrived Saturday. They were following up on a recent conversation with me at Oak Ridge in which I mentioned the possibilities of positions at Site W. They asked for details of the proposition such as type of work, rank or position, and salary. [8, pg.416]

1944-02-14 I wrote Gofman answering in detail the many questions he and Sheline raised in their February 1 letter about positions at Site W. Concerning their query as to when they might start work at Hanford, I suggested to Gofman that if he were one of Willard's three to five

supervisors directing research efforts, the starting time would be in July. I also said that most of my answers apply to Sheline as well as to him. [8, pg.428]

1944-02-29 Compton wrote that he is transmitting to General Groves and others in the Army who are concerned, copies of my memorandum on the possibility of the utilization of U233 by the enemy. **Compton added that it is his own judgment that such a U233 program probably would require more time and industrial effort to develop a conclusion than the Pu239 program. Also, he does not see any strong reason to believe that the Germans are actually engaged upon a program of U233 production.** He does agree, however, that work on its investigation should be kept alive and considers that my suggestion of encouraging the Canadians to follow this line is well worth following up. [8, pg.456]

1944-03-23 A "Status Report on U233 Program" has been written by Leonard Katzin and was submitted for distribution as a Laboratory Report today. Part I covers the discovery of U233, its properties and possible advantages over Pu239, and the present status of its isolation. Part II is concerned with the preparation of U233 and Part III summarizes the chemical extraction of U233 indirectly, in which Pa 233 is the intermediate, and directly. [8, pg.501]

D Extracts from *History of Met Lab Section C-I, Volume 3*

1944-05-08 My office received a memo to me from Allison on Friday asking if it would be possible for us to send 10 pounds of thorium carbonate to Montreal for use by Jules Gueron and A. G. Maddock. They are interested in the extraction of U233 from thorium. [9, pg.8]

1944-05-17 At 8:00 a.m. I held a meeting in my office of the Council for our section, attended by Albaugh, Baumbach, Cunningham, Davidson, Dawson, Dreher, Ghiorso, Hindman, Katzin, Orlemann, Simpson, Thompson, Watt, and Willard. I mentioned that the Argonne heavy water pile has been started with considerably less heavy water than expected. I reported that Fermi, using the one gram 94-239 sample we prepared for him, has found evidence for the absorption of neutrons in 94-239 to form 94-240; thus the properties of 94-240 will be of critical importance. [9, pg.22]

The Project Council Policy Meeting was held at 9:00 a.m. in Room 209, Eckhart Hall, attended by Allison, Chapman, Chipman, Compton, C. M. Cooper, Eastman, Fermi, Greager, Hilberry, Jeffries, Johnson, Mulliken, Oppenheimer, Spedding, Stearns, Stone, Szilard, Vernon, Warner, W. W. Watson, Whitaker, and Wigner. In his "State of the Nation" message, Compton gave the latest information on the Hanford schedule: The first pile unit will be operating at power in late August or early September. The first extraction plant is to be completed September 1, with first actual operations November 1. Problems still not completely solved include canning and film. Decontamination and purification are still not completely in hand.

Fermi initiated an extensive discussion of the difficulty of jacketing slugs which will withstand full power operation; the possibility of using long helium-filled tubes as a substitute was considered. Fermi pointed out that although the P-9 pile at Argonne has been brought to criticality, it has not been operated at power because of the lack of sufficient heavy water to cool it. This shortcoming will be remedied in another month.

Compton mentioned that the basis for the interchange of information with the Canadians is still under discussion. We are not at present free to exchange information on 49 and its chemistry. Whitaker stated that Clinton will be ready to test chemical separations with a full number of cells about June 15, and this will be followed by tests of the Hanford process. The last few groups of eight batches have given overall yields above 80%. Whitaker also raised the question of what to do with the 20 lbs of thorium in the pile. About 50 mg of U233 have been accumulated. Compton questioned whether it would be necessary to push it out at all, to which Whitaker replied that it is only a matter of gas pressure. Oppenheimer stated that he would be glad to study the U233 if the chemistry can be handled. [9, pg.23]

1944-05-19 At 9:00 a.m. I attended a meeting at which Fermi reported on recent values of nuclear constants of U235 and Pu239. Fermi has recently measured the absorption cross section of Pu239 for thermal neutrons by the transmission method. His absorber consisted of the mixture of one gram of plutonium oxide and two grams of graphite powder sample which we prepared for him. His measured value is 950 barns. Fermi also reviewed the present

status of the branching ratio (neutron capture to fission) of U235 and Pu239. He has found evidence pointing to a greater branching ratio in Pu239 than in U235. **This means 49 has a large n, γ cross section.** He has measured the number of neutrons emitted in fission per neutron captured to be slightly less for Pu239 than for U235; the number of neutrons emitted per fission is somewhat larger for Pu239, being 2.8 as compared with 2.4. [9, pg.25-26]

1944-05-31 Willard and I visited the sites (100 areas) where the plutonium production piles are under construction and the sites (200 areas) where the plutonium chemical extraction plants are under construction (Figs. 2 and 3). It is an awe-inspiring experience to see the thousands of workmen busily engaged in the building of these complicated edifices. These are located in a vast expanse of area (almost 500,000 acres in all) with the piles near the Columbia River for cooling purposes and the extraction plants somewhat removed. To reach these areas we drove over some of the flattest, most lonesome territory I have ever seen.

For lunch we were taken into the Hanford Camp where the construction workers live in rows of barracks, tents, and trailers stretched out in all directions. We ate in the largest mess hall (Fig. 4) I have ever been in—a sea of faces all being well-fed in shifts. We were told that there are some 40,000 residing at Hanford.

After lunch, Simon, who is serving as Hanford's Mayor, took us on a tour of the Hanford Camp. This included a visit to the jail, which was full of the motliest aggregation of tough-looking characters that I have ever seen. We learned that in order to find the large quantity of labor needed, it is necessary to hire "off the street," so to speak, and they have a large turnover with a substantial element of people of dubious character. We learned there was a murder last night and that this is a rather common occurrence.

After continuing our visits at the construction sites, which ran until quite late in the afternoon, we started back to Richland with Simon in his automobile. Being impatient with the secondary roads in the area (actually, there are no other kind), he decided to take a short-cut straight across the desert. In a particularly isolated area the wheels of Walter's car got trapped in loose sand. Fortunately, we discovered a water hole nearby, and Willard and I carried water over to pack down the sand to create a little better traction. However, Walter spun the wheels of his car before we had finished our compacting project, and he impatiently dug the car in deeper and deeper. With a great deal of persuasion and some restraint, John and I finally succeeded in slowing Walter down, and we managed to get free and make our way back to town. [9, pg.41]

1944-06-03 The report, "Metallurgical Laboratory Progress Report for April 1944," (MUC-SKA-665) was issued today. Items of special interest to me that are included in the report of the Nuclear Physics Division are:

"The construction of the P-9 reacting pile is essentially complete. All the main items of equipment are now on hand. The metal rods have been jacketed and welded and are now being given the final test. It will be started as soon as sufficient P-9 is on hand.

"The cross section for absorption of thermal neutrons by 49 has been found to be 950×10^{-24} cm^2 for neutrons of 2200 meters per second velocity. Assuming the ratio between the fission

cross sections of 49 and 25 to be 1.4 (as recently measured at Y) and assuming further that the capture branching ratio is equal for the two isotopes, one would find a cross section of about 900. There is, therefore, some indication that a may be slightly larger for 49 than it is for 25, although the difference is not outside of experimental error.

"The number of neutrons emitted by 49 and 25 has been compared. The ranges of the two types of neutrons for slowing down in graphite are equal within the experimental error. The number of neutrons emitted by 49 per neutron absorbed seems to be slightly less than the corresponding number for 25. A direct comparison of the ν values for the two isotopes is in progress.

"A second attempt has been made in order to determine whether the activity of a 25 sample irradiated at Clinton increases due to the formation of an alpha-active isotope. No evidence was found, and the result indicates that if 26 is formed, its lifetime must be greater than 600,000 years." [9, pg.52-53]

1944-06-05 Steacie, Goldschmidt, Maddock, and Greenwood from the Montreal Group arrived at the Met Lab for a visit and to participate in technical cooperation discussions on P-9 problems. I joined Steacie, Goldschmidt, and Maddock to discuss the chemistry of U233 and its separation from Th232. Meetings are scheduled through Thursday of this week. [9, pg.56]

1944-06-08 A summary meeting was held with Montreal Laboratory representatives following a series of conferences that took place while they have been here. The agenda for these conferences covered (1) separation of U233, (2) irradiation of materials at Site X, (3) protection of chemical operations and design of special laboratories, (4) radiation chemistry, (5) chemistry of film formation in cooling water, (6) liaison on analytical work, (7) corrosion chemistry, (8) chemistry of the homogeneous experiment, (9) metallurgy, and (10) canning.

At the summary meeting I was requested to provide (a) the latest type of alpha counter and FP-54 chamber, (b) two cans of thorium carbonate from the material already irradiated at Site X, and (c) U232 tracer material, separated or preferably in the form of a target. I was also asked to take up the question of putting repurified thorium compounds into the Clinton pile for neutron irradiation. [9, pg.8]

1944-07-04 Although this is Independence Day and was celebrated throughout the city, work went on as usual at the Met Lab.

At 10:30 a.m. I attended the Project Council Information Meeting on Nuclear Physics. Zinn presented a terminal report on the construction of the P-9 pile at Argonne. Start-up occurred on June 20 when 62 tons of satisfactory heavy water had been accumulated. The pile is designed for operation at 250 kW but has not yet been run above 190 kW. Preliminary measurements indicate that at 250 kW the neutron flux in the center of the pile will be 9×10^{11} n/cm^2/sec. By way of comparison, the flux at the center of the Site X pile is 4×10^{11} n/cm^2/sec at 1,700 kW.

The lattice dimension of oxides of the four heaviest elements show the following regularity: ThO_2 = 5.59; UO_2 = 5.46; NpO_2 = 5.42; PuO_2 = 5.39. Thus the crystal structure data give unambiguous evidence that the 5f shell is being filled in the elements starting from thorium. In analogy to the early members of the rare earths (lanthanides), these elements may be called thorides. [9, pg.120-121]

1944-07-05 Compton read a letter from General Groves concerning Project programs and personnel during the coming year. We are instructed to freeze our present level of personnel and to consider possible reductions in personnel that can be carried out without sacrificing effort on work believed to have potential value in the present war. No discussion of reduction in staff should at present be held with members of the Project in less responsible positions. Compton explained that the freeze means that personnel numbers are not to increase, but replacements for losses are in some cases allowable. He asked that consideration be given to reductions in staff as work goes into a stand-by basis—perhaps reductions between the limits of 25% to 75%. Timing will vary, but we might expect the reductions to start about September of this year and be completed by next January. There is no consideration of dropping effort on jobs with war significance. Hogness mentioned the difficulty of keeping strong men under stand-by situations. He also pointed out that the development of solvent extraction processes is one of our most important jobs, but there is some question about it being useful in this war.

Smyth urged that peacetime planning be pressed in order to avoid drifting into an untenable situation when, for instance, Germany collapses. Compton asked that each Division Director prepare a suitable program and personnel plans for longer-term work. He also agreed we must prepare a long-range program insofar as it is feasible to do so.

Compton read another letter from Groves requesting recommendations for a program in connection with the use of U233. Whitaker said that 60 slugs are now out of the pile, containing perhaps 150 milligrams of U233. Two slugs have been sent to Montreal. Fermi said that two or three milligrams of U233 are needed to get started and that Site Y has asked how they should go about obtaining 1-2 mg. Compton said he believes that we should provide Site Y with an estimated delivery date; the U233 work is still third priority class but now near the top. He asked that Hogness review the situation in the Chemistry Division and attempt to get a separation process going. If it turns out the Project should extract U233 from the full 60 slugs, the logical place would be Clinton Lab with the process development carried out at Chicago. Compton will also investigate the possibility of the Montreal people agreeing to do the extracting. [9, pg.124-125]

1944-07-07 Kay talked about progress at Hanford, emphasizing that the first material to be processed at Hanford will run between 5 and 50 grams of plutonium per ton of uranium. The pile may operate at 30,000 kW in September, 60,000 kW in October, and 120,000 kW in November. Building 200 T (first canyon) could start processing the first output of plutonium on September 15—Kay thinks it will be a miracle if we make it. The Hanford demonstration runs will be finished at Clinton on September 1, after which Clinton will study means of

increasing production. [9, pg.125]

1944-07-10 Katzin commenced setting up for the extraction of U233 from thorium carbonate cans irradiated in the Clinton pile. [9, pg.128]

1944-07-12 At 8:00 a.m. I held a meeting of the Council of Section C-I in my office...I brought up a number of items: (a) Security violations must be eliminated. (b) There has been a limited authorization for work along other lines, such as the U233 work. In line with this, four periods of the work are now recognized: (1) present, (b) stand-by, (3) transition, and (4) postwar. (c) A report is to be prepared by the sub-section chiefs on what should be done in the period up to October 1 (the stand-by period). This will be due July 24. These reports should include an estimate of the manpower required. (d) Manpower is now frozen. No new men can be added. Orlemann noted that the glass shop is the worst bottleneck in getting our work done.

I expressed my opinion that the need for semiworks operation within the next few months will be concerned with (f) the development of methods for the extraction of U233 and Pa233 from thorium... [9, pg.130-131]

1944-07-15 Katzin has worked out the procedure for extracting U233 directly from the Clinton pile-irradiated thorium carbonate, without preliminary Pa233 separation via MnO_2. Ether extraction will be used. The carbonate will be dissolved, siphoned over in 100-ml portions to the extraction vessel, NH4NO_3 added, followed by ether extraction. [9, pg.136]

1944-07-17 I gave a summary of my paper, "Inferences Concerning the Electronic Structure of 49 from Chemical and Physical Properties," which I dictated last Friday. I pointed out the probability that some sort of a transition group should begin in the neighborhood of the elements in the seventh period (elements 89-94). After reviewing the chemical and physical properties of neptunium, plutonium, thorium, and uranium, I suggested that:

> An attractive hypothesis is that this rare earth-like series begins with Ac in the same sense that the 'lanthanide' series begins with La. On this basis, it might be termed the 'actinide' series, and the first 5f electron might appear in Th. Thus, the ground state of Th might have the structure $5f^1 6d 7s^2$ beyond the radon core. With an 'actinide' series, U might have the electron configuration $5f^3 6d 7s^2$. It is very interesting to note that Kiess, Humphreys, and Laun in the recent report A-1747, in which they give a preliminary description of the analysis of the spectrum of neutral U atoms, come to the conclusion that the electron configuration of the lowest state of U is $5f^3 6d 7s^2$ (with the term symbol $^5L_6$), which supports the above view.

I also speculated about the chemical properties of the series members which we have not yet had an opportunity to study: $_{91}$Pa, element 96 (possible configuration $5f^7 6d^1 7s^1$) and element 95 (possible configuration $5f^7 7s^2$).

Spedding pointed out that while he agrees with me in the main, he thinks it would be extremely dangerous to depend on the existence of any super rare-earth group occurring, or that if such a group does appear that it would start at the same element for the various valency numbers. In summary, he stated:

> To sum up, due to the fact that the energy difference is rather small between whether an electron goes into the 5f or 6d and that the resonance effects are very large, the latter probably predominates in determining which level occurs lowest. Accordingly, any rule which might be derived for a new rare earth type series starting at actinium, thorium, or uranium would probably have more exceptions than regularities, and these exceptions should become more marked as the atomic number increases since the spectra becomes more complicated. I would therefore predict that it would be difficult to make any simple set of rules as to the valency states of the eka-uranium elements. [9, pg.141]

During the month of June the study of the heterogeneous heavy metal-light water system proceeded rapidly with the theoretical section of the Nuclear Physics Division at the Metallurgical Laboratory cooperating with experimental physicists at Clinton Laboratories. Lattices were tested that proved to be so close to the chain reacting condition that there is no doubt that with slight and practicable modications, a chain reacting system can be set up. The heterogeneous heavy metal-heavy water pile, which was shown to be chain reacting in May, has been modified during the month so that operation at power in early July is anticipated,

During the month a development of great interest took place in that an alteration of the isotopic constitution of an element by neutron absorption was demonstrated. Samarium, which had been exposed in the Clinton pile, was examined with the mass-spectroscope, and an increase of isotope 150 with a corresponding decrease in 149 was found. This identified 149 as the high cross section neutron absorber in samarium and has important consequences in the pile poisoning problem.

It was shown that beryllium and thorium of commercial purity can be extruded at commercial temperatures and pressures. [9, pg.144]

> There was an evening meeting of the Project Advisory Board in Room 209, Eckhart Hall, called by Compton, preceded by a dinner at 6:00 p.m. The program was scheduled to discuss (a) postwar plans for the Project as a guide for present changes in Project policy and organization; (b) the importance of light water moderated units in the overall Project program and the effect on transfers of associated personnel required if the program is to be pushed.
>
> The meeting, however, concentrated on something that came up which was much more immediate. **Robert Oppenheimer, who was attending the Board Meeting from Los Alamos, announced that E. Segre, O. Chamberlain, and G. W. Farwell have found strong evidence for the existence of the plutonium isotope**

Pu240, which undergoes decay by spontaneous fission! This was found in the neutron-irradiated Pu239 from the Clinton pile that we had purified for them.

It should be noted that this disclosure came as a great shock to everyone. However, over a year ago in my "Report of Month Ending March 15, 1943, Special Chemistry of 94," (CK-514), **I had written about the possibility of an n, γ reaction on Pu239 giving the isotope Pu240. I had said that a possible spontaneous fission decay of this isotope would seriously impinge upon our ability to use Pu239 as intended. Now it was learned that indeed this reaction takes place; and that since the neutron flux in the Hanford piles would be so high, Pu240 would be produced in so great a relative abundance that the neutrons resulting from its spontaneous fission would overshadow those from any α, n reactions on impurities that might be present in Pu239.**

Because of this new development, Site Y will now have to rethink how it will proceed in the design of a plutonium bomb. Furthermore, it was decided at this Board Meeting to demobilize Thomas's staff which is handling the coordination and general direction of the chemistry, purification, and final metallurgy of Pu239 because the planned extreme purification of plutonium would be futile—this could not prevent the emission of the unwanted neutrons. This meant that there would be no further monthly meetings at the Met Lab on this aspect of the chemical program. [9, pg.145]

1944-07-19 After the Project Policy Meeting this morning, Hogness and Warner came to my office to tell me that the purification program is no longer needed. I was standing in the hall in front of my office as they approached. They said that Compton had agreed I should be given the reason but that I was not to tell others. I said they didn't need to tell me the reason—**I assumed the spontaneous fission rate of Pu240 has been found to be so high as to overshadow the neutrons from the α, n reaction.** I went on to say that, since no one has given me this information, I feel free to pass my interpretation on to my men. [9, pg.147]

> The Project Council Policy Meeting met at 9:30 a.m. in Room 209, Eckhart Hall, attended by Allison, Chapman, Chipman, Compton, C. M. Cooper, Dempster, Doan, Fermi, Franck, Greager, Hilberry, Hogness, Jacobson, Jeffries, Johnson, A. V. Peterson, Spedding, Szilard, Thomas, K. Tracy, Vernon, Warner, Whitaker, Wigner, and Zinn.
>
> Compton took up in more detail the decision reached at the meeting of the Project Advisory Board Monday night, namely, that the need for exceedingly high purity plutonium no longer exists and that the present intensive work on purification can be dropped. Hogness asked if it would be proper to inform others on our staffs that it is the properties of plutonium that have made the change in emphasis necessary; adding that he was "thinking especially of Seaborg." Compton said he believed I should know. The section chiefs at Argonne also should know, he added, but it is highly essential to limit the information to the smallest number of people. Fermi mentioned the implications of such a finding must be known in order to plan properly for the construction of new piles.

Compton stated that there is still an urgent need for chemists on war jobs both within and without our Project. He mentioned that a committee is being formed of people who are not now working on this Project to appraise future potentialities and develop plans which can be acted upon by Congress.

At Compton's request, consultant Zay Jeffries set forth his ideas as to the future of atomic energy by reading a memorandum he has prepared. He likened the nuclear field to the electronics field thirty years ago, predicting a bright future for both. He suggested the word "nucleonics" as a name for the new nuclear field. Among the possible postwar uses he identified were tagged-atom experiments in scientific studies and industrial processes, related need for electronic instrumentation, piles for manufacture of artificial radioactive substances and power. In the latter connection he speculated on the use of piles in making desert or semi-arid land productive. He discussed the possible use of nuclear detonations to relieve geological faults and to form a sea-level opening across the Isthmus of Panama. Jeffries suggested the following objectives:

"(a) The projects relating to 49 and 25 should be prosecuted by the government, no matter when the war ends, to a point sufficient for military appraisal.

(b) The development of the nucleonics industry by private enterprise should be encouraged. The military by-products of the industrial developments should be made available to the government, and the use of government information patents should be made available to industry so far as the military situation may permit.

(c) A suitable agency, with both government and non-government representatives, should be established to guide and coordinate such nucleonics activities as may affect the military or other interests of the government."

After discussion of Jeffries' presentation, Compton expressed the Council's desire that plans for a permanent organization be pressed as rapidly and as specifically as possible. He asked Jeffries to organize a committee within the Project for preparation of a prospectus. [9, pg.149-150]

1944-07-21 Hogness asked Compton for help in obtaining information on naval power units (*e.g.*, submarines, destroyers, cruisers, and battleships) for the informal Power Committee chaired by Allison. He said that Allison has asked him to obtain such information as it would be useful for those Lab people who are now giving thought and attention to the development of new sources of power, such as nuclear. [9, pg.155]

1944-07-25 Hogness also states his belief that we are equipped with personnel and facilities to carry out laboratory work on the extraction of U233, the development work and large-scale extraction to be done at Clinton. [9, pg.159]

1944-07-26 At 8:00 a.m. I held a meeting of the Council of my section in my office...I noted that since the purification sub-section is being disbanded, the following reorganization is taking place: (c) Katzin has been placed in charge of the U233 work. His rank is that of Assistant Section Chief, and he will report directly to me. [9, pg.161]

1944-07-28 Allison sent a memo to Compton transmitting a review of the Met Lab activities in the four Divisions: Chemistry, Health, Physics, and Technical, as anticipated for a period beginning about October of this year. The summarizing comments are as follows:

"The Hanford studies, which are the items of highest priority, will be almost entirely laboratory-scale problems on process improvement. We anticipate that work on these problems at semi-works scale will be done at Clinton. Alternate processes for Hanford, such as the promising solvent extraction (hexone) method for plutonium recovery, will be carried to the semiworks state at Chicago, however. Certain studies on the effect of pile radiations on materials contemplated for Hanford, such as the current study of dichromate reduction, will also be of highest priority.

"Probably next highest priority will be accorded studies of the basic chemistry of plutonium and to methods of extraction of 23 studied on a laboratory scale. **The recent abandonment of attempts to reach high purity in our product has, in my opinion, increased somewhat the interest in 23.** This is because we are now committed to only one method of final assembly; and if this should fail, the military usefulness of our product is doubtful. **Under these conditions we would immediately give consideration to the conversion of our product to 23.** Work on extraction of a few milligrams of 23 to be tested at the Argonne Laboratory will probably carry over into the post-October period. [9, pg.163]

"In this same priority would be the preparation and chemical isolation (if possible) of Pu240 and U236. We now know that the former will be a constituent in our product, and the latter will be produced in our piles and in the final gadget. These atomic species in a pure state should be presented to the physicists for study as soon as possible. [9, pg.164]

1944-07-31 Stan Thompson returned to Chicago at 8:20 a.m. from his trip to Montreal, Canada. He told me that he spent considerable time with Goldschmidt and Gueron discussing the work being carried on at the Montreal laboratory. He described to me the procedure used there to extract the U233 from the two irradiated thorium carbonate slugs received from Clinton early in July—an ether extraction method similar to that used by Stoughton at Oak Ridge. They obtained about 8 to 9 mg total uranium which may or may not be fairly pure U233—analyses are not completed.

Although Thompson was not permitted to discuss plutonium work, they did volunteer information about what they have accomplished. [9, pg.165]

Thompson is of the opinion that it would aid their microchemical work considerably if our microchemists could spend a few days at the Montreal Laboratory. He also suggested we send them some micro lusteroid cones. He was told of plans for building a pile at Petawawa, which is about 110 miles from Ottawa. This pile will be operated with approximately 13 tons of heavy water and about 8 tons of uranium, the structure will be built of aluminum, and the uranium slugs inserted in the channels so that water can flow between the wall of the channel and the uranium cans. There will be a 1/8" aluminum jacket on the cans. There will be extra holes in the pile for inserting thorium to produce U233, which is central to

their program. The quantity of thorium and the form in which it is to be used have not been decided yet, although there is considerable talk about using thorium metal. [9, pg.166]

1944-08-01 James Franck sent a letter to Vannevar Bush, Carnegie Institution in Washington, D.C., pointing out the general feeling that exists throughout the Project concerning the need for a peacetime atomic program that will continue to supply the research necessary for both military and technical applications in the postwar period. Planning is important and may require at least a year's effort after the war ends. Franck points out that one of the major difficulties we face is the irreversible scattering of key men during the present production and trouble-shooting period. Should the war suddenly stop, there is danger of wholesale dissolution of projects and personnel before intelligent long-range plans will have been formulated. Franck proposes that an interim organization under the Government be created to act as a buffer between the wartime military program and a new peacetime organization with long-range goals. [9, pg.168]

1944-08-02 After regular hours, about 5:45 p.m., Katzin, Studier, and Hagemann began the job of extracting U233 from two cans of thorium carbonate that were irradiated for seven months in the Clinton pile. They fished the first can out of its shipping container and deposited it in the dissolver unit containing nitric acid. Following dissolution, ammonium nitrate will be added to the thorium nitrate solution to a total nitrate normality of 8-9 N. The uranium will then be extracted from this solution using ether extraction procedures.

The Purification Sub-section has been liquidated. A solvent extraction group, with Lawroski as its group leader, has been organized under Katzin to work on U233. [9, pg.169]

1944-08-09 Katzin and his co-workers have nearly completed the extraction of U233 from two cans of Clinton pile-irradiated thorium carbonate. The extraction run was started one week ago and has proceeded very well. Adequate planning and careful handling of materials have kept radiation exposures and contamination problems to a minimum. (This fact was given special notice by John Rose in last Saturday's Health Division Liaison Committee meeting.) It seems likely that at least 5 mg of U233 will be obtained from the extraction run.

In view of this success and since we would like to have larger quantities of U233 for experimentation, I sent a memo to Hogness requesting four more cans of irradiated thorium carbonate similar to the type previously received from Clinton. In addition I requested the irradiation of five cans of thorium carbonate material that has been completely purified of natural uranium by Katzin. This material is being canned and should be ready for shipment to Clinton within a few days. I asked Hogness to arrange for these cans to be irradiated for a period of several months in a high flux position within the Clinton pile. When processed, this material should provide us with U233 that is very free from natural uranium impurities. [9, pg.181]

An Executive Session of the Project Council was held this morning... Compton in his "State of the Nation" opening gave some impressions gained during his recent trip to Hanford and Los Alamos.

Compton then said that Groves would like a reviewing committee survey of new ideas for improved piles. These survey results might determine the future course of our Project. Compton asked if ideas could be ready by October 1. Spedding suggested there should be an adequate group of individuals whose primary effort would be to develop a clear picture of the future of nucleonics. Doan added that the picture of future possibilities would not be complete unless all fields of technical knowledge are covered, and Compton replied that the committee under Jeffries is preparing to develop such a picture.

Allison stated that beryllium, bismuth, and thorium are materials which will be important to our future. Whitaker said he would like some thorium metal to replace the thorium carbonate in the Clinton pile. Szilard added that if poisoning of the Hanford piles were done with about one ton of thorium metal, this would be an excellent source for larger amounts of U233. Allison proposed to authorize Spedding to produce a definite amount of thorium.

Whitaker announced that with the installation of two new fans for the air-cooled graphite pile at Clinton, the power level has been increased to three times the original design power level. One fan, however, was seriously damaged last night, and the production power level is now down by about 25% to 30%. The results in the Clinton extraction plant indicate the Hanford plants will operate with yields above 80%. [9, pg.184-185]

1944-08-12 Katzin, Studier, and Hagemann completed the extraction and purification of U233 from the two cans of thorium carbonate that were irradiated for about seven months at Clinton. **Preliminary indications are that about 6 mg of U233 have been isolated from the two cans.** [9, pg.189]

1944-08-15 A revised organization chart and several recommended organizational changes for my section were sent by Manning to Hogness. The number of sub-sections is reduced to two—namely, the Separation Processes Subsection and the Basic Chemistry and Services Subsection. The group doing work on U233 is not included in either of the subsections. [9, pg.191]

Compton wrote to James B. Conant in Washington about the transfer of nucleonics to postwar conditions. (In a footnote, Compton explains that the word "nucleonics" has recently become widely used throughout the Project to designate the field of research, development, and application of atomic nuclear reactions. It, according to Compton, comes from the word "nucleon" which is the name for the elemental particles in the atomic nucleus.)

Compton believes the most important applications of nucleonics will be for some time in the area of military weapons; however, other applications will grow rapidly such as radioactive material for medicine, agriculture, and scientific purposes, and the use of atomic power especially for naval vessels and perhaps for heating of cities. He suggests that the U.S. Engineers may be the best group to continue its Project management role into the postwar period. But, in order to provide for coordination of the various aspects of nucleonic work throughout the nation, he suggests that a Committee (or an individual) reporting directly to the President, be given the responsibility for overall supervision. The Committee would

function in a way similar to the Military Policy Committee and would see that appropriate controls are put into effect for regulating nucleonic work.

Compton suggests an immediate program to encourage universities, industries, and private laboratories to undertake their own activities in nucleonic research. This would include the transfer of technical information from the Project to the public making available special materials. Authorization to use the tools built under government contract for the production of tracers and other useful materials would greatly aid medical and scientific developments. Public education on the significance of nucleonics is urgently needed in order to prepare for the postwar adjustment.

Regarding the future of the Metallurgical Project, Compton suggests a sharp distinction be made between continuing military work including continued support of the programs at Sites W and Y and the development of nuclear weapons and non-military work such as nuclear physics and properties of neutrons; fundamental chemistry of products and by-products; long-time instrument developments; the preparation, properties, and application of radioisotope tracer materials; scientific, medical, and industrial applications of radioactive materials. Compton says it would appear appropriate to encourage the University of Chicago to see that (non-military) scientific use be made of the tools now in its charge. Compton concludes his letter by saying that the problems associated with the interim period and postwar adjustments, especially as applicable to the Metallurgical Project, will be the main subject for discussion at the August 21 Advisory Committee Meeting. [9, pg.193-194]

1944-08-16 Katzin calculated, using alpha-particle counts by Hufford, that the specific activity of the U233 extracted from the irradiated thorium carbonate is 19,800 disintegrations per minute per microgram. This figure, however, is subject to revision, as the extent of natural uranium impurity is not yet known. [9, pg.197]

Allison sent a memo to Franck referring to statements made at the June 20 Project Information Meeting that, (a) a pile with solid moderator will inevitably go bad, and (b) it is improbable that operation with graphite as a moderator can go on for two years. G. D. Graves of du Pont has questioned these statements and pointed out to Allison at a recent meeting that such statements, if left to stand on the record, could seriously embarrass the Project unless they are extremely well-founded in fact. Allison suggested that if similar considerations come up at the Project Information Meeting on August 22 that the record should be made clear as to exactly how large an extrapolation from our current limited knowledge is involved. [9, pg.199]

1944-08-17 Leo Szilard wrote to Vannevar Bush in Washington, enclosing the text of a personal letter he wishes to send to Lord Cherwell of the British Cabinet in England, whom he knows well. The letter is for the purpose of bringing to the very influential Cherwell's attention some of the possible ways Germany might produce atomic weapons. Szilard says in his letter to Bush,

> The possible use of atomic bombs by the Germans is perhaps the only factor that

> may reverse the present favorable trend of the war in Europe. **It would therefore seem very important to find the industrial installations in Germany which are likely to be used for this purpose and to destroy them.** I believe that the British efforts in this direction would be greatly intensified if Cherwell took a personal interest in them.

Szilard's letter to Lord Cherwell is six pages long. In one paragraph he states,

> Private communications originating from Switzerland which reached me two years ago indicated that the Germans knew by the middle of 1942 how to make a chain reaction go and that Heisenberg, who was in charge of that work had some conspicuous successes along this or the other parallel line of work during that year and consequently was put in full charge of all the work (made director of the Kaiser-Wilhelm Institute for Physics) late in 1942. This was about the time of Stalingrad after which the Germans must have realized that they may have to win this war by other than ordinary methods. **Unless I completely misjudge the psychology of the Germans, they must have gone full scale into this work soon after Stalingrad at the latest.**

Further on in his letter Szilard remarks,

> I am rather convinced that a properly organized effort of the British Intelligence will lead to the discovery of industrial installations for the manufacture of the relevant materials in Germany but when the location of the German factories is discovered, it may be found that we would have to pay an exceedingly high price for their destruction by large-scale parachute invasion or other such methods. Some of us might think that these factories have to be destroyed practically at any price but in order that you should be in a position to advise the War Cabinet how far to go in this respect you ought to have first-hand knowledge of the action radius within which life will be destroyed if a small atomic bomb is detonated above a city. Clearly you are the only member of the Cabinet who can have convictions based on his own computations rather than on official 'reports' or other forms of 'hearsay'. If you check the calculations of others or make calculations of your own you will have a firm conviction of your own and will be able to arrive at a balanced recommendation and also be able to assist the War Cabinet in reaching what may be a very difficult decision. [9, pg.199-200]

1944-08-19 Franck answered Allison's letter of last Wednesday about the statements made at the Project Information Meeting of June 20 regarding the expected short operating life of a graphite-moderated pile. Franck points out that the statements made at the meeting should not embarrass the Project as Graves from du Pont implies they might, in that it was foreseen by Wigner right from the Project's beginning that a risk would be involved in the use of graphite as a moderator. So far the experience with the Clinton pile shows that the risks are

not so great that a pile cannot be run long enough to turn out amounts of plutonium exceedingly useful for the war effort. Franck points out, however, that it is absolutely necessary to monitor the change in properties of the graphite in piles at Hanford during their operation. It is impossible to predict the degree of deterioration of pile graphite based only on an equivalent of three days of operation of the Clinton pile at Hanford radiation levels. [9, pg.202]

1944-08-21 Katzin summarized for me the work his special group has completed on the extraction of U233 from the two cans of thorium "carbonate" that were irradiated for about seven months in the Clinton pile. Each can contained about 164 grams of thorium and a small undetermined amount of natural uranium. The initial extraction was quite successful. Approximately 6.5 mg of U233 were extracted based on the half-life 120,000 years for U233. Initial measurements (subject to correction) give a specific activity of 20×10^6 d/m per mg, which corresponds to a half-life of 170,000 years. Gofman, Stoughton, and I estimated the half-life of U233 to be 120,000 years in our original work on U233. Katzin reports the range of U233 alpha particles, based on absorber foil measurements, to be 3.35-3.40 cm in standard air. A more precise determination, however, is expected using Jaffey's differential alpha-particle range chamber. Preliminary calculations using data from thermal neutron fission cross section measurements give a U233/U235 cross section ratio of about 1.3, based on the value 120,000 years as the half-life for U233. The data are consistent with an isotopic purity of 70% for the U233. **A more accurate value for the ratio of neutron fission cross sections, U233/U235 will be obtainable when a more precise half-life value determination is made following a mass spectrographic analysis of the isotopic composition of the sample.** [9, pg.205]

1944-08-22 The successful separation of over 6 mg of U233 from Clinton-irradiated thorium, described in Katzin's memo to me yesterday, was reported. I said that the question the half-life for U233 is still not settled but may be as high as 170,000 years instead of our earlier determination of 120,000 years. W.C. Johnson said that initial analyses show that there is about 0.05 ppm natural uranium impurity in the starting thorium "carbonate" leading about 10% natural uranium in the U233. (If this is correct, the half-life may be about 153,000 years.) [9, pg.209]

Spedding reported on the work at Ames relating to the production of cesium and thorium metal. I asked how much thorium metal he is planning to make. He replied that it could be produced on a large scale, but the quantity must be decided by Washington. [9, pg.210]

1944-09-06 I attended the evening meeting of the Separations Process Subsection of Section C-I at 7:45 p.m. In Room 209, Eckhart Hall. Hagemann told us about the extraction of U233 from the third and fourth cans of thorium carbonate irradiated by neutrons for several months in the Clinton pile. About 8.5 mg of U233 were obtained. Specific activity determinations carried out by Hufford by counting a weighed sample of oxide gave 23,000 d/min/microgram as the best value for pure U233. I mentioned that the Montreal group has been obtaining much

lower values than this—perhaps impurities still present in their U233 account for these low values.

Hellman gave an account of the extraction of uranium and thorium into ether from solutions of varying NH4NO_3, Th(NO_3)4, and HNO_3 concentrations. Hyde reviewed the extraction of uranium and thorium by hexone; he has found that thorium decay products are extracted by hexone to a greater extent than by ether. Reinhardt spoke on the effect of lanthanum, calcium, and ammonium nitrates as salting agents for extraction of plutonium into ether and hexone. He has found that their effectiveness in both solvents is in the order named. Van Winkle talked about the work being carried out on isolation of natural Pa231 and Io230 from residues obtained from commercial uranium production. He gave a summary of the existing knowledge of protactinium and ionium chemistry. Separation and isolation of protactinium and ionium by means of UF_4, which carries ionium but not protactinium, will be the next step in the investigations. [9, pg.228-229]

1944-09-09 I received a copy of a September 8 memo from Dempster to Stearns giving the results of a mass spectrographic assay to determine the ratio of the isotope 233 to the isotope 238 in a sample of the 7.2 mg of uranium isolated from the thorium carbonate irradiated by neutrons for several months in the Clinton pile. **His data indicate the percentage of U233 to be 85$\pm$1.5% of the total uranium.** [9, pg.232]

1944-09-13 Today the first uranium slugs are being loaded into the first production reactor at Hanford—the 100E pile. Fermi is there to oversee and direct the operation. [9, pg.235]

1944-09-18 Also on Thursday, Allison sent a memo to Mulliken of the chronology of the Project. Thirty-six highlights are identified from January 1939 (chemical evidence for fission by Hahn and Straussman announced by Bohr) to September 12, 1944 (loading of metal into first production pile at Hanford). Other highlights include:

- March 28, 1941. California group demonstrates that 49 is fissionable at thermal energies.
- February-March, 1944. Milligram and gram amounts of 49 from Site X are available for experimentation.
- July, 1944. Spontaneous fission of plutonium-240 discovered at Site Y, and purification program abandoned for plutonium. Ceiling placed on Met Lab personnel. [9, pg.238-239]

I received a memo from Davidson giving a summary of existing data on the thermodynamic properties of oxides and fluorides of uranium and plutonium from uranium—again results are promising.

Burton reported work of Section C-II on the problem of sudden release of energy accumulated in the pile of graphite by displacement of carbon atoms in the crystal lattice. The possibility of annealing was mentioned. Sugarman reported on fission product work in Section C-III and indicated that the distribution curve of fission products for the heavy group from

neutron-irradiated plutonium appears to be identical to that from U235 except for distinctly higher yields at the extreme heavy-weight end of the curve. [9, pg.241-242]

1944-09-20 I sent a memo to Hogness requesting that four more cans of the irradiated thorium carbonate be shipped to us from Clinton as soon as possible. The U233 to be extracted from it is needed mainly for our own experiments; the previously extracted U233 is needed for use at Site Y and by Anderson at Argonne. [9, pg.248]

1944-09-28 In Montreal. W. W. Watson, the Metallurgical Project representative stationed in Montreal, and I visited the Montreal Project. Most of my discussions were with the chemists—Paneth, E. W. R. Steacie, Bert Goldschmidt, and J. Gueron were present in practically all the discussion. The more important topics I covered with them today are:

- Thorium metal vs. carbonate for irradiation (Sutton and Bewick).
- Separation of U233 or Pa233 by adsorption (L. G. Cook, W. E. Grummitt, and G. Wilkinson).
- Solvent extraction (Reid, L. Yaffe, Fitch, Russell, A. G. W. Cameron, and Mungen).
- Organic complexes (A. G. Maddock, A. C. English, and Musgrave).
- Fluorine method (Maddock).
- Purification of U233 (Yaffe, English, and Cruikshank).

In the afternoon I gave a talk on the hazards connected with the alpha-particle radiation from U233 and the gamma-radiation from Pa233 and made a few suggestions on how these might be minimized. In the same talk I discussed the latest measurements in Chicago on the specific activity, half-life, alpha-particle range, etc., of U233. Also, I gave some speculations as to a probable decay chain for U233 (the 4n+1 series) and also some speculations on the electronic structure of elements 89-95, inclusive (*i.e.*, my actinide concept). [9, pg.257-258]

1944-09-29 In Montreal. I held further discussions with members of the Montreal Project, covering the following:

- Extraction of protactinium from ores (H. G. Heal, Cook, and F. Morgan).
- Fission products of U233 (Yaffe, Wilkinson, and Grummitt).
- Barium branching ratio in U233 fission (Yaffe, Wilkinson, and Grummitt).
- Rare gas experiments - measurement of helium, krypton, and xenon (W. J. Arrol).
- Fission chambers (Maddock, Miller, and B. Pontecorvo).

At 4:30 p.m. Watson and I met to review decisions with Arrol, Goldschmidt, Gueron, Huffman, Maddock, Paneth, and Steacie. The more significant items discussed were:

Metal versus carbonate. I indicated that, for the immediate task of making one gram of U233 per day with material around the outside of the pile, everything is in favor of thorium carbonate.

Extraction of Pa233 versus U233. It was agreed the product should be separated as U233—no point in separating as Pa233.

Elimination of protactinium by adsorption or precipitation. Direct separation by adsorption is not practical. The precipitation method involving MnO_2 is fairly certain to work.

Solvent extraction. This is clearly the best method for extracting U233 from thorium carbonate. Ether is probably the best solvent so far developed, but work on other solvents should proceed.

Purification of U233. The details of how to purify U233 extremely well were discussed. As soon as there is a sample of quite pure U233, the Montreal group wants to measure the decay period. As soon as there is a sample absolutely free from beta particles, the group will launch an investigation to find the disintegration products and look for half-life periods and radiations of these products.

Extraction of protactinium from ore and determination of valences. Montreal chemists have been working with the most insoluble residues of Canadian uranium ore in search of protactinium. I suggested the more soluble carbonate fraction might better be used. I pointed out it will be especially interesting to look for an oxidation state of 4 and possibly state 3 in the protactinium.

Fission products of U233. Montreal chemists would like to have the 10 mg of the U233 they sent to Anderson at Chicago retained in Chicago and irradiated by neutrons in one of the piles for at least a month. Then it should be sent to Montreal for a study of the fission products of U233. I agreed to prepare the sample for irradiation in a convenient form.

Fission chambers. The Montreal group has measured the fission cross section of U233 by using UF_6 and BF gases in their fission chambers. The spontaneous fission rate of U233 has been measured by Pontecorvo at equal to or less than 3 neutrons per minute per gram, this value obtained even with U238 impurity. A discussion was held on the difference between the Montreal and Chicago measurements of the fission cross section of U235, The Montreal value is 4.6 x 10-24 cm2 and the Chicago value is 3.9 x 10-24 cm2. There seems to be no explanation for the discrepancy.

In a general discussion it was decided that for the present there should be no division of work between Montreal and Chicago. Montreal will carry on investigations of all problems connected with U233 production while we at Chicago will consider only those problems which interest us. [9, pg.258-259]

1944-09-30 I visited Watson's office and prepared notes on my discussions with members of the Montreal Project on Thursday and Friday.

I boarded a train for Toronto and Chicago. [9, pg.259]

1944-10-02 Last Wednesday morning (9/27), a few minutes after midnight, marked the first power run of a Hanford chain-reacting pile. Two weeks earlier, Fermi had brought the production pile (100B) into criticality for the first time but without water cooling. During this midnight run of about two hours, the power level exceeded anything that had ever been attained in a pile before, either at Oak Ridge or Argonne. But following the two-hour run, to everyone's consternation, the power level began to decline slowly and steadily of its own account. By Wednesday evening the chain-reaction ceased completely. Then early the next morning the chain-reaction started up spontaneously, and by evening the power was up to its former high level. Soon the pile began shutting itself down again. **Many hypotheses were advanced, the most acceptable being that the pile had generated some kind of poison to reduce the reactivity. If it were indeed a poison, then it would have to have a half-life of around 9.7 hours to fit the pile's behavior pattern. The fission product Xe135, with a half-life of 9.4 hours, was thus suspected.** Zinn at Argonne and Doan at Clinton Laboratories were immediately notified and asked to run experiments to determine if a Xe135 effect on the operation of the piles there could be detected. Zinn soon confirmed that the production of Xe135 in the chain-reaction does have a poisoning effect. Now the question arises, must the Hanford piles be modified if they are to produce Pu239 on a grand scale? [9, pg.260]

Approximately 8 mg of U233 have been recovered from thorium carbonate irradiated about seven months in the Clinton pile. The U233 was found to be 85% pure upon analysis with a mass spectrograph. A corrected half-life determination on the U233 sample gives a figure of 138,000 years. A corrected cross section for U233 as compared with U235 (23:25) gives a ratio of 0.96 for thermal (paraffin-moderated) neutron fission. The +6 oxidation state of neptunium has been identified by means of sodium neptunyl acetate. [9, pg.264]

"23 Group", Group 9: Uranium-233 Separation. L. I. Katzin, Assistant Section chief in charge of Group 9's U-233 work. [9, pg.265]

Farrington Daniels sent Mulliken suggestions for several possible applications of "nucleonics," including high-temperature radiation chemistry, process radiation, radioluminescence, and weather modification applications. He suggests the development of a gas-cooled pile operating at temperatures up to 2000°C to obtain good thermodynamic efficiency. The high temperature and heat produced might be used to replace electric furnaces and might find other applications for producing chemicals such as calcium carbide, nitric oxide, phosphorus, and possibly even alcohols from carbon monoxide to provide automobile fuel. The intense radiation available in a pile would make possible the consideration of photochemical reactions on a large scale—such as the production of resinous materials from petroleum products and possibly even carbohydrates. [9, pg.268]

The ionization from pile-produced materials might be used with an electrostatic precipitator to remove dust and smoke. Daniels also suggests that intense radiation in the form of radioactive gases or particulates could be dispersed in the air to dispel fog or possibly even to induce rainfall. [9, pg.269]

1944-10-03 In addition to this meeting, I attended the Project Council Information Meeting on

Physics which met in Room 209, Eckhart Hall. Zinn described the 105B Hanford pile start-up problems. The initial 9 MW start-up power level at 0.00088 excess k fell off to 3.7 MW because of poisoning by a 9.4-hour Xe135 (the daughter of 6.6-hour I135). With no slug or pile modifications, this poisoning effect will limit the full operating power of the Hanford pile to 216 MW. A modification providing 1.85% excess k would completely override the poisoning effect by destroying the Xe135 by neutron absorption and should thus permit pile operation at any power level. The poisoning effect has not yet been oberved at Clinton It may be caused by resonance in the thermal neutron energy region.

H. L. Anderson reported on experiments concerning the thermal neutron (2200 meters per second) absorption cross section for plutonium in plutonium nitrate dissolved in deuterium nitrate, heavy water solutions, that was determined to be 1150 barns $\pm$ 3%. This result is in fair agreement with the value of 1050 barns obtained at Los Alamos. The fission cross section is 773 barns (alpha = 0.49). The resonance cross section is found to peak at 6730 $\pm$ 240 barns. **Anderson also gave values of 650 barns and 535 barns, respectively, for the thermal neutron absorption and fission cross sections of U233.** The cross section for U233 is higher than the cross section for U235, which in turn is higher than for Pu239, over the neutron energy range from 0.5 to 2900 ev.

"Chemical Research - Extraction and Properties of U233, Report for Period Ending September 10, 1944," (CS-2165), was issued today. A summary of the contents is as follows. Studier, Hagemann, and Katzin have extracted 7.2 mg of U233 from two cans of thorium carbonate irradiated seven months at Clinton. The extracted material is 85% pure U233 with the rest being natural uranium. Four cans are undergoing extraction by the same ether-extraction method used for the first two cans.

Katzin, in cooperation with Dempster, Lapp, Crawford, Hufford, Jaffey, and Weissbourd, has determined that the specific activity of pure U233 is close to 23,300 disintegrations per minute per microgram, corresponding to a half-life of 146,000 years. The range of alpha particles for U233 has been determined as 3.38 $\pm$ 0.01 cm in air. The fission cross section for cadmium-absorbable neutrons slowed in paraffin is 1.1 times that of U235. [9, pg.271]

1944-10-04 The Project Council Policy Meeting for October was held today at 9:00 a.m. in Room 209, Eckhart Hall. In attendance were Allison, Bartky, C. M. Cooper, Daniels, Dempster, Franck, Hamilton, Hogness, Howe, Jeffries, Mulliken, A. V. Peterson, Spedding, Stearns, Stone, Szilard, Vernon, C. J. Watson, W. W. Watson, Whitaker, Wigner, Wirth, and Zinn. Allison announced that Compton flew to Hanford yesterday and that up until yesterday afternoon the Hanford 105B pile has operated twelve hours at 10 MW. The xenon-poisoning phenomenon was discussed. Wigner estimated that the maximum power that could be attained at full Hanford pile loading (2000 tubes) for 1.2%, 1.8%, and 2.25% excess k would be 200 MW, 400 MW, and 800 MW, respectively. It is possible that additional excess k can be had by making changes in slug canning, removing thimbles, etc.

Hogness said that Site Y would like the top people in Sugarman's Section III, but Allison recommended maintaining the present status until the pile poisoning problem has been solved.

Allison said the Met Lab has lost about 175 technical people over the past two months with Chemistry being the big loser. Clinton has lost about 150 technical employees, primarily to Hanford.

Questions concerning cooperation with England ("Evergreen") were raised by Allison, in particular, whether or not the U.S.A. should give England information on the xenon poisoning phenomenon. W. W. Watson agreed there are problems and cited our specific directions to exchange all information on U233 chemistry but none on Pu239 chemistry, even though the "Evergreen" people obviously understand the relation of the U233 to the Pu239 chemistry very well. He said all evidence points to England pushing vigorously ahead after the war. **Stearns noted that if England does emphasize nucleonics after the war and we do not, the U.S.A. will be at a serious disadvantage. He raised the question of whether we should wait for requests or should volunteer information to England. Both Peterson and Zinn spoke out in favor of volunteering needed information. Zinn said England will have the finest research pile in the world within a few years and we should keep on a good cooperative basis in order to obtain information on their results.** Cooper suggested that a copy of the directive covering cooperation with "Evergreen" be inserted in the minutes of the Project Council Policy Meeting minutes. However, because of the wide circulation of the minutes, it was later decided to issue the directive, including further restrictions by McKinley, on a smaller selected distribution list. [9, pg.273]

1944-10-09 I wrote to Goldschmidt in Canada announcing that the report on our recent U233 work has been issued under Report No. CC-2165. I make several comments on the reported work, in addition to the points I made in my talk during my visit to the Montreal Project at the end of last month. I quote our more accurate value for the half-life of U233, 146,000 years, based on our specific activity of 23,300 d/min/pg. I mention the as-yet unidentified insoluble residue we have observed when dissolving thorium metal in hydrochloric acid in light of similar experiments performed at Montreal and described by Goldschmidt during my visit there. [9, pg.276]

1944-10-10 Charles Cooper presented a discussion on "New Goals for the Metallurgical Project" at the meeting of the Tolman Postwar Policy Committee today. Cooper points to the possibility that our job may soon, if not already, be completed with regard to assuring successful W operation. He asks, "with this prospect before us, is it not our immediate and major responsibility to obtain agreement upon a continuing course of research which must be pressed if the United States is to maintain the lead in the field which we presume it now possesses?" Cooper summarizes the current Project status as follows:

> By some two years of great effort under emergency conditions we have arrived at a situation which may be summarized thus. A process has been developed which very certainly will produce quantities of 49 adequate for experimental purposes and probably sufficient to meet situations immediately foreseen. The present process is very complicated and unduly expensive, both as to manpower requirements and the materials which are involved. **The present approach is not adaptable to**

generation of power nor is it designed to manufacture or recover effectively possible byproducts of operation. The product will be contaminated with undesired isotopes which could possibly be eliminated if other methods, as yet only sketchily developed, were to be employed. We cannot be entirely certain until final trials have been made that the product will be as useful as anticipated, and it may well develop that other elements or isotopes will turn out in the end to be much more useful than the one we have prepared to manufacture.

In order that our country may be as well prepared as possible, it would appear from the above that research should be pressed along the following lines.

(1) All possible effort should be made to discover what material and isotope is most promising for our present ultimate use.

(2) As soon as point (1) can be settled, the development of adequate manufacturing facilities for the chosen substances should proceed.

(3) While the developments indicated under (1) and (2) above are in progress, studies aimed at improving the country's position with respect to production of the present isotope and studies aimed at the production of power and of reaction by-products should be vigorously proposed.

Cooper points out that the development of usable electrical power from nuclear reactions "poses a problem much more difficult technically than the one which we have had to solve in connection with our present manufacturing objectives." Many of the problem areas are identified such as high temperatures, corrosion resistance, the need for new materials and alloys for fuel fabrication, structural purposes, and shielding. A wide variety of engineering, scientific, and manufacturing organizations will be required. The Met Lab has many of the facilities and scientific and engineering personnel required, but Cooper points out it is not possible to attract or even hold able persons in the Project because of the future being so uncertain. He says unless the Project "can be given a greater degree of certainty by a new directive indicating clearly new objectives, it will hardly be possible to maintain even our present working organization much beyond the first of the year." [9, pg.277-278]

1944-10-12 Work of U233 Group

Extraction of U233. Four more cans of carbonate have been extracted (in addition to the first two). There are approximately 4 mg of U233 per can. (Hagemann, Studier)

Survey of Solvents for Extraction of U233. (Hyde, Wolf)

Methods of Concentration of U233 Following Extraction. (Hellman and Wolf)

Determination of Physical Constants for U233. The latest values of the specific activity and corresponding half-life of pure U233 are 23,300 disintegrations per minute per microgram and 146,000 years. The range of the alpha particles is 3.38 $\pm$0.01 cm at standard conditions. The fission cross section for neutrons slowed by paraffin and absorbable by cadmium is 1.1 times that for U235. Checks on material from the Montreal Laboratory corroborate the half-life determination. (Instruments Group)

Methods of Dissolving Thorium Metal. (Katzin)

Analysis of Ore Samples for Protactinium. Isolation of Milligram Amounts of Pa231 and Ionium. First steps toward isolation have been taken. (Van Winkle, Sedlet) [9, pg.285]

1944-10-13 Technical Council Members received a memo from Stearns enclosing a list of rules for the interchange of information between the Evergreen Area (the Canadian-British group at Montreal) and the U.S.A. Project groups. Three of these rules of concern to me are the following: (a) **"Information connected with the transformation of thorium to element 23 and the separation and measurement of the physical and chemical properties of thorium and 23 will be freely interchanged."** (b) "Information necessary for the guarding of the health of the operators at the Montreal Plant will be exchanged. Information concerning the toxic effects of either 49 or fission products will not be included in this exchange." and (c) "Information concerning the chemistry of element 49, the method of separating 49 (including all engineering details), and the purification together with the chemistry of the fission products will not be transmitted until further instructions are issued." [9, pg.286]

1944-10-16 Report MUC-GTS-1068 reviews the investigations conducted under Contract OEMsr-206 related to measurement of the fission properties of U233, Np237, and Np239. The discovery of U233 and measurements of its radioactive properties and its slow neutron fission are described in Reports A-153 and A-192. The discovery of Np237 and the demonstration that it does not undergo fission with slow neutrons is covered in Report A-151. Contemplated work on the fission properties of Np239 was never done. The work on the preparation of U234 through the extraction of UX1 (reported in A-172), the search for spontaneous fission in U234 (reported in A-268), and the measurement of the fast neutron fission cross sections of Pu239 and U233 (reported in A-269) were also done under Contract OEMsr-206. [9, pg.288]

1944-10-17 I presented the status of our U233, protactinium, thorium, and neptunium work and future plans. The growth of Pa233 by alpha-particle decay from Np237 has been observed and is being followed, I said. The U233-Th229 decay chain is being followed. I reported that the structure of NpO_2 has been identified with Zachariasen's help and that neptunium in the +6 state is being studied in the form of NaNpAc3. Np(III) cannot be extracted with hexone and in this respect is analogous to Pu(III). I said that intensive work has been started on the use of dibutyl carbitol for the extraction of thorium and uranium. A black residue remains after dissolving thorium metal (from Ames) in HCl. X-ray analysis shows the residue to consist of about two-thirds ThO_2 and the remainder thorium nitrate. [9, pg.292]

Sugarman, representing Section C-III, talked about the radioactive poisoning effect discovered in the Hanford pile when the pile was first brought up to a power level of 9 MW. **The poisoning effect is caused by short-lived Te135 which decays to 6.6-hour iodine and eventually into 9.4-hour Xe135; the latter probably has a high thermal neutron absorption cross section.** Xenon-135 normally decays to a 20-30-year cesium that becomes stable barium. The yield of this fission chain is reported to be about 4.3%.

Johnson introduced the speakers from Clinton Laboratories. Elliott spoke about the xenon poisoning problem also. His measurement on the half-life of I135 gives 6.9 hours instead of 6.6 hours. The fission yield was found to be 5.7% instead of 4.3% for the chain beginning with tellurium. Sugarman suggested the resulting difference may be due to a difference in counting techniques. Problems concerning the accuracy of the physicists' neutron flux values were also discussed. Elliott also described experiments with neutron-bombarded LiF to produce tritium. So far only helium has been found.

English described three problems being studied, two concerning the Hanford process and one concerning U233. The two problems in the extraction step are to study losses caused by complex formation and to study the action of interfering agents such as hydrazine. Nitrite ($NaNO_2$) is not a good reducing agent for the Hanford process especially if much Pu(VI) is present in the process solution. Oxalic acid seems fairly good. English said $NaNO_2$ seems to kill the effects of hydrazine: this is probably due to the $NaNO_2$ acting as an oxidizing agent. The work at Clinton on U233 is progressing. English reported results of the dissolution of thorium metal with HNO_3 and with HF in concentrated HNO_3. Some residue remains and probably contains silicon. I cautioned English that when one uses HF, a complex will be formed that will interfere with the extraction process. English said HCl will also dissolve the thorium but this is undesirable and causes increased corrosion problems. With regard to the residue, Zachariasen asked Spedding how the thorium was melted. Spedding said beryllium oxide crucibles were used (not nitride as Zachariasen had earlier suspected). [9, pg.293]

1944-10-18 Wigner said that the immediate progress at Los Alamos is not currently limited by a lack of understanding about the theoretical physics of the bomb. Compton remarked that since the loss of a large fraction of our Met Lab experimental physicists to Site Y will so greatly weaken our ability to go forward with new pile studies, effective utilization of theoretical physicists in Chicago may not be possible. It would therefore seem desirable to transfer the theoretical physicists to Site Y also. Franck and Mulliken felt differently about the matter. Franck pointed out that Chicago needs a strong theoretical group for consultation, and Mulliken said such a transfer would seriously hurt the work on the preparation of Project records.

Compton reviewed the situation at Hanford and said that a week-long operation of the 105B pile at about 30 MW showed no new surprises. Xenon production in the pile is still the best explanation as to the cause of the poisoning effect.

Whitaker said the large new fan at Clinton has been repaired and the X-10 pile is operating at 4 MW. He said careful measurements were made of about 7% of the center pile materials irradiated for two months. This material was pushed out and replaced with fresh material. An increase in reactivity was observed that corresponded closely to the amount of xenon calculated to be present in the old materials which had been pushed out. [9, pg.295-296]

1944-10-23 I read a memo Pye wrote to me reporting on his recent trip to Clinton. He states that the amount of material (slugs) being processed in the canyons per run has recently been increased from one-third ton to one-half ton. Processing, using two decontamination cycles

and a crossover cycle prior to isolation, continues to be satisfactory. Eleven runs have been made involving recycling experiments in order to build up the plutonium concentration in the lanthanum fluoride-plutonium solution to a level where the Hanford isolation procedure could be evaluated on a plant scale. The recycling procedure used was to metathesize and dissolve the combined lanthanum fluoride-plutonium precipitates from two runs; precipitate the plutonium as the peroxide; dissolve this in Room D; and with the supernatant added, recycle this entire solution to the lanthanum fluoride-plutonium precipitation step as the lanthanum precipitating agent in 204 Building. The Hanford isolation equipment has been "copied" at Clinton, and two isolation operations have been carried out successfully in 204 Building under conditions which closely approximate Hanford conditions.

Pye reports that Stoughton's group is investigating methods for dissolving thorium and ThO_2 and has determined that a solution of 8 N HNO_3 and 0.3 to 0.5 N HF [or 0.005 N (NH4)2SiF_6] offers promise. Clinton Laboratories' future plans include the recovery of all U233 from the remaining cans of irradiated Th(CO_3)2, the development of a method for obtaining irradiated materials in the future on a continuing basis, and the development of procedures for isolating very pure U233. The isolation procedure contemplated for the latter will consist of immediate ether extraction of the irradiated thorium carbonate to remove uranium and other impurities. The aqueous phase solution will be then aged to permit the growth of U233 from its parent Pa233; the U233 will then be removed in a pure state by a second ether extraction. A continuous ether extraction system is planned for eventual use. [9, pg.304]

1944-11-01 I reviewed recent results on the physical constants of U233 it appears that the fission cross section is about the same as for U235 (550 barns); the total neutron absorption is only around 620 barns, giving a favorably small value for the parameter alpha (0.12). The number of neutrons per fission is 1.07 times the U235 value. The number of neutrons given off per neutron absorbed is 1.13 times the U235 value. **These values are within the range to enable U233 to be made from thorium by a chain reaction on the U233 (*i.e.*, to make breeding possible)—extremely important because it may make it possible to be independent of uranium once a supply of U233 for starting purposes is on hand.** [9, pg.320]

A meeting of the Division Directors took place today. Some of the topics that were considered are the following: (3) Allison states that Compton has issued a directive requesting that the separation of U233 from 40 slugs of thorium carbonate, irradiated at Site X, be done entirely at the Metallurgical Laboratory. Provision for doing this should be made. The remaining slugs on hand at Site X will be irradiated, and fifteen will be required for shipment to Montreal. Allison requested that 25 mg of U233 be set aside for the use of Professor Koch at the University of Illinois. [9, pg.320]

Hogness issued a summary of the manpower distribution in the Chemistry Division (148 total). My section employs 73 men. The three research sub-sections or groups of Section C-I are divided in the following way.

[9, pg.321]

		Number of Men	
		Sept.	Oct.
Albaugh (Site W work, 26 men)	R. Thompson, extraction and decontamination	11	10
	Pye, concentration and isolation	8	5
	Gilbreath, process development	7	6
	Lawroski, solvent extraction	4	4
Cunningham, (Site Y work, 38 men)	Simpson, high vacuum work	11	10
	Hindman, basic chemistry	10	10
	Dawson, recovery	9	8
	Ghiorso, instruments and physical measurements	10	9
	Katzin, 23 work	6	6

1944-11-03 "The 23 separated from the thorium carbonate from the X pile by extraction with ether has shown 85% purity with respect to U238. The half-life has been set at 140,000 years, the alpha-particle range as 3.38 cm in air. Samples have been sent to both Y and Argonne for additional work." [9, pg.323]

1944-11-06 About mid-morning I was visited in my office by W. P. Grove and A. Paneth from the Montreal Project, who are here on a three-day visit. They were accompanied by Captain Chapman of the Chicago Area Office. We first held a general discussion on the radiation hazards connected with U233 work. In answer to Grove's question, I gave the opinion that the tolerance level for U233 in the lungs is something like 10 to 50 micrograms. I showed them a copy of our "mandatory product safety rules" that apply to any alpha-particle emitter. We also discussed precautions in handling beta- and gamma-activity, and I described our "hot" laboratories. I mentioned our system of continuous cleaning of laboratory desks, equipnent, floors, walls, and ceilings and our system of changing to special laboratory shoes upon entering the building. We discussed particularly our rule of handling plutonium only when working with gloves and our rule that plutonium in any appreciable amount at all can be handled only in hoods under forced draft. After this discussion, we went through Rooms 5 and 6 where the U233 work is done and Rooms 34, 35, and 37 where the plutonium recovery work and other work is done. After lunch I again met in my office with the visitors and Captain Chapman. Also present was Dr. Nickson who described in detail our radiation monitoring system

A memorandum on secrecy (MUC-TRH-188) was sent to Compton, signed by 19 people who hold the rank of Division Director or above—leaders of the Met Lab, Clinton Labs, and the Ames, and California Projects. The memorandum gives three reasons for lifting to a certain extent, the Project secrecy restrictions: (1) If the enemy has the weapon and uses it first, the lack of forewarning could cause panic among our Allies. (2) Our unexpected use of

the weapon could have dangerous postwar effects stemming from the sudden, unanticipated conclusion of the war and the lack of postwar plans which take account of the existence of this new weapon. (3) The enemy may be able to create division among the Allies by pointing to the existence of big production plants in which are fabricated weapons which they can contend are not designed for the present war.

The writers/cosigners advocate that a general statement be made to the public concerning the existence of a new weapon and stating that work is going on in this country for manufacturing it and that it is bound to affect the relations of the nations in the future. They further advocate that, since these considerations may transcend those of the military, active steps should be taken to have them laid before the highest authorities of the land and before all those who will advise on the specific conditions of postwar control. [9, pg.325]

1944-11-07 At 2:00 p.m. I attended the Project Council Information Meeting on Physics at which Zinn presented the followin nuclear constants for U233, comparing them with the constants for Pu239 and U235:

	49	23	25	Ratio 23/25
σ_{abs}	1150	620±20	640±13	
ν	2.94	2.60±0.13	2.41±0.10	1.079
σ_{f}	820	550±20	548±10	1.004
α	0.47	0.11±0.05	0.17±0.03	
η	2.05	2.4	2.14	
σ_{c}	330	62±28	92±17	

[9, pg.326]

1944-11-08 With regard to U233 it was stated that, in order to evaluate Herbert Anderson's data obtained at Argonne, the specific activity of the samples he used must be determined accurately; this will require about 800 micrograms of oxide. It was mentioned that Site Y has detected Pu240 by mass spectrographic analysis—the sample used had about 5 parts of Pu240 per 10,000 parts of Pu239 present.

The pile at Site W has been operating at 90,000 kW for the past two weeks. It appears likely that the pile cannot be operated above 225,000 kW at the present potential loading capacity. The present plan is to run until the concentration of plutonium approaches 30 g/ton and then commence extraction of some of the plutonium. [9, pg.327]

Allison reported that two directives have been received by Compton. First, responsibility for waste recovery of uranium has been assigned to Clinton and responsibility for recovery of U233 has been assigned to the Met Lab. The second directive is that Allison's responsibilities at Chicago will end in one week on the 15th and Stearns will take over. Allison is to go to Los Alamos. [9, pg.331]

1944-11-15 Katzin and Davidson held their first meeting with Wigner, Weinberg, Ohlinger, and Young for the purpose of becoming oriented in the chemical problems involved in the design

of conversion piles (U233-fueled, D2O-moderated piles for the conversion of thorium to U233).

I attended an evening meeting of the Separation Process Subsection of my section. Studier talked about gamma- and x-ray activity found to be associated with pure U233 analogous to that previously reported to be associated with 94-239. Aluminum and lead absorption curves have been run on a 16-mg sample purified by ether extraction and by BaSO4 and lanthanum fluoride precipitations. Gamma-rays have been detected at 420, 230, and 150 kev energy, also x-rays of 87, 19, and 14 kev. The presence of electrons, probably arising from internal conversion of the gamma-rays, was demonstrated by magnetic-deflection counting techniques. I offered a probable relationship between the energies of the radiations and the quantum levels in the Th229 daughter atoms which are involved.

Hyde reported some exploratory investigations of extraction of uranium and thorium by cellosolves, esters, ketones, and diethyl ether (in connection with U233 extraction studies). Hellman spoke on the effect of thorium, aluminum, lanthanum, copper, manganese, and ammonium nitrates as salting agents upon the extraction of uranium and thorium into ethyl ether. There is a pronounced increase in uranium extraction at comparatively low aluminum nitrate concentrations, indicating its value for preliminary extraction. He said that $NH4NO_3$ is indicated as a salting agent for final purification where the amount of thorium extracted is an important factor.

Katzin reported on the probable decay chain for the "neptunium series," of which U233 is the third member. Based on preliminary experiments and by analogy with the known series, he postulates the following decay chain:

Katzin then presented experimental evidence in support of that part of the proposed decay chain following U233 and indicated the possibility of branching decay leading to isotopes of elements 85 and 87. [9, pg.337-338]

1944-11-18 a. Katzin reports that cans nos. 7-10 of thorium carbonate have been extracted, yielding about 4.5 mg of U233 per can. He also describes studies of salting-out action of various nitrates on thorium and uranium, solvents for uranium-thorium separation, the radiations from U233 and the determination of the decay chain of U233. [9, pg.341-342]

The Jeffries Committee, appointed by Compton last July to prepare a report on the future prospects of atomic energy, submitted to Compton its 65-page report, "Prospectus on Nucleonics." Members of the committee are Fermi, Franck, Hogness, Jeffries (Chairman), Mulliken (Secretary), R. Stone, and C. A. Thomas. The report begins with a review of the history of nuclear science and the principles of chain reacting piles. **With regard to the future of nucleonics, it explores the question of whether there is enough uranium to supply the power needs of the U.S. by nuclear reactors, points to the use of thorium as a supplementary source of power from fission, and describes the possibility of converting hydrogen into helium to give a virtually unlimited source of energy.** The report identifies the possibility of nuclear generators as sources of power in deserts, polar regions, on ships, or even in excursions into space. It suggests the production of new heavy elements—new

neptunium isotopes or elements beyond plutonium—that may be suitable for use in piles or other nuclear power devices. The report looks forward to the production of radioactive isotopes in piles in large quantities for use in tracer studies and for medical purposes. With regard to the nearer future of nucleonics the report explores: Physics. Nucleonics to be the liveliest part of theoretical and experimental physics with applications of radiation and radioactive tracers to other branches of physics. Chemistry. Study of transuranic elements will be pursued; use of radiation in chemical and physico-chemical processes (replacing catalysts or heat); use of tracers in reaction kinetics - C14 for investigating the mechanism of organic reactions; study of fluid flow. Biology and Medicine. Radioactive iodine in thyroid, tracer elements in studying basic problems of animal and plant metabolism - study of photosynthesis, use of radioisotopes in diagnosis. Metallurgy. Use of tracers to study diffusion, inclusions, distribution of minor constituents. Engineering and Construction. Problems of wear and lubrication (tracers); leak testing with radioactive gases; tracers for tracing flow in sewers or dams; embodying radioactive materials in paints and floor covering to eliminate static; radioactive sources as substitutes for industrial and clinical x-ray machines; illumination by mixing radioactive materials with luminescent material. Agriculture. Study of poisons and micronutrients. **Power. Limited amount of available uranium precludes a widespread use of pile power for energy production in competition with coal, oil, or falling water.** However, the (by-product) heat from piles built for plutonium production can be utilized for central heating of large areas, thus freeing oil for premium uses. On the other hand, should not conclude that the cost of pile power is necessarily going to be high as compared with other power sources. **Pile power may also be used for transportation "while pile-powered interplanetary ships still belong to the realm of scientific day-dreaming, pile-powered battleships or submarines have been considered as likely applications of nuclear power in the relatively near future."** Explosives. Changing direction of sea currents, destroying hurricanes, removing danger of earthquakes or volcanic eruptions, blasting of waterways, *e.g.*, Panama Canal. Impact of Nucleonics on International Relations. Danger of a blitzkrieg. Need for central authority to exercise control over nuclear power—necessity for all nations to make every effort to cooperate now in setting up an international administration with police powers which can control the means of nuclear warfare. Control of Materials. Should be possible to give considerable scope to free enterprise and still have the government hold a tight rein on the important factors. Postwar Organization of Nucleonics in America. Nation must maintain its leading position in nuclear research and nuclear industry. "There should be government supported nucleonics laboratories having ample facilities for both fundamental and applied research." Without a healthy development of a nuclear industry, nucleonics research will be insufficient to guarantee the leading position of this country. The report concludes with a statement of broad objectives: to stimulate widespread research in nucleonics in the United States, to encourage the development of a free nucleonics industry in the United States, to coordinate the governmental activities in nucleonics with the scientific and industrial developments in this field in such a way as to insure maximum national security, to strive for the establishment of an efficient international supervision over all military aspects of nucleonics.

Suggestions for the accomplishment of these objectives are given as follows: (a) The projects relating to plutonium, U235, and perhaps U233 should be prosecuted by the Government, no matter when the war ends, to a point sufficient for military appraisal. (b) The development of the nucleonics industry by private enterprise should be encouraged. The military by-products of the industrial developments should be made available to the government, and the use of government information and patents should be made available to industry so far as the military situation may permit. (c) Scientific education and research should be encouraged in existing university laboratories, and new research laboratories for nucleonics with special facilities should be created at universities. (d) A suitable agency, with both government and nongovernment representatives, should be established to guide and coordinate such nucleonics activities as may affect the military or other interests of the nation. (e) Enlightenment of public opinion on the scope and significance of nucleonics should start as soon as possible to bring about realization of the dangers for world security caused by the new scientific and technical developments, and to prepare for decisions which will have to be taken to meet this danger. (f) Cooperation with friendly nations in all these problems—particularly the last named one—should be given serious and prompt attention. [9, pg.341-343]

1944-11-20 In the evening I was the speaker at the first of the revised biweekly Chemistry Division Monday meetings, held at 7:45 p.m. in Room 251, Ryerson Hall. In the first of two talks on "The Heavy Elements," I discussed the nuclear properties of elements from atomic number 80 to 96, with emphasis on elements 89 to 96. Aspects covered were characteristics of heavy nuclei, energetics of radioactivity, systematics of type of radiation and half-life, neptunium series, neutron fission in the "Big Three" (U233, U235, and Pu239), and prospects for useful nuclei other than the "Big Three." The talk included a description of our recent work on the decay of the 4n + 1 series (see Fig. 29). This also includes the 4n, 4n +2, and 4n+ 3 decay series and suggests the nomenclature of classical radioactivity for the new 4n + 1 series. I included a description of the recent observation by James and Ghiorso of an isotope of element 96 produced by helium ion bombardment of Pu239 in the Berkeley 60-inch cyclotron. I said this isotope, which has a halflife of a few months, decays by the emission of alpha particles with range 4.6 cm in air, and exhibits chemistry characteristics of the +3 oxidation state of a rare earth, may be 96242 produced by the reaction Pu239(α, n)96-242. I also predicted that there is a good chance that Pu241, produced by successive neutron capture in Pu239 [*i.e.*, by Pu239(n, γ)Pu240 and Pu240(n, γ)Pu241) may be a beta emitter, leading to the possibility of observing the daughter isotope, 95-241. [9, pg.345]

1944-11-21 Figure 30. The four radioactive decay series. November 1944.

Uranium-233 Studies. Finally, I reported on our systematic study of the influence of salting-out agents on thorium and uranium solutions. I indicated that best results are obtained with calcium nitrate, ammonium nitrate (the old stand-by), and aluminum nitrate. I also mentioned our survey of solvents.

I described the gamma-rays and x-rays we have found for U233. (Here I showed the slide [Fig. 291 of the four radioactive decay series that I had presented last night at the first

biweekly Chemistry Division meeting.) I stated that: "The new radioactive disintegration chain of which U233 is a member, has now been almost completely identified. It starts with Np237, which is transformed by alpha emission into Pa 233 which in turn gives U233 by beta emission. U233 is converted by alpha emission into Th229 which has a half life of 10^3-10^4 years. From the growth of activity of isolated Th229, which has now been followed for several weeks, it is clear that an element with a lifetime of at least several months occurs in the disintegration chain below Th229. **The alpha activity of Th229 contains at least five different groups, which is consistent with the assumption that the stable final product is Bi209.** The daughter element of Th229 is Ra225. The latter emits alpha particles (perhaps also betas) and gives a short-lived emanation (86-221). In the figure, the 'neptunium series,' which may also be called '(4n+1)-series' is represented tentatively alongside the three old established series, the uranium-(4n+2), thorium-(4n), and actinium-(4n+3) series." [9, pg.348]

1944-11-22 Absorption cross section of U236. The cross section for the formation of U237 from U236 appears to be less than 135 barns. Poison chain: The cross section for Xe135 is of the order of 3 megabarns. The fission yield according to Sugarman is about 5%. [9, pg.351]

Compton asked that those present consider whether the work from here on should continue to be organized around the immediate future or whether some other objective should be followed. In the discussion of possible objectives that followed, **Jeffries pointed out that the objective of Chicago has been achieved only in part since plutonium does not have all the desirable properties that had been anticipated. The same apparently holds for U235. Perhaps, then, the only way to provide adequate insurance that the objective really be achieved would be to be ready to convert Pu239 over to U233.** This could provide a clear-cut objective.

Compton summarized the discussion, stating that (a) The primary Pu239 objective has reached the point where we should advise the Army that our part of the 49 task is largely complete. (b) There is general agreement that a large amount of work in utilization of nuclear energy is still before us, such as hexone extraction process development and conversion of 49 and 25 to 23. A discussion of pressing the work on U233 followed, and Compton indicated that agreement by the Army to proceed with moving and setting equipment for a metal fabrication laboratory would have to wait until after his meeting with Colonel Nichols this week. [9, pg.352]

1944-11-23 Katzin and Davidson attended the second meeting on the design of the homogeneous U233 pile, conferring with Wigner, Young, Weinberg, and at times, Ohlinger and Szilard. Problems discussed included handling and cooling the thorium absorbing blanket, production of water vapor and gases in the reacting unit, mechanism of ridding of fission product gases, cooling of the reacting solution, effect of heat and energy release on the materials, *e.g.*, solvents used for processing the highly concentrated fuel solution, without preliminary cooling. [9, pg.353]

1944-11-24 "The supply of several milligrams of pure U233, which has been made available for

the first time, has opened up a whole new field of exploration in radioactivity. It is now clear that U233 starts a decay chain leading to a whole set of new isotopes of the heavy elements which have been missing gaps in the isotopic chart. It is now possible with pile-produced U233 to fill in these gaps and work has been started on determining the lives and properties of these new isotopes.

"The sample of U233 purified in the Chemistry Division has been used at the Argonne Laboratory for the determination of the cross section for neutrons and the fission yield. Fission chains can be produced from U233 as well as from U235 or Pu239, but the details will be given in reports from Argonne. Solvent extraction methods are being perfected for the extraction and purification of U233 from irradiated thorium." [9, pg.355]

1944-11-27 Dempster and I talked by phone about the preparation of plutonium samples to be used for mass spectrometer identification of Pu240 and also the preparation of U233 samples to be used to determine their isotopic composition.

Daniels sent a memo to Stearns about the Chemistry Division progress outline for November, summarizing work in Section C-I on (9) isolation of 18 more mg of U233 from irradiated thorium, (10) radiations from U233 and its decay chain.

"Hexone extraction of U233 from thorium is satisfactory in the presence of an excess of thorium and aluminum salts. Experiments are under way to find the most suitable ketone. The new neptunium decay series is being filled in with new isotopes which have become available as a by-product of the pile. For the first time a decrease in elastic modulus has been observed in graphite which has been subjected to long exposure in the pile. It is not known yet whether this phenomenon, connected with the Wigner effect, is to be interpreted as favorable or unfavorable. **The latest measurements on the properties of xenon connected with the poisoning of the pile are: yield—5.6 percent, cross section for slow neutrons—3.1 million barns, and half-life—9.2 hours.** New developments in analytical techniques include the use of porous graphite electrodes for spectrographic analysis which act as wicks to feed in solution at the proper rate, the application of the polarograph to the determination of plutonium in uranium solutions, and the simplification of analysis of zirconium and iron by extraction with organic solvents." [9, pg.358-359]

1944-11-28 The discharge of the first uranium slugs from the Hanford pile was completed today. The plutonium content is low because the uranium did not receive full production-level irradiation. [9, pg.360]

"As material comes to the Hanford extraction plant, following pile irradiation and 'cooling,' it consists of cylinders of uranium metal encased in aluminum. In each metric ton of uranium metal there are expected to be about 250 grams each of plutonium and fission products. The radioactivity of the fission products, after 100 days of irradiation at 2500 kW per ton and 40 days 'cooling,' is about 3.5×10^6 curies per ton of uranium. After 60 days cooling the activity is about 2.5×10^5 curies, and further cooling has less and less effect. The extraction problem thus becomes a double one—chemical extraction of 250 grams of plutonium from 1000 kg of uranium, and separation of plutonium from fission activity. Accepting the value

of 10 millicuries as the maximum amount of beta and gamma radiation that may be worked with directly by a chemist, with some shielding, 'decontamination' from fission activity must be accomplished by a factor of $2.5 - 3.5 \times 10^7$ for material cooled 40-60 days. Furthermore, this extraction and decontamination must be performed entirely by remote control, behind biologically adequate shielding (*e.g.*, 8 feet or more of concrete). [9, pg.361]

1944-11-29 Katzin gave a brief review of the work on the homogeneous U233 pile, which is to consist of about a cubic meter of heavy water with 7 or 8 kilograms of U233 in solution. **Since the pile is to be run at 100,000 kW, the cooling problem is severe, and since the power per unit volume is much higher than any other pile, the problem of poison by fission products is a much more serious one. Apparently, the fission products will have to be removed almost daily.** [9, pg.365]

1944-11-30 "During all this time a competent staff of personnel must be maintained which is qualified and willing at a moment's notice to devote its entire attention to any unforeseen problems which may arise at the Hanford Engineer Works or Y. **To maintain such a staff intact with a sufficient degree of morale, it is necessary that they work on some forward-thinking problem. Most promising of the problems at the present time seems to be in the field of (1) 'converter' piles and 'breeder' piles;** (2) chemical procedures for handling metal much faster than planned in the original flowsheet; (3) the continued investigations of the effect of radiation on graphite." [9, pg.368-369]

1944-12-01 Hogness issued a summary of manpower distribution in the Chemistry Division which shows that there are 157 people in the Division as of December 1, including 80 in my section. The groups under Albaugh, Cunningham, and Katzin are divided as follows:

		Number of Men	
		Oct.	Nov.
Albaugh (Site W work, 32 men)	R. Thompson, extraction and decontamination	10	9
	Katz, concentration and isolation	5	3
	Gilbreath, process development	8	6
	Egan, semiworks	0	9
	Lawroski, solvent extraction	4	4
Cunningham, (Site Y work, 39 men)	Simpson, high vacuum work	10	10
	Hindman, basic chemistry	10	10
	Dawson, recovery	8	8
	Ghiorso, instruments and physical measurements	9	10
	Katzin, 23 work	6	6

[9, pg.370]

1944-12-03 Also at Clinton, 15 cans of $ThOCO_3$, specially purified from uranium, went into the pile in a position giving 1.4 times the average flux. [9, pg.371]

1944-12-04 The first production run of irradiated Hanford uranium slugs is scheduled to be received in the separation plant's underwater storagee area today. [9, pg.371]

Compton asked Stearns for the following information to aid in deciding whether or not to authorize an immediate program for the production of 23 for possible use in the present war: "(1) A feasible time schedule for the production of 23 in quantities of 400 grams per day, assuming the availability of the required amounts of 25 or 49, (2) Additions to the research staff required for such a vast project, (3) Assuming as an alternative that the program is to be carried out, but not for immediate application in this war, what work should be done by the Met Lab before June 30, 1946." **Compton mentioned that he has received a memo from Clinton Labs with regard to future developments which includes a proposal for construction of a small experimental 23 "producer" at Clinton as a part of a larger program toward 23 production.**

At 7:45 p.m. I gave my second talk on "The Heavy Elements" at the new biweekly Monday evening Chemistry Division Meeting in Room 251, Ryerson Laboratory. After recapitulating my November 20 talk on nuclear properties, I completed that aspect by discussing the work of James, Morgan, and Ghiorso on our possible observation of an isotope of 96 from Pu239 plus helium ions, possible isotopes of 95 from Pu239 from deuterons, and our proposed look for 95241 from Pu241 beta decay or, later, 95-243 from Pu243 beta decay (the Pu241 and Pu243 to come from intense neutron irradiation of Pu239). **I mentioned the U232 found in pile-produced U233 in the work of Ghiorso, Katzin, Studier, and Hagemann.** [9, pg.372]

1944-12-05 The first uranium slugs are to be charged into the second Hanford plutonium manufacturing pile, the D pile, today.

At 10:30 a.m. I attended the Project Council Information Meeting on Physics. Others present were Borst, H. Brown, Burton, Compton, C. M. Cooper, Dancoff, Daniels, Darrow, Dempster, Estermann, Franck, Friedman, Goldsmith, Greninger, Hamilton, Hill, Jesse, Langsdorf, Lapp, Lewis, Lichtenberger, Manning, Maurer, Moon, Mulliken, Nordheim, Ohlinger, Rabinowitch, Seitz, Shonka, Simpson, Spedding, Stearns, Stephenson, Stern, Sugarman, Szilard, Wakefield, Watson, Wattenberg, Way, Weinberg, Whitaker, Wigner, Young, Zachariasen, and Zinn. Some of the high points are as follows. Weinberg presented the results of calculations on the properties of a pile run at high temperature (2000°C) as envisioned in the "pebble" pile suggested by Daniels. The advantage to be gained by such a pile is the elimination of the 40 difficulty (production by neutron capture in 49) by the continuous distillation of plutonium from the uranium carbide spheres. His calculations indicate the prospects for such a pile are poor because of the loss in k at the elevated temperatures. **Young reported on his study of a design of a U233 "breeder," *i.e.*, a pile that would operate on U233 and breed more U233 in a surrounding thorium shield than is consumed in the pile.** He concludes that it is a promising possibility but that some types of losses may have been overlooked. There are many technical problems that have not been investigated. Nordheim

reported on a small pile designed for Site X, which would be a 23 breeder or converter. He visualizes a pile using uranium enriched by a factor of 12.5, moderated by heavy water with a graphite reflector, and surrounded by thorium.

At the end of the meeting Compton gave the following information he has just received from Los Alamos; (a) neutrons per fission for U233 = 2.45±5%, which is equal to the value previously obtained for U235; (b) the relative abundance of 94-240 to 94-239 as measured in metal obtained from Site X is determined by mass spectrograph to be 3.4×10^{-4}. [9, pg.373]

1944-12-06 On the subject of materials, I mentioned we have a pound of depleted uranium in which there is a 25-fold depletion of U235. We also have about 20 mg of ionium (20% ionium, 80% thorium mixture) from Berkeley that may contain as much as one mg of protactinium; it was originally obtained from a process for isolating UX1 from tons of uranium to make 10 micrograms of U234. It has been decided that six cans of neutron-irradiated extra pure thorium (free of uranium) will be brought up from Clinton on January 1. Forty cans of ordinary thorium (irradiated) have come from Clinton for extraction—about 4 mg of U233 per can.

I brought up the subject of working hours of the Met Lab academic personnel. The Army has been examining attendance records and says that few people are working the established 48-hour week. Only a few arrive at 8:30; by 9:00 a small number more have come in. On the other hand, the signature sheets show that relatively few check out after 6:00 p.m. or come back to work at night. This laxity probably will result in a very strong request for our men to go to other sites where manpower is badly needed. I also requested that no newspapers and magazines be left lying about because the Army objects to the reading of these on working time. [9, pg.374-375]

1944-12-08 At Clinton today, 14 additional cans of $ThOCO_3$, specially purified from uranium, go into the pile in a position giving 0.3-0.4 times the average flux. [9, pg.384]

1944-12-12 At 9:45 a.m. I arrived in Pendleton, Oregon. I was met and then continued by automobile to the Hanford Engineer Works. There has been much construction at Hanford Village since my visit last May. During the afternoon I made a tour of the pile and chemical separations areas. The one pile now operating is running at a power level of 125,000 kW but is expected to go to its rated level of 250,000 kW when all 2,000 tubes have been filled. Each pile has 200 tons of metal instead of the originally planned 100 tons. It will take 200 days operation at 250,000 kW to attain 250 grams of plutonium per ton of metal.

One canyon is finished and is running with tracer now. The canyon is a tremendous construction 860 feet long. Looking down at the control boards the length of the canyon, one sees nothing but 860 feet of valves, meters, indicators, controls—a fantastic sight. Everything is handled completely by remote control. The insides of the cells cannot be seen except by means of a thoroughly shielded periscope handled by an operator from an overhead crane. Each cell has three staggered seven-foot concrete block covers (15 tons each) which can be removed by an operator in a thoroughly shielded overhead cab. The periscope enables the operator to use remote control wrenches and other tools to replace defective equipment.

Slugs coming out of the pile glow red from the photons shooting out. (Their temperature is, of course, much below red heat.) They are dropped into a deep water channel and caught in big buckets which carry them into large lead coffins, after which they are hauled out into the desert five or ten miles to cool.

The piles are in the 100 area and the extraction canyons in the 200 area. Both run on a 24-hour schedule. Extraction and decontamination takes place in Bldg. 221B. Concentration (the crossover step) takes place in Bldg. 224B, and isolation of the product occurs in Bldg. 231. The Control Labs (Bldg. 222B) are also in this area. The 300 area is near Richland—about 20 or 25 miles away from the 100 and 200 areas. This area contains the research building (No. 3706) where John Willard and his group work: the test pile, the canning building, the cold semiworks, etc. (see Figs. 36-47).

I had dinner at the Perlmans' and then spent the night at the Richland transient quarters. [9, pg.387]

1944-12-14 At an afternoon meeting of Division Directors in Eckhart Hall, **Stearns and Wigner reported that a letter has been received from Compton asking if it is possible to make plans for converting 100 grams of Pu239 into at least 70 grams of U233 per day.** Wigner outlined the sandwich pile which his Physics Division considers to be the most practical for quick results. (Plutonium is sandwiched between thin aluminum sheets folded in the form of a bellows and placed in a cylindrical tank two feet in diameter and eight feet high. Ordinary water is forced through the bellows. Surrounding the plutonium cylinder and the water cooling is a concentric cylinder of thorium metal, oxide or oxide slurry.) The plutonium must be decontaminated from fission products by a factor of 10^7 once every ten days to remove neutron poisons and to make it possible to refabricate the plutonium; high yields of plutonium (99.9%) are required to make the desired yields of U233 possible. A report to Compton is to be written by January 6. Daniels was asked by Wigner to bring back a report from the Chemistry Division on the feasibility of obtaining high efficiencies (yields of the order of 99.9%) in purification and decontamination by the solvent extraction method. [9, pg.399-400]

Katzin wrote a summary of the U233 work for my use at the Project Council Chemistry Information Meeting on Tuesday. Topics covered are (1) Additional information has been obtained on salting-out action of various nitrates on thorium and uranium. Data obtained are in harmony with the interpretation that high extraction of thorium into ether results in "salting back" of the uranium into the aqueous phase. The desirable combination of high salting of uranium and low salting of thorium into ether is shown most outstandingly by calcium nitrate, by ammonium nitrate at high concentrations, and by aluminum nitrate at low concentrations. [9, pg.400]

(2) Tests on solvents for use in the uranium-thorium separation have been extended to the extraction on protactinium. It has been found that diisopropyl ketone will extract protactinium to the extent of 50%. (3) Calculations and analysis of data obtained on the 4n+1 decay chain indicate that as little as 10^{-8} parts of U232 formed by the $n, 2n$ reaction on

U233 or Pa233 may produce sufficient amounts of radio-thorium and the subsequent members of the thorium decay chain to complicate the experiments on the 4n+1 series. So far, it has been learned that RdNp(Th229) probably has a half-life of several thousand years and NpX (Ra225) has a half-life of about one month. (4) If a radium-thorium separation is made and the radium fraction set aside for several weeks, the ThX and its daughter activities decay out leaving the presumably pure neptunium series members. Experiments on material that has been handled in this fashion have confirmed the presence of Pb209 (NpD) with the known half-life of 3.3 hours. Isolation of the bismuth fraction from the aged NpX has given an alpha activity of approximately 45 minutes half-life. Although no accurate alpha-particle range curve is available from which to determine the branching ratio, there are definitely two differing ranges of alpha particles detectable, one of which, no doubt, corresponds to NpC and possesses a very long range. Unsuccessful attempts to isolate members of the series between NpX and NpC, correlated with the rate of growth of NpC and NpD, indicate that all other half-lives involved are probably shorter than ten minutes. No decision can be made yet concerning possible branching at NpX and other points along the chain. [9, pg.401]

1944-12-15 In the afternoon Manning, Katzin, and Davidson met with Daniels to discuss **the feasibility of obtaining high efficiencies in purification and decontamination operations required in connection with the converter pile being designed for the conversion of 100 grams of plutonium per day to U233 with a yield of at least 70 grams.** It was concluded by everyone that at the present state of solvent extraction development the best efficiency would be about 95% for a decontamination factor of 10^2 making it necessary to fabricate the plutonium by remote control, and that the required yield of 99.9% and 10^7 decontamination is impossible. [9, pg.401]

1944-12-18 At 2:00 p.m. there was a meeting in Daniels' office to discuss the problems of the converter pile. Those present were C. M. Cooper, Daniels, Davidson, English, Katzin, Maloney, Manning, and Tepe. **All agreed that the efficiency in chemical processing of plutonium that Wigner hopes for, namely 99.9%, is impossible if decontamination is to be** 10^7**.** If decontamination of 100-fold is permitted, requiring remote control for plutonium sandwich refabrication, a yield of 95% might be hoped for. Other types of piles were discussed with English advocating the homogeneous heavy water pile, Cooper the homogeneous pile using circulating water with plutonium nitrate in solution, and Daniels the high temperature pebble pile using bismuth as coolant. [9, pg.403]

1944-12-19 At 9:30 a.m. I attended the Project Council Information Meeting on Chemistry in Room 209, Eckhart Hall. Others present were Arnold, Bartky, Boyd, Burton, Cohn, Compton, Connick, Coryell, Daniels, Dempster, Doan, English, Fred, Hilberry, Huffman, Jeffries, W. Johnson, Manning, Mulliken, Nickson, Rabinowitch, Spedding, Stearns, Stone, Sugarman, Szilard, Turkevich, Wakefield, Watters, Whitaker, Wigner, Zachariasen, and Zinn. I was first on the program and reported on investigations of separation processes for Hanford, on basic chemistry, and U233 as follows:

(3) U233 program—Katzin's group has made preparations for the extraction of 37 cans of

irradiated thorium carbonate shipped from Clinton, and 28 mg of U233 have so far been extracted. We have redetermined the half-life of U233, based on the new isotopic composition of 96% U233 and 4% U238 measured by Dempster (the old measurement was 86% U233). **The new value is 160,000 years.** I presented the latest information on the neptunium decay series. [9, pg.403-404]

I reviewed a memo (MUC-GTS-1201) prepared for Daniels on the pile proposed by Wigner for converting Pu239 to U233. In this memo I note that the losses envisioned by Wigner in the operation of the pile are such that the 30% operating margin is essentially completely used up in the physical losses caused by neutron absorption by fission products, materials of construction, etc. A complete 100% yield is necessary for all chemical procedures to stay within the required 70% efficiency.

I include the following table, based on the assumptions that the converter will run on a cycle in which either one-tenth or one-twentieth of the material must be processed daily to eliminate neutron-absorbing fission products:

Chemical yield (%)	Production loss (grams)	Overall efficiency (in percent, 10-day cycle)	Overall efficiency (in percent, 20-day cycle)
100	0	70	70
99.9	2.8	68.2	69.0
99.5	14	61.4	65.4
99.0	28	54.6	61.4
98	56	44.9	54.6
95	140	29.2	41.0
90	280	18.4	29.2

[9, pg.405]

I point out that an efficiency of 99.9% for a decontamination by a factor of some 10^7 **followed by refabrication of an aluminum sandwich is totally impossible by any known process and probably also by any conceivable process.** It is the consensus that a process giving a 95% yield is an optimistic possibility which means an efficiency of not better than 41% for a 20-day cycle. This might involve a combination of precipitation and solvent extraction procedures.

I suggest that if the decontamination requirement can be lowered to a factor of 100 (presuming it is feasible to have remote-control refabrication of the fuel), a chemical yield of 98% is possible by a solvent extraction procedure alone. **I also mention, as an alternative possibility, a homogeneous reactor with ordinary water that would require a minimum of decontamination (only to remove fission product poisons) and would avoid the hold-up due to fuel fabrication operations;** however, little is known about this approach, and hence this could require the most research work.

With regard to the processing of the thorium blanket in Wigner's converter pile, I indicate the problems seem straightforward, **although there would be a delay in harvesting the uranium because of the need to await the decay of the 27.4-day protactinium precursor.** I suggest the use of thorium metal as the best practical form.

I mention some of the heavy isotope problems that will arise in the operation of a converter unit. Pu240 will be formed at the rate of 40 g/day if the rate of fission of Pu239 is 100 g/day. If Pu240 has a low fission cross section (and a low capture cross section), the average cross section of the plutonium in the pile will decrease at a rate of 15 barns per day. If Pu240 has a capture cross section of 10 barns, there may be 10 g of Pu241 in a unit after 100 days operation.

In summary, I say we favor the heterogeneous arrangement with refabrication by remote control, in which case an "individual" chemical cycle yield of 98% may be possible, corresponding to an overall yield of some 55% U233 on a 20-day cycle. Thorium metal seems to be the best material for the thorium absorption blanket.

I indicate this summary was prepared at my direction by Katzin and Davidson. [9, pg.406]

1944-12-20 Daniels sent Wigner a memo about his conception of a high temperature bismuth-cooled converter pile to be fueled with pellets of plutonium oxide spaced in a lattice composed of pellets of beryllium oxide. The U233 is formed in an outer shell through which thorium oxide powder is passed slowly.

At 4:00 p.m. I attended a meeting on conversion piles under Wigner's chairmanship. Compton was also present. At Daniels' request, I presented technical details of the chemistry involved in the processing of fuel from Wigner's sandwich pile. **Wigner was disappointed in the low yields, but we indicated such yields would be inevitable.** C. M. Cooper described a heat exchanger which he has proposed for homogeneous water piles. Greninger outlined his idea of sandwich fabrication. Daniels discussed the high temperature bismuth-cooled pile.

At the conclusion of the meeting, there seemed to be the general opinion that there is probably a good chance that a converter pile can be made which will be certain to operate with an efficiency of at least 50% for the conversion of Pu239 to U233. All effort must be directed toward trying to make an attractive and practical case for this pile to be presented by January 6.

The evening meeting of the Basic Chemistry, Recovery, and Instrument Groups of my section, at 7:45 p.m. in Room 209, Eckhart Hall, was attended by Arnold, Brody, Cunningham, Daniels, Dixon, Ghiorso, Hagemann, Hufford, Jaffey, Katzin, Krueger, Larson, Manning, McLane, Morgan, Nickson, S. Peterson, Phipps, Sedlet, Sheft, and me. I turned the meeting over to Cunningham to preside as usual. Jaffey gave the first of a series of lectures to be presented at these meetings to acquaint chemists with the essentials of nuclear physics. His topic was the concept of nuclear cross sections. Near the end of Jaffey's lecture **Katzin asked whether the unit of cross section referred to is a barn or a barn door, to which I replied that the unit-is called a barn because it is as large as a barn door in comparison**

with actual cross sectional areas of nuclei. The term was originated by Fermi. I also called attention to the fact that the series of lectures on the fundamentals of nuclear physics begun by Jaffey will include concepts with which any chemist having worked on the Project should be thoroughly familiar. [9, pg.408]

Compton said that he recently discussed the U233 program with Tolman and Conant and concluded that studies looking toward its production should probably be the major effort of the Project, although not with the expectation of use in this war. **The development of nuclear power is longer term, not for this war; hence plans can be delayed for a few months and need not be ready by February 1, 1945.** Compton stated that it is of the greatest importance that we emphasize our need for fundamental scientific information in our recommendations to the Army. He believes a general feeling exists throughout the U.S. that war research has greatly advanced our knowledge of fundamentals, whereas the actual case is quite different since most war developments have been in the application of existing knowledge. Our duty is to correct this impression. Wigner complained about the lack of liaison with Site W and Y that has developed. Greninger agreed that information from Site Y has dropped off since Cyril Smith ceased his visits to Chicago. Compton agreed to have John Wheeler attend the February Physics Information meeting to help bring us up-to-date on Site W activities. [9, pg.412]

1944-12-21 The isolation and purificiation of the 25 mg of U233 for Los Alamos is now complete. The material has been shipped. [9, pg.412]

It should, of course, be possible to use essentially the same types of methods to decontaminate U233 and U235 and mixtures of these isotopes with each other and/or with Pu239 (in the presence or absence of U238) from the fission products and from other isotopes formed when these (U233, U235, etc.) are used for running chain reactions.

Daniels sent a memo to Wigner stating that I am preparing a more complete report on the research on solvent extraction required in order to meet the new drastic requirements for recovery and decontamination in connection with the "sandwich" converter pile. Daniels quotes significant parts from my memo of December 19, MUC-GTS-1201, as an interim report.

In addition I conferred with Wigner about converter piles and explained again the difficulty of achieving complete recovery of plutonium and high decontamination. Wigner indicated he will think about the matter. [9, pg.413]

1944-12-26 I wrote to Allison at Los Alamos describing the following samples we have sent him. (1) 100 micrograms of 93-237. This material may contain about 0.05% Pu239 by weight. It came from Clinton uranium at a level of 5 grams Pu239 per ton. (2) 25 milligrams of U233. The actual amount is more than this because the new measurement by Dempster indicates that the isotopic composition is 96% U233/4% U238 rather than 88% U233/12% U238 on which the original quantity was calculated. I point out the difficulties in his suggestion that it might be feasible to measure the amount of U238 in our U233 by counting the UX beta particles. It would not be possible to do this directly, without a chemical separation,

because of the x-rays and gamma-rays arising from the disintegration of the U233 itself; it might be barely feasible to look for UX in the separated UX1 fraction containing Th229, although the Ra225 daughter of Th229 gives rise to the whole remainder of the 4n+1 series and makes the measurement of UX1 difficult. (3) 10^7 disintegrations per minute of a mixture which seems to be 50-50 by weight of Pu238 and Pu239 on the basis of alpha-particle range measurements; the Pu239 probably was introduced into this deuteron-produced Pu239 by contamination in the laboratory. [9, pg.423]

1944-12-27 Katzin mentioned the problems in separating the uranium, thorium, and radium in the 4n+1 series. Thorium-229 unfortunately is carried on BaSO4 to approximately 30%, so BaSO4 is not too good to use to precipitate the radium out first. I suggested precipitating the thorium first, perhaps with zirconium iodate or ceric iodate, following which the radium could be precipitated with barium sulfate. [9, pg.424]

Extraction-Decontamination: Ader, Kelley, Ames, Post, and Hopkins (it was indicated that Post, Kelley, and Ader were drafted from the laboratory and are now in the Army with the expectation that they will be assigned back here). These men are highly trained in radiochemistry, and will be needed for any extraction-decontamination research for piles converting plutonium to U233. Recovery: Fineman, Anderson, Britain, Asprey, and Stewart. These men have a high degree of skill, are needed for the important ongoing program of recovering, purifying, and preparing special forms of plutonium. Solvent Extraction: Hausman and Schraidt. Shotgun Tests: Brownell, Lewis, and Cressman (not C-I men). [9, pg.424-425]

I attended the evening meeting of the Separation Process Subsection of Section C-I at 7:45 in Room 209, Eckhart Hall. Others present were Albaugh, Arnold, Blaedel, Bradt, Cunningham, Daniels, Egan, Ghiorso, Gilbreath, Hagemann, Hyde, Hyman, Jaffey, Jones, Katzin, Kraus, Larson, Manning, Morgan, S. Peterson, Sheft, Walling, Winner, Wolf, and others. Davidson discussed the breeder and converter piles currently being proposed. **The structure which has been proposed for the so-called "breeder pile" is of the homogeneous type. A solution containing 3.5 kg U233 in 400 liters of D2O (or possibly H_2O) would be continuously recycled through a chamber surrounded by a thorium reflector or blanket (to produce more U233 from Th232). Davidson pointed out that this type of structure has some advantages from a chemical standpoint. While it would be necessary to decontaminate the circulating U233 solution either at intervals or continuously, the fact that the fission products would be formed in a solution might simplify the chemical processing problem.**

Davidson mentioned that more serious consideration has been given to the possibility of using the fission of Pu239 as the source of neutrons to form U233 from Th232—this is a converter pile. The homogeneous type of pile has been considered for this scheme also. Davidson described the alternative sandwich-type converter pile devised by Wigner. He detailed the chemical and other problems in the use of the sandwich pile as described in my memo to Daniels of December 19. **He also stated that difficulties could arise from the production of U232 in the U233.** [9, pg.425]

1944-12-28 I completed a ten-page report (MUC-GTS-1218) entitled "Conversion of Pu239 to U233" and sent it to Hogness. It gives a more detailed presentation of the chemical problems involved in the pile for converting Pu239 to U233 than covered in my earlier memo to Daniels (MUC-GTS-1201 of December 12, 1944).

I begin by revealing the complications that will be introduced by the nuclear side-reactions in relation to the problem of final chemical purification of U233. I point out that if care is not taken to control small amounts of heavy isotope impurities, **the final material may have the same limitations as the material (plutonium) whose deficiencies are being remedied.** At the time the material is worked up the thorium will contain elements 90, 91, 92, and in some cases, 93 and 94. In addition to U233, there will be produced as relatively stable long-lived isotopes, Pa231, U232, U234, and, in the case where 94 is present, U235, Pu239, Pu240, and Pu241. **Of particular concern is the presence of U232 that has four short-lived (hence long-range) alpha-particle decay products, which means that the problem of purifying the material from light element impurities assumes importance again as it originally did in the case of Pu239.** There is also the yet unanswered question of spontaneous fission in U232 or one of its daughters. All of these factors make it seem extremely important that the factor of possible fast neutron reaction (the source of U232) in the thorium mass be minimized, including the possible necessity of processing the thorium at shorter intervals than other considerations might dictate.

In the area of chemical considerations, **I point out that should a pile containing thorium enriched with uranium be adopted, the chemists would be confronted with the very interesting, although somewhat staggering, problem of devising a means for separating elements 90, 91, 92, and 94 from each other and from fission products with a decontamination factor of 10^7—with all these separations done rapidly and with high yields.**

I then review the problems of chemical decontamination, assuming the sandwich pile proposed by Wigner, with a 20-day operating cycle. I urge that for great simplification of chemical processing, a decontamination factor of only 10^2 (rather than 10^7) be required, which would certainly justify the expenditure of a great deal of effort to develop a remote control process for sandwich fabrication. As a basis for estimating the manpower requirements for development of a solvent extraction process that would give 98% plutonium recovery and 10^7 decontamination, I examine the following factors that would require laboratory investigation: (1) Development of a method for complete dissolution of the irradiated plutonium sandwich. (2) Selection of a solvent that would be stable at the high radiation levels encountered. (3) Determination of distribution coefficients for plutonium and fission product activities. (5) Study of feed and subsequent process solutions for undesirable valence or state of aggregation of plutonium. (6) Development of a method for final concentration of plutonium solutions. (7) Determination of optimum recycle operations to obtain maximum plutonium recovery. I also review the many possibilities for and the magnitude of plutonium losses.

On the basis of a 50-50 probability of success I estimate that it would require over 100 men and at least 6 months to develop a process that would give 98% recovery with 10^7

decontamination. I bring up the simpler problem of 10^2 decontamination, suggesting that a yield of 99% rather than 98% might be attained. In conclusion I briefly cover the problem of U233 separation from thorium and indicate the problem is much simpler; we have had the benefit of experience in carrying out the separation on a laboratory scale in Katzin's group. [9, pg.426-427]

"An additional 25 mg of U233 has been extracted from eight cans of thorium carbonate irradiated in the pile at X. This U233 has been purified and shipped to Y. A continuous extraction process is being devised which should make it possible to work up the remaining 29 cans containing U233 within the next few weeks. It is expected that the remaining cans will yield about 130 mg of U233, but after these have been worked up there is no further source of U233 in sight at the present time. These experiments on the production of U233 constitute important pioneering work for any later development of conversion piles.

"**A fundamental study of the new neptunium decay chain was reported last month. As a result of these studies it has been found that some U232 is formed in the pile along with the U233 as a by-product according to one or more of the following reactions:**

$$\begin{aligned} \mathrm{U}^{233}(n,2n) &\rightarrow \mathrm{U}^{232} \\ \mathrm{Pa}^{233}(n,2n) &\rightarrow \mathrm{Pa}^{233} \rightarrow \mathrm{U}^{233} \\ \mathrm{Th}^{232}(n,2n) &\rightarrow \mathrm{Th}^{231} \rightarrow \mathrm{Pa}^{231}(n,\gamma)\mathrm{Pa}^{231} \rightarrow \mathrm{U}^{232} \end{aligned}$$

U232 has a short half-life (approximately 50 years) giving off alpha rays and producing decay elements which fit into the natural thorium decay series. This series includes several short-lived alpha-emitters. Among these alpha-emitters is ThC' which gives off alpha-rays of exceedingly high energy. This fact introduces complications, in that the tolerance limits for some of the lighter elements may have to be made more exacting for the ultimate use. **U233 appears to be better than Pu239 and purification from light elements would seem to be less exacting by a factor of ten or so. However, this advantage may be completely nullified and, in fact, the situation may be worse if appreciable amounts of U232 are present in the U233.** The possibility of high spontaneous fission rates for U232 and its decay products must also be taken into account. These possibilities must be considered in designing piles for converting plutonium into U233. It seems likely that the formation of U232 can be sufficiently minimized by placing the thorium only at the outside of the pile where there are no fast neutrons and by reducing the length of exposure before the U233 is extracted from the thorium." [9, pg.429]

1944-12-30 "In the Chemistry Division, the column for product isolation by solvent extraction has been successfully operated, and this method should prove to be of great value if a 49 to 23 conversion program is undertaken. An additional 25 mg of U233 have been prepared and shipped.

"The main efforts of the Laboratory have been toward the preparation of (1) **a report to the Project Office on the feasibility of a conversion of 49 into 23 as a war-time insurance measure**; (2) the preparation of a report to the Project Office making recommendations for a research and development program for the Laboratory from June 30, 1945, to the end of the war; and (3) the preparation of final reports, looking toward the termination of the present contract as of June 30, 1945." All information on Section C-I in the Chemistry Division portion of the report is derived from Report MUC-FD-39, which was submitted to Stearns on Thursday. Total expenditures for the month of December were $861,462. Personnel employed at the end of the month were 1,522 (581 technical), a net decrease of 44 during the month. [9, pg.430-431]

1945-01-01 The six cans of thorium carbonate, specially purified from uranium, were removed from the Clinton pile for shipment to Katzin here at the Met Lab.

Hogness issued a summary of the manpower distribution in the Chemistry Division as of January 1, 1945. It shows the following for my section accounted for out of a total of 77 men:

		Number of Men	
		Nov.	Dec.
Albaugh (Site W work, 30 men)	R. Thompson, extraction and decontamination	9	7
	Katz, concentration and isolation	3	3
	Gilbreath, process development	6	8
	Egan, semiworks	9	0
	Lawroski, solvent extraction	4	11
Cunningham, (Site Y work, 37 men)	Simpson, high vacuum work	10	9
	Hindman, basic chemistry	10	11
	Dawson, recovery	8	6
	Ghiorso, instruments and physical measurements	10	10
	Katzin, 23 work	6	7

[9, pg.432]

1945-01-03 At 8:00 a.m. I held a meeting of the Council of Section C-I in my office, attended by Albaugh, Cunningham, Davidson, Egan, Ghiorso, Gilbreath, Jaffey, Jones, Katzin, Kraus, Lawroski, Manning, Schaffner, Simpson, Stewart, and Roy Thompson. I reviewed the activities of the last few weeks concerning the calculations on technical extraction problems involved in the use of converter piles. I mentioned that the problems are more complicated than at Hanford since the necessary decontamination factor will be about 10^8 (at Site W it is 10^7) and about 2 kg of plutonium per day must be handled as compared with 1/4 kg per

day per canyon at Site W. I also mentioned the high yields (98% to 99%) required because of the repetitive nature of the decontamination process and the fact that only the solvent extraction process can give such yields. **A further problem, not solvable by chemical processes, I went on, is that for every 140 grams of Pu239 present there are 40 g of Pu240 formed in addition to 100 g of fission products. If Pu240 will not fission with slow neutrons, the result will be that in successive decontaminations the dead isotope will be carried along, taking up valuable space, and contributing nothing.** It may even cause considerable trouble by the reaction 94-240(n, γ)94-241. This, in effect, would be a completely unremovable poison although its effect may be somewhat mitigated since 94-241 is probably slow neutron-fissionable. [9, pg.433-434]

I had a meeting with H. W. Koch, a research physicist at the University of Illinois, and Dancoff regarding preparation of samples for neutron measurements by Koch using the University of Illinois cyclotron as the neutron source. The program calls for Cunningham to prepare the following samples for the measurements: (1) a 62"x 62" ordinary uranium, 100 mg, on an aluminum plate with a cellulose acetate cover by January 10; (2) same type of plate with depleted U238 by January 17; (3) same type of plate with U233 by February 5 (Zinn may use the U233 first); (4) same type of plate with Pu239 (after the U233 plate, unless we cannot get the U233 extracted by January 31). Koch will be here to watch the preparation of the U233 or Pu239 plate about February 1-5. We will send a man to the University of Illinois with the U233 plate to show their physicists how to handle it for the neutron cross section measurements. [9, pg.434-435]

1945-01-04 III. U233 problem. (Most of the material listed here is covered by various categories already included in the outline. The material is gathered together to emphasize the magnitude of work related to this problem.)

A. Separation studies.

III. A. 1. Continued study of solvent separation from thorium, protactinium, and uranium from fission products.

2. Investigation of possible complexing agents to help in separations.

3. Investigation of solvents for sulfate and possibly chloride, as well as nitrate, looking towards use in breeder.

4. Continue work on theory of salting-out action in solvent extraction of inorganic salts, for use in predicting behavior, agents, etc.

5. Concentration procedures in uranium extraction for a. Straight solvent. b. Complex in solvent.

6. Precipitation procedures to separate a. Uranium from protactinium and thorium. b. Uranium from protactinium alone. c. Protactinium from thorium and/or uranium.

7. Development of selective carriers for uranium that do not carry thorium or protactinium.

8. **Work on ThF_4-UF_6 methods for continuous removal of product.**

9. Decontamination of highly active U233 solutions (from breeder piles).

10. Study types of fission products removable as volatile fluorides in terms of poisons and competitors for neutrons.

B. Complete the characterization of the U233 decay chain (4n + 1 series), including radioactive constants of daughter isotopes.

C. Nuclear properties.

1. U233

2. U233 decay products.

3. By-products which may be formed during production of U233, *e.g.*, U232 and its decay products.

D. Basic chemistry of thorium, protactinium, and uranium in relation to

1. Decontamination.

2. Purification

a. For satisfactory use in pile.

b. For final purification of U233 for use as weapons. [9, pg.439-440]

Volume 19, Production and Separation of U233. Seaborg, Editor; Katzin and Stoughton, Associate Editors. Editorial Board: Seaborg, Katzin, Stoughton, a Berkeley representative, and perhaps B. Goldschmidt as consultant. [9, pg.441]

1945-01-09 I also told Stoughton about the plan for him to be an associate editor of the Proceedings of the Metallurgical Laboratory volume on "Production and Separation of U233" and suggested that he bring information on the samples (of thorium carbonate being irradiated in the Clinton reactor) next week. Stoughton continued the conversation with James and Katzin. [9, pg.447]

1945-01-10 Katzin summarized, by memo, information on U233 for my use at the Project Council Information Meeting on Chemistry next Tuesday. He reports that 25 mg of U233 have been extracted from eight cans of irradiated thorium carbonate, purified and sent to Site Y. A continuous batch extractor, semi-automatic in operation, was used. There was some difficulty with the appearance in the ether of an organic material which held uranium in the ether phase.

The U233 on hand emits about one alpha particle in two thousand that is from U232, corresponding to about one part of U232 in 10^7 of U233. The yield seems high in comparison with the expected $n, 2n$ yield in uranium and should be investigated because of the possible bearing on the purity requirements of U233. The previously reported data on the new specific activity of U233 based on the new determination of the isotopic ratio are restated. Studies on extraction of thorium and uranium by a series of esters are summarized, showing the required large extraction of uranium and low extraction of thorium. A

detailed study of the protactinium extraction by diisopropyl ketone shows a rather different mechanism for extraction of the nitrate than in the case of uranium. [9, pg.449]

Katzin mentions that a program has been initiated on the study of slurries of thorium oxide in anticipation of their possible use in U233 production units. [9, pg.450]

1945-01-15 Compton wrote to Stearns to thank him for sending the report on the feasibility of the 23 converter pile. The matter has been placed in the hands of the Army with the statement that further active steps along this line will await a request from them. **Compton also mentioned that in discussing the problem with Colonel Nichols, the impression has been gained that it is unlikely that we shall receive a request to proceed further with the 23 converter pile.** Nichols has expressed himself, however, as actively interested in a research and development program that would lead to the more efficient use of uranium and thorium in producing a useful product. [9, pg.453]

1945-01-17 Compton announced that all program recommendations for next year have been submitted to him. He finds that the proposals fall into two categories: (1) essential products and services required by, *e.g.*, Sites Y and W, and (2) research and development. **With regard to the latter, Compton said that he was interested to find that three different laboratories have proposed breeder piles for U233. He stated that while we have not been authorized to carry out the program activities as proposed, the indications are that the level of support will be relatively constant and we will have a wide range for making decisions about their priority.**

There was a discussion of Compton's proposal that the research and development budget be set at 50% of the essential products and services budget. No conclusions were reached. The readjustment to a research laboratory and the need for a scientific advisory committee to stimulate and correlate fundamental research in the fields of nuclear physics and chemistry were also discussed. Compton asked Smyth and Franck to get together and formulate a formal statement on the need for a review group. That statement will be referred to General Groves.

Compton reported on a discussion with Nichols at Clinton on the possibilities of a U233 converter, based on the report prepared early this month which describes the feasibility of such a converter. Compton said that he told Nichols we prefer the breeder, whereupon Nichols said he thinks it is more important that we go ahead on the breeder than on the converter and to lay our plans along this line. [9, pg.457]

1945-01-19 In a memo to Warner, I propose that the Editorial Committee for Volume 19A of the Metallurgical Project Record, "The Production and Separation of U233," consist of Warner, Katzin, Rollefson, Spedding, Stoughton, and Seaborg (Chairman). I ask that Bertrand Goldschmidt be appointed consultant to the Editorial Committee. I also give the tentative Table of Contents of the Volume and authors of each of the eight chapters. [9, pg.460]

1945-01-24 Daniels said that Pa231 would be formed in a converter pile by an $n, 2n$ reaction on Th232. I estimated that perhaps 0.1% of the neutrons in the center of the pile would be used

up by an $n, 2n$ reaction. Near the periphery, where the neutrons are slow, the $n, 2n$ reaction would not occur. This would still be a better source of Pa231 than our present one. [9, pg.464]

1945-01-25 Katzin talked about the status of the search for U232 in U233 produced by the neutron bombardment of pure thorium carbonate. Hagemann described the work on the 4n+1 series, and Katzin gave the current situation with respect to the search for "neptunium emanation (Nn)."

It was decided that at the next meeting Ghiorso or Jaffey will discuss standard beta-particle counting geometry. It was also decided that for the next meeting we will schedule talks on (a) the search for x-rays from our 5 mg sample of U233, (b) the search for x-rays from Pa 231, (c) absorption curves derived from counts on Pa 233 beta particles, gammarays, and electrons, (d) Katzin's work on the 4n+1 series, (e) Crawford's work on Hanford neutron-irradiated plutonium.

Transfers of materials arranged at this meeting were (a) Kraus to give 5 micrograms of Pa 231 to Ghiorso for alpha particle energy measurements on his pulse analyzer, (b) **Katzin to give purified U233 to Jaffey and Crawford to look for U232 on their differential alpha-particle range chamber**, (c) Hindman to give 0.4 mg of Np237 to Florin to look for 93236, (d) a sample of Hanford neutron-irradiated plutonium prepared by O'Connor, Florin, and Simpson to be given to Jaffey and Crawford to look for 94-240 using their differential alpha-particle energy chamber. [9, pg.465]

1945-01-27 Wigner sent the following memo to Compton, with copies to the Reading File:

> Bits of news are reaching us occasionally about rumors at W concerning the validity of the advice which this Laboratory gave the Company operating W. The present letter is prompted by our hearing two bits of news of this kind within two weeks. Rumors of the kind referred to unintentionally impair not only the reputation of the Laboratory but also that of the persons involved. It puts us into an awkward position since we follow secrecy regulations and hence cannot contradict these rumors in the majority of cases.
>
> The first rumor we heard about within two weeks was that, had the Company followed our advice in connection with the number of tubes at W, the production rate would have been even lower than it is now. It is true that our original report (January 1943) called for 1700 tubes while the piles actually built contain 2004 tubes. This gives an effect of 0.2% in the multiplication constant. However, the Company intended to operate, and did operate for a long time against our oral advice, with 1500 tubes. This gave a loss of 0.1% in the multiplication constant as compared even with our original report. Furthermore, our original report called for much longer slugs which would have given not 0.2% but 0.7% rise in the multiplication constant. As late as May 1944, MUC-GY-9 and CP-1729 called attention to the waste in the multiplication constant occasioned by

the unnecessarily thick end caps. It is clear, therefore, that the rumor referred to is unfounded.

The second rumor which we heard within two weeks claimed that, had the Company followed our advice concerning the number of safety rods, it would have been very difficult to operate the pile. The exact opposite is true. The blueprints originally submitted by the Company specified 19 safety rods and this number was increased at our repeated insistence to 29 rods. Fortunately, we heard this rumor from somebody whom we know to be cleared to receive information on this point and we could set it right.

It is suggested that rumors of the above kind be officially discouraged as long as the present secrecy restrictions prevent us from answering them. [9, pg.469-470]

1945-01-30 Jones requested that Furney arrange for the transfer of 1 mg U233 from Katzin to Dempster for mass spectrographic analysis. He mentioned that the sample is approximately 97% isotopically pure. [9, pg.473]

1945-01-31 "Metallurgy Laboratory, Report for January 1945" was prepared by the Laboratory Director's office. The summary includes the following items of interest:

"A report appraising the feasibility of converting 49 into 23 was completed and transmitted to the Project Director, who in turn presented the report to Col. Nichols. The Division Directors have prepared and submitted outlines recommending plans for the work of the Laboratory beginning July 1, 1945. The final reports will be submitted during the month of February.

1945-02-01 Ghiorso described the differential alpha energy chamber which can be used on 94240, 94238/94239 mixtures, and U232/U233 mixtures; the accompanying pulse selector circuit was explained.

Activities to be considered for scheduling at next week's meeting are: (1) Search for characteristic x-rays from a 50-mg sample of U233. (2) Search for characteristic x-rays from Pa234 (and Pa233). (3) Beta, gamma, and conversion electron absorption measurements on Pa233. (4) Katzin's next run on the decay products of U233 (the 4n+1 decay series).

I received a memo from Mulliken suggesting that, in connection with the setting of editorial standards for the Metallurgical Project Record, there is an opportunity to take a few steps forward in simplifying and improving nomenclature in the field of nucleonics. He asked my reaction to the following proposals.

1. Names of radioactive families. Thorium or (4n +0) family becomes "One or Tetron-one or Polytetron-one"; uranium or (4n +2) becomes "Two or Tetron-two or Polytetron-two"; actinium or (4n +3) becomes "Three or Tetron-three or Polytetron-three."

My first impression to most of these suggestions is not very favorable.

Jones sent Furney the adjusted figure for the U233 inventory (193.6 milligrams, raised from 175.9 milligrams), based on the redetermination of the half-life from 146,000 years to 162,000 years.

The proposed program for the Metallurgical Project, 1945-46, is outlined in broader form in the Proposed Budget part of the memorandum, (b), as follows:

1. Fundamental Research
   (a) Basic laws of nuclear structure and mass-energy transformations.
   (b) Nuclear properties and the physics, chemistry, and metallurgy of fissionable isotopes, fission product isotopes, and artificial radioactive isotopes.
   (c) Nuclear properties and the physics, chemistry, and metallurgy of structural materials.
   (d) Interaction between radiation and matter and its effects on materials and on physical and chemical processes.
   (e) Fundamental studies in heat transfer, energy removal, energy utilization, and associated fields.
   (f) Basic investigation of the detection of radiation and of elementary particles.
   (g) Studies of the mechanisms of toxic poisoning of biological systems.
   (h) Investigation of the mechanisms of radiation damage to biological systems.
   (i) Use of radioactive materials for physical, chemical, metallurgical, and biological "tracer" research.
   (j) Fundamental chemical studies in connection with chemical processes.
2. Applied Research
   (a) Construction and operation of high-neutron and radiation-flux density piles of enriched "breeder" type with its necessary chemical plant.
   (b) Process design of new units and their associated chemical plants, particularly process design of "profit operation" "breeder" plants.
   (c) Consultation and process studies on power piles.
   (d) Evaluation of present Project information with respect to new military possibilities.
   (e) Evaluation of industrial possibilities.
   (f) Determination of tolerance dosage of toxic and radioactive materials.
   (g) Determination of tolerance dosage for "fast" and "slow" neutron, beta, gamma, and neutrino irradiations.
   (h) Development of tests for incipient toxicological or radioactive materials damage.
   (i) Development of tests for incipient "radiation" damage.
   (j) Development of prophylactic and curative procedures.

Daniels issued a summary of the manpower distribution in the Chemistry Division which shows the following for my section (78 men):

		Number of Men	
		Dec.	Jan.
Albaugh (Site W work, 29 men)	R. Thompson, extraction and decontamination	7	6
	Katz, concentration and isolation	3	0
	Gilbreath, process development	8	8
	Lawroski, solvent extraction	11	14
Cunningham, (Site Y work, 38 men)	Simpson, high vacuum work	6	9
	Hindman, basic chemistry	11	12
	Dawson, recovery	6	6
	Ghiorso, instruments and physical measurements	10	10
	Katzin, 23 work	8	8

1945-02-03 In response to Farrington Daniels' request, **Davidson and Katzin sent him their personal views on what they consider to be the major initial chemical research problems to be solved in the design of homogeneous, "circulated," D2O-moderated, converter or breeder piles.** The problems that they identify as being common to a majority of designs and types of structures are: (1) recovery, decontamination, and recycling of reacting material with extreme efficiency, (2) problems of the absorbing blanket, (3) general mechanical problems of homogeneous structures, and (4) general nuclear problems. [9, pg.488]

1945-02-05 The first delivery of plutonium to Los Alamos from Hanford was made last Friday.

Today the second Hanford plutonium manufacturing pile (Pile D) reached its rated power level. [9, pg.488]

1945-02-06 In a memo to Jaffey, Crawford evaluated an earlier proposal to measure the half-life of U232 indirectly by observing the rate of emission of alpha particles from such a sample over an extended period; the problem is complicated by the presence of the thoron daughter which would diffuse out of the sample and exit alpha particles under a geometry greater than the rest of the emitters in the sample. Crawford presents calculations that show that the method proposed to slow the diffusion of the thoron from the sample until it decays cannot work reliably. [9, pg.490]

1945-02-07 In view of the increasing emphasis in our section on converter pile problems involving 23, I then asked Katzin to describe some of the latest problems that have been considered. (Katzin and Davidson have been keeping in touch with the physicists, especially Wigner, on these matters.) Katzin made the following observations: (1) **At the present time the sandwich structure is not being considered very much because it is less efficient than other types. The heavy-water homogeneous pile is being more thoroughly considered.** (2) In the heavy-water pile, the evolution of D2 and O_2 gas is a serious problem at the minimum

planned power level of 100,000 kW. If nitrate is used, the problem of nitrate stability enters. (3) Solvent extraction seems to be the best process for removal of fission product poisons. (4) Other problems include the possibility of insoluble compounds settling out as a result of the 100 grams per day production of fission products and the possibility that, in the nitrate solution, the plutonium may polymerize and not solvent-extract.

Katzin also mentioned that other, non-homogeneous, types of structures are being considered. They involve running a pile at higher neutron temperatures to avoid too high a loss of neutrons in the materials of construction. [9, pg.492]

1945-02-08 At 8:30 a.m. the Heavy Isotopes Group met in my office and the meeting was attended by Florin, Ghiorso, Hagemann, Hindman, Jaffey, James, Jones, Katzin, Larson, Manning, McLane, Morgan, O'Connor, Studier, and me. La Chapelle and Magnusson came in later. Studier gave a tentative flowsheet for treating 26 mg of U233 (which has been decaying since November 18) in order to isolate the various daughter fractions for the purpose of studying the decay schemes. There was then a general discussion of the problem. O'Connor describes his and other proposed work on the sample of Hanford neutron-irradiated plutonium from Site Y sent to us for decontamination. [9, pg.493]

1945-02-13 Marian Pinckard was hired to work as a technician in Katzin's U233 group. [9, pg.496]

1945-02-16 The third Hanford plutonium manufacturing pile (Pile F) was charged with uranium for the first time yesterday. [9, pg.501]

1945-02-17 Katzin summarized for me, by memo, the work of Group 9 for my use at the Project Council Chemistry Information Meeting next Wednesday. He gives the following information: About 600 micrograms of Pa231 have been isolated from 6 kg of carbonate residues obtained from uranium extraction procedures. About 150 micrograms of this have been purified and are being used for basic chemical experiments. A larger-scale isolation from 15 kg of carbonate residues, rather richer in protactinium, has been started. A 36-inch countercurrent extraction column employing ether has been set in operation to separate U233 from irradiated thorium. The column handles a can of thorium carbonate (200 g) in five hours. About 15 cans have so far been extracted. Reinvestigation of the usefulness of calcium and magnesium nitrates as salting agents has indicated that these two salts are probably the best yet found for uranium-thorium separation by ether procedures. [9, pg.503]

1945-02-20 At 8:30 a.m. I attended the Project Council Physics Information Meeting. Items of interest reported were: (7) Dempster said he has reanalyzed the sample of U233 submitted by Katzin and finds it to contain 95.8% $\pm$ 0.4% U233. (8) Evidence has been obtained at Site Y that neutrons are emitted following fission but within 10^{-9} second after fission. (9) **Weinberg reported on breeder piles operating at higher neutron energies at which the capture-to-fission ratio would vary from that observed with thermal neutrons.** (10) Seitz talked about the graphite stored-energy problem. There appears to be some danger of an explosion due to local heating in the pile that would bring about a rapid release of the stored energy, but there seems to be no danger from this source until the stored energy in

the graphite reaches a value of 50 calories per gram. The graphite in the Hanford pile has now reached the 30 calorie per gram level and is still rising. It is hoped that it will level off because of self-healing before it reaches the 50-calorie level; if not, it perhaps will be desirable to attempt to heal the pile by controlled heating. [9, pg.506-507]

1945-02-21 General Groves is in Chicago to discuss with Compton next year's research program as proposed for the Metallurgical Project.

In discussing the U233 work, I mentioned that 600 micrograms of Pa231 have been isolated and 150 micrograms purified and used for study of the basic chemistry of protactinium. The continuous and automatic countercurrent extraction column for the ether extraction of U233 from irradiated thorium was described, as was the work on magnesium and calcium nitrates as salting-out agents for the separation of uranium and thorium. [9, pg.508]

1945-02-22 Van Winkle reported that the extraction of Pa231 (0.5-1 mg) is nearly finished; Larson has started a new extraction on 15 kg of carbonate residue.

Activities to be considered or scheduled at next week's meeting are: (1) the search for x-rays, by Ghiorso, in about 120 mg of U233; (2) the next extraction of decay products from U233 (the neptunium series); (3) discussion of the results of the Berkeley uranium plus alpha-particle bombardment; and (4) Jaffey to report on the subject of Geiger counter coincidence corrections. [9, pg.510]

1945-02-23 Manning and I reported on the Physics and Chemistry Project Council Information Meetings we attended on Tuesday and Wednesday. During the review of what was said on cross section measurements, I interpolated some predictions concerning the n, γ cross sections of the heavy elements and suggested the following classification:

Few barns (one to ten)	Many barns (~100)	
(even-even)	(even-odd)	(odd-odd)
Io230 (predicted)	U233	Pa231 (predicted)
U236	U235	Np237
Th232	Pu239	95-241 (perhaps)
U238		
Pu240 (perhaps)		

[9, pg.511]

Report CC-2636, "Chemical Research—Extraction and Properties of U233, Report for Period Ending December 15, 1944," was issued and contained the following information:

Radiation spectrum of pure U233. Studier has taken aluminum and lead absorption curves on a 16-mg sample of U233. These indicate the presence of gamma-rays of 130, 230, and 420 kev energy and an abundance of from one to ten per million alpha particles. Lower energy radiations (90, 19, 14, and 8 kev) range in abundance from about two in 10^5 alpha

particles to about one in ten alpha particles. Magnet bending experiments have demonstrated the presence of conversion electrons.

New half-life determination for U233. Hyde, Hagemann, and Katzin have carried out a new half-life determination in which the specific activity was determined by an isotopic dilution alpha-particle counting method. The isotopic ratio of the uranium sample was determined mass spectrographically by a revised method in which the isotopic ratio (U233 to U238) was brought to a value near unity. The isotopic purity of U233, according to these latest measurements is about 97%, rather than 87.5%. The alpha-particle counting results indicate a longer half-life (than hitherto assumed), namely, 162,000 years. [9, pg.513]

Determination of the decay chain of U233. Ghiorso, Hagemann, Katzin, and Studier have tentatively established the following identifications of members of the 4n+1 series: RdNp(Th229), an alpha emitter of probably several thousand years half-life; NpX(Ra225), an alpha emitter of about one month half-life; **NpC(Bi213), an alpha emitter of about 44 minute half-life**; NpD(Pb209), a beta emitter of 3.3-hour half-life, an activity which was previously known. When a sample of Ra225(NpX) is allowed to stand in an alpha counter for several hours and then removed, the counter shows signs of contamination. This is undoubtedly due to active deposit from neptunium(Em221). Attempts to determine the half-life of the emanation by following its growth in freshly purified NpX indicate that its half-life must be quite short. It is not possible to state whether the emanation is alpha or beta active. Attempts to isolate polonium or element 85 from solutions of NpX have been unsuccessful, indicating that any isotopes of these elements that are members of the chain are quite short-lived. The half-lives between NpX and NpC seem considerably shorter than ten minutes.

Extraction of U233. Hagemann, Hellman, and Studier have found an unidentified organic compound that is either an impurity or some reaction product of diethyl ether which complexes uranium and prevents its transfer from ether to an aqueous phase. [9, pg.514]

1945-02-25 The third Hanford plutonium manufacturing pile (Pile F) was placed in operation today. [9, pg.515]

1945-02-26 I received a letter written February 24 from Stoughton about his participation in the reparation of Project Volume 19A, "The Production and Separation of U233," and the corresponding collected papers volume (19B). He suggests an additional topic for Volume 19B to the nine already decided upon, "Production of (small amounts of) U233 by separation of Pa 233 from thorium after a short neutron bombardment of the latter, followed by decay of the Pa233 to U233." He also indicates that in view of his past and current work he could be-co-author of volume 19B, chapters (1) "Discovery of U233," (4) "Extraction Theory," (7) "Chemistry of Protactinium," and (10) the additional topic he has suggested. [9, pg.515]

1945-02-27 I had a phone conversation with Stoughton at Site X about the $ThOCO_3$ cans in the Clinton pile. He said 15 cans went into a good position (1.4 times average flux) on November 10 for the Canadians, another 15 cans in a similarly good position on December 3, and 14 cans in a poor position (0.3 to 0.4 times average flux) on December 8. Only these 44 cans

out of 80 passed the heating test at 230°C needed for the good position. Stoughton thinks only half of our present 125 cans will past the test. He will make this test on all our cans in order to have some ready (at least 15 are necessary for a whole replacement) in case the Canadians call for five cans to get Pa233 (as Watson told Katzin). I told Stoughton about the Canadians' primary purpose to get Pa233 [preferred material would be freshly discharged after three months irradiation] and that we might get some of their more strongly irradiated cans provided we can keep them supplied with Pa233.

Mulliken issued a "Preliminary Outline of Metallurgical Project Record," listing the titles and editorial committee for each of the 20 technical volumes and the four supplemental volumes: "Project Handbook, Project History, Patents Abstract, and the Index and List of C Reports." I am listed as being on the editorial committee for Volumes 9A and 9B, "Fission Products and Radiochemistry," (editors Coryell and Sugarman) and as the co-editor (with manning) of Volumes 14A and 14B, "Chemistry of Transuranium Elements," and (with Katzin and Stoughton) Volumes 17A and 17B, "Production and Separation of U233." The number designation of these and the other volumes have been undergoing change, but have now, presumably, assumed their final form. [9, pg.516]

1945-02-28 I wrote Whitaker about the irradiation of thorium for Evergreen and the Metallurgical Project, passing on the information from Watson that the Canadians, around the middle of March, intend to ask for four or five of the thorium cans that have been in the Clinton pile since last November or December. I point out that their interest is primarily in Pa233 and hence they would prefer freshly-discharged material with three months of irradiation. I suggested that since we have a large number of needs for U233, the remaining 10 or 11 cans from the particular stringer should be processed either here or at Clinton, and new cans be put in the pile later to supply the Evergreen people with an additional source of Pa233 and to keep up a continuous manufacture of U233 for the Metallurgical Project. [9, pg.517]

Manning sent a memo to Daniels listing the activities of Section C-I in two categories—those directly related to the development of new piles and those that are indirectly related to new piles. Under the first category are shown (1) Study of the chemical and nuclear properties of heavy isotopes [7 men]. (2) U233 and related problems [5 men]. (3) Extraction of protactinium from ores and study of its chemistry [2 men]. (4) Decontamination of plutonium from its fission products (2 men]. (5) Volatilization at high temperature of plutonium and fission products from uranium carbide [1 man]. (6) Colloidal behavior of plutonium and its fission products [1 man]. In the second category are (1) Solvent extraction methods for separation and isolation of plutonium from uranium and its fission products [17 men]. (2) Basic chemistry of plutonium [8 men]. (3) Basic chemistry of uranium (1 man]. (4) Maintenance of electronic instruments [5 men]. (5) Recovery of laboratory plutonium for re-use (5 men]. (6) Chemistry of neptunium [4 men]. (7) Chemistry of actinium [1 man].

"Metallurgical Laboratory, Report for February 1945," (MUC-JCS-209) was issued. The summary section includes the statement, "**Most of the research and development work of the Laboratory is exploratory work in connection with breeder piles.** Consideration is being given to the homogeneous thermal breeder and the heterogeneous resonance and

high energy breeder type of pile." The activities of Section C-I described in the report are excerpts of Reports CN-2688, "Chemical Research - Separation Processes for Plutonium," and CN-2689, "Chemical Research - Basic Chemistry of Plutonium," which will be issued sometime next week.

Compton received a registered letter from General Groves in Washington. I think it was somewhat of a blow to Compton, for this is what Groves wrote:

> I have given careful consideration to your proposed research program for the year 1946 which we discussed during my last visit to Chicago.
>
> Since the basic consideration for any work performed under the direction of the Manhattan District must be winning the present war, **it is necessary that the efforts of the District not be diverted in any way to post-war problems.** The Military Policy Committee has concurred in these views. The Committee is also of the opinion it should not assume responsibility for the post-war period.
>
> I am in complete agreement with the recommendation that some commercial firm be found to take over the responsibility for the operation of the Clinton Laboratories. As you point out, they must be continued in operation for the production of vital materials and to carry on research development essential to the solution of recurring problems.
>
> The work at the University of Chicago and at the Argonne must be restricted to three items:
>
> a. The solution of problems involved in the operation of the Hanford Engineer Works.
> b. The carrying of such work as may be desirable for the benefit of the Los Alamos project.
> c. **Such research as is necessary to determine the value of thorium including the possibilities of its use practically.**
>
> The work under paragraph c above must be confined to research and not extended to extensive developmental engineering until our knowledge of the problems involved is much greater than it is now or can be in the months to come.

[9, pg.518-519]

In answer to Post's question as to how much Pu240 could be tolerated in purified plutonium for Los Alamos, **I said they have no choice in the matter; whatever is produced must be tolerated.** I added that on the basis of the original purification program only about 0.001% could have been tolerated on account of its neutron emission. [9, pg.521]

1945-03-01 The following activities are scheduled: (a) the next run on 23 decay products will be made Monday, March 12; (b) a check run will be made for U233 gamma-rays and x-rays on 60 mg of U233; (c) La Chapelle will prepare a sample of 100 micrograms of Np237 for

Zinn; (d) a run will be made on the Np237(n, γ) reaction with cadmium on Thursday, March 8 or Monday, March 12.

Jones sent transfer forms to Furney for 61.5 mg of U233 as oxide to Site Y. Jones indicates that the material is ready for shipment. [9, pg.522]

The summary of the manpower distribution for the Chemistry Division that Hogness issued today shows the following for my section:

[9, pg.523]

1945-03-02 General Groves and Conant are in Chicago.

I received a March 1 memo from Dempster giving the result of the mass spectrometer comparison of the abundance of U238 in the 23 sample kept for this purpose. He finds a value for U233 of 95.8$\pm$0.5 percent. This is in agreement with two earlier approximate values. [9, pg.523]

1945-03-03 I read a copy of a March 2 memo from Daniels to Wigner summarizing the present research program in the Chemistry Division related to homogeneous water piles, listing the following investigations and manpower involved: (1) Chemical and nuclear properties of heavy isotopes - 7 men. (2) U233 - 4 men. (3) Decontamination of plutonium in absence of uranium - 3 men. (4) Colloidal behavior - 2 men. (5) Fission products - 4 men. (6) Decomposition of aqueous solutions by radiation - 6 men. (7) Solvent extraction applied to high energy piles - 4 men. (8) Protactinium - 2 men. Total, 32 men. [9, pg.523]

1945-03-06 I say we have sent 61.5 mg of isotope U233 that is 95.5% U233 and 4.5% U238 (thus 2.8 mg U238). I discuss our lans to take another more careful look at the possibility that the Th229 daughter of U233 undergoes decay by beta-particle branching; at that time we should have more to say about his interest in the possibility of analysis for U233 content in these samples by means of beta-particle counting. **I suggest we may have a better solution to this problem through some Clinton neutron-bombarded thorium carbonate that was completely purified of its natural uranium impurity before bombardment. We are now ready to extract the U233 and will make an accurate determination of its half-life.** [9, pg.528]

1945-03-07 At 8:30 a.m. I held a meeting in my office of the Council of Section C-I, attended by Albaugh, Cunningham, Davidson, Egan, Ghiorso, Gilbreath, Hindman, Jaffey, Jones, Katzin, Lawroski, Manning, Simpson, Stewart, and Roy Thompson. I opened the meeting with a reminder of the three instruments meetings scheduled for later today. **I announced that Zinn, Dancoff, *et al.*, have found eta for U233 (neutrons emitted in fission per neutron absorbed) to be greater than 2.3 and the n, γ cross section is probably less than 10 barns.** [9, pg.531]

I received a copy of a memo from Zachariasen to Mulliken suggesting that neither the term "thoride series" or "actinide series" be adopted for general use in the Project publications. He argues that it is impossible to state at this time that the term "thoride series" may not

become justifiable since, so far, valence state observations do not extend beyond plutonium. At the same time, he considers as inappropriate my term "actinide series" (I first proposed this at the July 17 meeting with C. A. Thomas on "Final Purification and Metallurgy of Product," and stated it on page 55 of the report of that meeting, CK-1968), for thorium is certainly not actinium-like. He does concede, however, that if there are nine elements following plutonium, all of which are trivalent (and not all of them tetravalent), there would be some justification for the use of "actinides" even though there is a gap in the series at thorium.

Captain Chapman requested of the District Engineer, Oak Ridge, six grams of U235 containing not more than 25% U238. He indicates that one gram is desired for the purposes I listed in my memo to Stearns last Saturday. The other five grams are desired for cross section measurements and other neutron work by the physicists, including use as a comparison standard for measuring neutrons emitted per thermal neutron absorbed by the isotope U233. [9, pg.532]

1945-03-08 At 8:30 a.m. I held a meeting in my office of the Heavy Isotopes Group, attended by Florin, Ghiorso, Hindman, Jaffey, James, Jones, Katzin, La Chapelle, Larson, Magnusson, Manning, McLane, Morgan, O'Connor, Studier, Van Winkle, and later Cunningham. James described his compilation of information on energies, Auger electron yield, etc., and L- and K- x-rays of heavy elements; and Florin talked about his progress on working up the uranium plus He 4 target (sample TotB, Berkeley bombardment). Work on x-rays of U233, Pu23 , and Np237 was discussed.

The following activities are scheduled: (a) Studier-Ghiorso run on x- and gamma-rays from 60 mg sample of U233, (b) a run about Monday, March 19, on U233 decay products, (c) La Chapelle to prepare a 100 microgram sample of Np237 for Zinn, (d) the Np237(n, γ) run with cadmium cut-off will be made next Monday. [9, pg.534]

1945-03-09 The third plutonium production pile (F) at Hanford reached its rated power yesterday. [9, pg.534]

Compton received a March 6 letter from Latimer indicating he will be very glad to receive the 1 mg of U233 that I told Connick we could transfer to the Berkeley project. [9, pg.535]

1945-03-10 Dempster wrote a memo to Stearns about some aspects of the future program of the Met Lab that should be pursued because of their obvious military importance. Stating that they represent examples of the content of such a program he lists the following:

(1) What are the possibilities of a small country developing breeder piles that could make that country an international menace in 25 years from the present time?

(2) What are the possibilities of small piles operating in various neutron velocity ranges as power sources suitable for special purposes?

(3) Is there any possibility of finding new methods of enriching naturally occurring isotopes that could be developed and used secretly by a foreign power or private organization?

(4) What methods can be used to detect unauthorized use or international traffic in special materials or secret activities in various parts of the world leading to the accumulation of "products" in the hands of aggressor nations? [9, pg.535]

The first meeting of the Laboratory Steering Committee took place in the afternoon. Those present included Bartky, Compton, Daniels, Dempster, Franck, Greninger, Hilberry, Stearns, Stone, Szilard, Wigner, and Zinn. At the meeting Wigner summarized the present knowledge concerning the various breeder piles. Szilard spoke about his concern that the first bomb that we detonate may start a race in atomic armaments between us and other nations, in particular, Russia. He believes that for the safety of this nation it is necessary for us, within the next couple of years, to produce ten times as much fissionable material as is now planned (*i.e.*, ten tons instead of one ton) in order to insure an agreement with Russia to prevent its future use in war. [9, pg.536]

1945-03-12 Szilard prepared a draft memorandum (MUC-LS-61) on the importance of the U.S. drawing up **a program for the production of ten tons of "heavy elements" within the next few years (ten times the planned amount), in order to convince the Russians that we are so advanced in the field of atomic weapons that no other nation has a chance to catch up to us.** Szilard feels that such evidence of superiority is our best chance to convince the Russians that they should agree to a system of joint control by the U.S., Great Britain, and the Soviet Union on the manufacture of "heavy elements" everywhere in the world. [9, pg.536-537]

1945-03-13 In a memo to Stearns, Daniels comments on the Steering Committee meeting of last Saturday afternoon. He urges that we begin thinking in terms of piles of a million kW in spite of the serious engineering difficulties involved. He suggests that a series of lectures on piles to which the section chiefs and a few others are invited will be helpful. He agrees with Szilard on the importance of making available, within two years, ten times as much fissionable material as has been contemplated. He then urges an actual demonstration of a high-temperature pile for the generation of power as a means of increasing our prestige with the public and our support by the government. [9, pg.537]

1945-03-14 I asked for a report on the present situation on Pa231. Katzin responded that we have 1 mg in the laboratory in a few hundred milliliters of solution and three additional batches containing 1 mg each that have gone through the first step of the semiworks isolation procedure.

Hilberry asked Stearns to arrange the transfer of the one mg of U233 to Latimer in Berkeley, provided it is possible from Stearns' point of view. [9, pg.538]

1945-03-15 At 8:30 a.m. I held a meeting in my office of the Heavy Isotopes Group, attended by Florin, Ghiorso, Hindman, James, Katzin, Larson, Manning, McLane, Morgan, O'Connor, Van Winkle, and later Magnusson. Florin described his progress in working up sample TaB (uranium plus 40 Mev alpha particles, Berkeley cyclotron bombardment), and Studier described his work on x- and gamma-rays of U233.

Activities schedules were (a) further work by Studier-Ghiorso on U233 x-rays; (b) a run on U233 decay products about Monday, March 26; (c) James to work on the plutonium plus alpha particles Berkeley target (sample 49aB) to be received next week); (d) a practice run next Wednesday to look for 94-241. [9, pg.540]

1945-03-17 Presentation schedule:

April 21	Chemical Problems	G. T. Seaborg or W. M. Manning
April 28	The Heavy Water Breeder Pile	E. P. Wigner or H. S. Brown
May 12	The 1 to 100 ev Breeder Pile	E. P. Wigner
May 26	The High Temperature Breeder Pile	
June 9	The Fast Fission Breeder Pile	L. Szilard
May 21	Uranium and Thorium	Z. Jeffries

[9, pg.543]

1945-03-20 (5) **Dancoff reported measurements of eta of U233 by comparison with U235; a value of 2.48±7% was obtained.** Langsdorf plans to determine eta of U233 by a method similar to the danger coefficient method.

Katzin wrote a summary of the work of Group 9, Section C-I, for my use at the Project Council Chemistry Information Meeting tomorrow. He states that: (1) Extractions are in progress on 60 kg of carbonate residues which may yield as much as 5 mg of Pa 231. (2) The 36-inch extraction column was used to complete the extraction of 23 cans of irradiated thorium "carbonate." **The product was specially purified and a portion used by Zinn for measurements of fission constants which indicate great promise for U233 "breeding."** (3) Electron microscope studies have been made of thoria particles in connection with slurry studies. (4) A flowsheet has been drawn up for solvent separation of uranium, protactinium, and thorium; it may prove possible to use a single solvent, diisopropyl ketone. (5) Thermodynamic data are being used to develop a theory for the mechanism of immiscible solvent extraction of uranium from nitrate solutions. [9, pg.547]

A special meeting of the Project Policy Council was held in Room 209, Eckhart Hall, from 5:05 p.m. to 6:30 p.m., attended by Chapman, Chipman, Compton, Daniels, Dempster, Doan, Eastman, Greninger, Hamilton, Harrell, Hilberry, Howe, W. C. Johnson, McKinley, Mulliken, Smyth, Spedding, Stearns, Stone, Tracy, W. Watson, Whitaker, Wigner, Wirth, Zinn, and later Franck. Compton explained that they were called together because of developments with regard to the future programs that have come up in the last few weeks. He then proceeded to read a memorandum (MUC-AC-2633) he has written to the Directors of the Argonne, Clinton, and Metallurgical Laboratories.

The memo states that the entire Metallurgical Project faces heavy cutbacks with the Laboratory at Chicago due for the greatest loss—a cut of 80% by July 1 of this year. Compton stated that the general order of preference in keeping staff is (1) retain men for certain top priority jobs at Site Y; (2) Met Lab will have first right to retain their own men;

(3) Clinton Labs, with only a slightly curtailed budget, will be next in line for taking Met Lab personnel; (4) men remaining available after Clinton and Met Lab needs have been taken care of can be transferred to other DSM projects. After that they will be available to other war projects.

Compton said that he is not too unhappy at this way of doing things as a strong group will be maintained at Clinton until the close of the war, Chicago will be able to retain certain key personnel, and Argonne can be kept operating. He explained that the Military Policy Committee and the OSRD have taken the position that the future is the responsibility of some other group. The War Department has been requested by Bush and Groves to set up a committee to consider our program, but this committee will not be able to take any action which will change the position we are now in. [9, pg.548]

1945-03-21 I gave the status of extraction of U233 from irradiated thorium and mentioned the 60 mg we turned over to Zinn for the cross section determinations reported by Dancoff at yesterday's Physics meeting.

Items of interest presented by other speakers are: (1) Perlman reported that the results of the chemical process at Hanford are very successful with production yields equaling or exceeding those obtained in small-scale experiments. Overall yield at present is 90% with prospects for improvement by another 2%. Total decontamination is 8×10^7; this is well in excess of the value of 10^7 that was the original aim. (5) **English gave a report on the problem of the composition of converter and breeder homogeneous pile solutions.** The requirements are minimum neutron absorption, stability of solution, and reasonable solubility of plutonium. [9, pg.550]

At 7:45 p.m. in Room 209, Eckhart Hall, I attended a meeting of the U233 Group (Group 9). Others present were Albaugh, Ames, Asprey, Blaedel, Bradt, Cunningham, Davidson, Dixon, Fineman, Fields, Ghiorso, Gilbreath, Greenlee, Hagemann, Hausman, Hellman, Hindman, Hyde, Hyman, Jaffey, Jones, Katzin, Krueger, La Chapelle, Larson, Manning, Morgan, Phipps, Post, Reinhardt, Robinson, Sedlet, Seifert, Schaffner, Sheft, Simpson, Stewart, Studier, R. Thompson, Van Winkle, Westrum, Winner, Wolf, and others. I announced the meeting tonight will be taken over by the U233 group. I then called on Katzin to conduct the meeting. He began by reviewing the methods for obtaining cross sections and fission constants for fissionable isotopes, illustrating the discussion by describing the measurements made on U233, including (a) the experimental determination of the U233 absorption cross section by the use of matched absorption cells, one containing pure DNO_3 and the other a solution of U233 in DNO_3, and (b) the measurement of the fission cross section by comparing the number of fissions obtained from a known weight of U233 (thin film) placed in a neutron beam with the number of fissions observed for a known weight of U235 placed in the same beam. When one uses the results of these two measurements, a value for eta of 2.49 for U233 is obtained.

Studier described some results on the measurement of the electromagnetic radiations of U233 using material that was purified by six successive ether extractions, one peroxide pre-

cipitation, and one uranyl acetate precipitation. The purified material amounted to 125 mg of which 60 mg were used for the measurements. Absorption curves were run on a modified Geiger counter, using copper, aluminum, and lead absorbers; these suggest 250, 60, 17, 12, and 8 kev components.

Hagemann described the construction and operation of the oneinch diameter, sixty-inch long ether extraction column being used for the isolation of U233 from dissolved, irradiated thorium carbonate slugs. Hellman presented a thermodynamic evaluation of the extraction of uranium into ether. [9, pg.551]

1945-03-22 Hawkins wrote to J.O. Pyle, Director of our Security Division, about the possibility that on V-E Day, celebration or civil commotion may endanger the security of the Metallurgical Laboratory. Additional guards are to be on duty at the Laboratory. He recommends that the guards be instructed that "any member of the Metallurgical Laboratory who becomes objectionable while at work, or who comes to work in an unfit condition," should be removed or prevented access to the premises. He expects that undesirable conduct may arise due to indulgence in alcohol.

English, who is visiting from Clinton Labs, and I conferred about the data from Stoughton on our sample irradiations. All samples that have been irradiated since Stoughton took over the supervision are receiving in the range of 65-90% of maximum flux; the maximum is 2.3 times the average flux. Shapiro has found that the maximum neutron flux is 3 to 4 x 10^5 neutron/sec per watt [determined by gold monitors and by the reaction Pu(n,f)Ba]. For the production of U233 and the uranium carbide samples, the flux is 80% of the maximum 3 x 10^5 nv/watt; this will apply to most future samples. For relative monitoring, silver wire is used (1 inch long, 10 mil diameter). [9, pg.553]

1945-03-23 At 8:30 a.m. I held a special meeting in my office of the Council of Section C-I which was attended by Albaugh, Cunningham, Davidson, Egan, Ghiorso, Gilbreath, Hindman, Jaffey, Jones, Katzin, Lawroski, Manning, Simpson, Stewart, and R. C. Thompson. I opened the meeting by announcing the night number of the telephone in my office has been changed to Butterfield 1405. I explained this is a prelude to what I am going to say next, which is worse. **I then announced that the Met Lab staff is to be cut by 80% on July 1. I said that the War Department is afraid to authorize any more money for any program not directly involved in fabricating the final device.** [9, pg.553]

1945-03-25 General Groves made a brief stop at the Lab today between train connections. He, Captain Chapman, Captain McKinley, Daniels, and I had a fifteen minute conference in my office. **Groves wanted to know why we have not shipped 100 mg, rather than 60 mg of U233, to Site Y. We explained that this was all we were asked to ship. We pointed out that we could get a larger supply and would plan to do so if he desires. Groves explained that he had to know the value of eta with high precision in order to make a proper decision. Zinn has reported it as being between 2.5 and 2.3, but he needs a more precise value.** We explained that one difficulty is that we do not have any highly enriched U235 with which to make a direct comparison. He suggested that we prepare larger amounts

of U233 for use at Argonne or Los Alamos to measure its fission properties in order to check the work of Zinn. He also asked us to find out from Zinn the best accuracy attainable at Argonne and to give him an answer by telephone tomorrow morning. [9, pg.556]

1945-03-26 Early in the morning Daniels and I conferred with Zinn about what can be done to meet General Groves' request for a more accurate value of eta for U233. I said we could prepare within a week, 55 additional milligrams from several laboratory sources. Furthermore, the total present stock could be increased by working up the 15 cans of thorium carbonate now in the Clinton pile, each of which contains 5 mg; this could be done within two weeks after they are delivered to us. For accurate measurements, in addition to lumping all the U233 together, it will also be necessary to make available to Argonne a minimum of 200 mg of highest purity U235. We then went to McKinley's office where Zinn talked with Groves by telephone. Groves seems to think it would be best to have the work done at Argonne. He suggests it would be advisable for Zinn and another person, probably me, to go to Site Y for a conference regarding the best means of obtaining quickly an accurate value of eta. [9, pg.557]

1945-03-27 Zinn, Daniels, and I sent a confirming memo to McKinley about yesterday's discussions with McKinley and with General Groves about the steps to be taken to make more precise measurements of the nuclear constants of U233. [9, pg.557]

1945-03-28 On the matter of manpower distribution, I said that the personnel shifts I gave last week will take place as planned except that a demand from Groves for U233 will shift Hagemann from writing reports to extracting U233 from 15 more thorium cans. I mentioned that it will probably be about two weeks before final decisions can be made on just what will happen to the Project. I added that Warren Johnson will be here from Oak Ridge next week to interview those people who might be available to go to Clinton; nothing has been heard from Los Alamos or Y-12. [9, pg.558]

Daniels asked how long it would take to determine for sure whether or not Pu240 is fissionable with slow neutrons. In response I indicated that a new uranium alpha-particle bombardment now in progress at Berkeley should provide the key to this question. The results of the bombardment should be available in six to eight weeks.

Following Daniels' comment on the tremendous importance of knowledge of the fissionability of Pu240 to the design and successful operation of breeder piles, I predicted that Pu241 will undergo a slow neutron fission and pointed out also that in the series of analogous nuclei, 91-231, 93-237, 95-241, it is known that the first two do not undergo slow neutron fission. In the case of 95-241, however, though the even number of neutrons in the nucleus is unfavorable toward fissionability, the high charge-to-mass ratio might be sufficient to cause this nucleus to undergo slow neutron fission. I emphasized that 94-241 is analogous to 92-235 and is again therefore likely to undergo slow neutron fission. [9, pg.560]

Ghiorso suggested that as these "new babies" are uncovered, I should provide cigars for the section. Davidson said that a big celebration should follow the identification of element 100.

I said that I would gladly sponsor such a celebration. Katzin remarked about the probable difficulty of separating the last three or four members of the actinide series. I countered with a statement that element 97 should follow cerium quantitatively—*i.e.*, it should have +3 and +4 oxidation states. Katzin asked whether or not europium would carry element 98, to which I replied that element 98 probably would not be chemically analogous to europium and I tabulated the rare earth and actinide series as follows:

La	Ce	Pr	Nd	61	Sm	Eu	Gd	Tb
Ac	Th	Pa	U	Np	Pu	95	96	97

I added that the resemblance between elements in the same vertical column is most marked with the general exception that the members of the actinide series, due to larger ionic radius, are more easily oxidized to higher oxidation states than the corresponding rare earths. [9, pg.561]

1945-03-29 The following activities are scheduled: (1) Jaffey and O. Van Winkle to make runs on U233 decay products and on Pa231(n, γ)Pa232 this week. [9, pg.561]

Zinn called me about a teletype he received from Site Y, stating the half-life of U233 to be 167,000 years. The 61-mg sample of U233 will be returned here from Site Y tomorrow morning. Fermi made his measurements at Site Y on pressed U233 oxide powder in the form of a plaque. **He determined the neutron absorption cross section to be 560 barns (compared with 513) and the fission cross section to be 475 barns.**

Daniels, Hilberry, and I conferred about arrangements to get the 15 cans of irradiated thorium carbonate from Clinton so we can start as soon as possible to get the additional U233 to meet General Groves demand for a more accurate neutron absorption cross section. Since Captain McKinley has not yet made arrangements to get the 15 cans of neutron-irradiated thorium carbonate from Clinton, Hilberry phoned Whitaker and asked that the cans be shipped at once. We have started to assemble all of the U233 available in our section. [9, pg.562]

1945-03-30 Daniels called me about a call from Tom Jones that the 15 thorium carbonate cans pushed from the pile this morning are exceedlingly hot and he wondered if they could be handled by the technicians for immediate shipment to us. I assured Daniels that the material will cool very quickly. I also conferred with Dr. Rose in the Health Division and convinced him that we are prepared to handle the hot material as soon as it gets here. [9, pg.564]

1945-04-02 Group 9 - 23 Work
Leonard I. Katzin - Assistant Section Chief
French T. Hagemann - Research Associate
Nison N. Hellman - Research Associate
Raymond G. Larson - Research Associate
Martin H. Studier - Research Associate
Michael Wolf - Research Associate
Earl K. Hyde - Research Associate

Quentin Van Winkle - Research Assistant
Marian Pinckard - Technician

No group assignment - Protactinium Work
Roy C. Thompson - Research Associate
Eugene Hausman - Research Assistant [SED] (half-time)
Bernard B. Brody - Research Assistant
John G. Malm - Research Assistant [9, pg.567]

Number of Men Feb. Mar. Albaugh Thompson, extraction and 6 6 (Separation decontamination Processes, Gilbreath, process development 7 7 27 men) Lawroski, solvent extraction 13 12

Cunningham Simpson, high vacuum work 9 9 (Basic Chemistry, Hindman, basic chemistry 11 11 36 men) Stewart, recovery 6 6 Ghiorso, instruments & physical 9 9 measurements

Katzin 23 work 7 7 (23 Work, 8 men)

Administration Seaborg, Manning, Jones, 6 6 Albaugh, Cunningham, and Katzin 74 73

Word has reached us that construction of the Hanford Engineer Works is completed and it is now engaged in total operation. Yields of plutonium in the March runs in the 200-T Area continue to be very satisfactory. The losses in the canyon (221 Building) average about 3%, in the crossover cycle (224 Building), about 4%, with an overall decontamination factor of about 3.4×10^7. The plutonium peroxide isolation procedure in the 231 Building has operated very satisfactorily. [9, pg.568]

1945-04-03 Fifteen cans of irradiated thorium carbonate were received from Clinton Labs to be processed here on an urgent basis in order to recover the U233 for the measurements General Groves wants. [9, pg.568]

1945-04-09 Daniels had a conference with me and other members of our section to go over the schedule for the production of U233. Including the hoped-for 70 mg in the material now being recovered from the 15 cans of irradiated thorium carbonate received from Clinton last Tuesday, we expect we can complete the delivery of 200 mg this coming Thursday. This is ahead of the tight schedule promised General Groves two weeks ago. [9, pg.574]

1945-04-11 At 8:30 a.m. I held a meeting in my office of the Council of Section C-I, attended by Albaugh, Cunningham, Davidson, Egan, Ghiorso, Gilbreath, Hindman, Jaffey, Jones, Katzin, Lawroski, Manning, Simpson, Stewart, and R. C. Thompson. **I opened the meeting by giving a list of reports and notebooks we cannot trace. I then announced that we will have to turn our entire stock of U233 into various forms for Zinn to enable him to determine eta for General Groves before April 26, the date of the San Francisco conference.** On the future of the Plutonium Project research program, I emphasized the importance of this determination by stating that everyone in Section C-I must be available for helping on it. Since a portion might be in metallic form, Florin will practice the HF dry

fluoride method for making the UF_4 needed for metal production. I added that the possibility of making wet UF_4 should not be overlooked as there is the possibility that the dry method may cause some loss of material. This, of course, is something that cannot be allowed to happen. I suggested Howland might be a good man for making the wet UF_4. I said that Fried and Westrum would work on the actual production of the metal. Depleted U238 oxide is to be used for practice runs since if we do get metal from these runs, Zinn probably can use it as a blank. [9, pg.576]

Katzin: (1) Isolation of pure Th229 and weighing [determination of half-life]. (2) Cooperation in the measurement of the slow neutron fission cross section of Th229. (3) Determine the n, γ cross section of Pa233. This is a toughy. (4) Isolation of Pa231 and chemistry of protactinium. (5) Continue work on neptunium (4n+1) decay series. (6) Investigate transmutation produced upon bombarding U233 with alpha particles and deuterons. [9, pg.577]

Van Winkle made a partial report on work he and Jaffey have done on an absorption curve on a very pure gamma-ray from Pa232 formed from neutron capture by Pa231. Ad-libbing until Jaffey arrived, I reviewed the purpose of the measurement of the cross section for the reaction Pa231(n, γ)Pa232. The thorium scheme [Th232(n,2n)Th231 -> Pa 231] is analogous to the U238(n,2n) scheme, even to close agreement in half lives and energies. It is Jaffey's problem to establish even further analogy by measuring the neutron absorption cross section on Pa231 which differs in mass from Np237 by exactly six.

Jaffey apologized for being late, explaining that he lost a factor of 100 in a calculation he was making for this meeting and had to redo all of them. He presented the equations and calculation used to arrive at the neutron cross section for Pa231 of 290 barns. He said that, as in the case of Np237, this cross section might account for all the U232 formed, which experimentally is found to have about the same activity ratio to U233 as the Pu238 has to the Pu239. [9, pg.578]

1945-04-12 We completed the isolation of the 200 mg of U233 today and turned it over to Zinn for measurement of eta at the request of General Groves.

President Roosevelt died at Hot Springs, Georgia, at 3:35 this afternoon. I first learned of this tragic event from Howard Lange who came by to tell me after hearing the news on his radio. All of our people are in a state of shock. The major radio stations turned their attention to this news for the remainder of the afternoon and throughout the evening. President Harry S. Truman was sworn into office about six o'clock (EST) this evening. [9, pg.580]

1945-04-13 In a memo to Compton on the significance of power piles in the research program, Daniels gives the opinion that it is unnecessary to regard power piles as limited to the distant future and that it will be possible to run a battleship or light a city with power generated by a pile within a year or two, or as soon as fifty or more kilograms of plutonium become, available as a starting material. [9, pg.580]

1945-04-16 Katzin wrote a summary of the work of Group 9, Section C-I, for my use at the Project Council Chemistry Information Meeting. It contains the following information:

The 36-inch counterflow extraction column has been rebuilt to give three times as great flow rates and used to process 20 additional cans of material which were irradiated at Site X at the 4000 kW level for several months. In addition, six cans of material from an earlier irradiation have been extracted, all within about one week. The activity level of each "hot" can was some 40-50 curies of Pa233 beta-particle activity, with approximately half that amount of gamma-ray activity.

The U233 obtained from the series of extractions, together with other material on hand, was prepared for measurements of physical constants of U233 by Zinn. The work included purification and determination of accurate specific activities, as well as making fission foils, making oxide powder for powder experiments and loading the cells and making solutions in "deuterium nitrate" for experiments with the velocity selector apparatus. Similar preparations were made of highly enriched U235 and highly depleted U238.

Solvents other than heptyl alcohol found suitable for protactinium extraction are normal octyl alcohol and 2-ethyl hexyl alcohol. **Experiments show that the slow neutron capture cross section for Pa231 is very high, around 290 barns, indicating that thorium used for production of U233 must be very free of Pa231 in order to avoid the formation of excessive amounts of U232. It reaffirms the possibility of forming U232 in even pure thorium, if there are sufficient fast neutrons present to produce Pa231 via the n,2n reaction on thorium.** [9, pg.584]

1945-04-17 (5) Zinn reported on the continued work on cross sections at the resonance peaks of some of the rare earths. He discussed the measurements on the value eta for U using the 200 mg our group has isolated. A value of 2.35 for eta was reported to Groves. (7) Zachariasen spoke about the effect of irradiation on Hanford pile graphite. (8) Lewis discussed fission product analysis by means of the mass spectrometer. This offers a powerful means of making mass assignments. (9) **Weinberg reported on calculations related to plutonium piles that use neutrons of intermediate energy. He believes eta becomes 2.7 at just about 0.4 ev. Under such conditions, breeder operation is possible.** (10) Friedman discussed the problem of U234 accumulation in U233 breeder piles. [9, pg.586]

1945-04-18 I mentioned our unsuccessful search for short-lived beta-particle emitting isotopes of plutonium, the Pa231 cross section work, oxidation-reduction potentials of Np III/IV and V/VI, plutonium chemistry studies of PuOBr and our chemical extractions that have provided 200 mg of U233 for Zinn's "thick solution" experiments and will provide an additional 100 mg so that Langsdorf will have 300 mg for his pile poisoning experiments to determine directly eta. [9, pg.587]

Items of interest presented by other speakers were as follows: (4) **English spoke on the solubility of uranium and plutonium salts for homogeneous piles. Stoughton's calculations on the Pa233 n, γ cross section were also reviewed.** (8) Burton discussed the graphite problem in operating piles. It has been found that an energy storage amounting to 80 cal/g could be removed by heating to 1000°C, whereas the total energy storage was 102 cal/g.

He said Zachariasen has found a 1% increase in lattice spacing of irradiated graphite. [9, pg.587-588]

At 7:45 p.m. in Room 209, Eckhart Hall, I attended a meeting of Sub-section 1 of our section: The meeting was devoted to a description of the high temperature pile proposed by Daniels and intended primarily as a power source. Daniels described the structure to us as a perforated cylindrical refractory block, about 5'7" in diameter and 5'7" high, fabricated out of a homogeneous mixture of 99.6% BeO and 0.4% UO_2 (the latter being the oxide of U233) surrounded by a thorium blanket. The best coolants would appear to be steam or bismuth. The unit would require 30 kg of U233 to start and would operate at 100,000 kW and at a temperature perhaps as high as 2000°C and burn some 100 gm of U233 per day. [9, pg.588]

1945-04-21 In the afternoon there was a Group Leaders' meeting on New Piles in Room 251, Ryerson Laboratory, at which Katzin and Blaedel spoke on the use of solvent extraction to purify the fissionable fuel material from poisonous fission products on a repetitive basis in high intensity piles. **Katzin reviewed the problems of purification of the pile and blanket materials in U233 converter or breeder piles, and Blaedel described the purification of plutonium fuels from homogeneous or heterogeneous pile systems.** [9, pg.591]

1945-04-25 I commented that the Army will get us 5 mg of protactinium at $100 per mg. This is pretty cheap. It should arrive in two days, and Katzin will have two days to analyze it. Katzin said that most of one barrel of our own protactinium material has been through the semiworks and is now being worked up in the lab. This should yield 4 to 6 milligrams. Roy Thompson stated that the semiworks should finish the two remaining batches in less than two months. [9, pg.593]

1945-04-26 Hindman reviewed his run on the specific activity of 90 gt and 35 gt plutonium which is nearly finished. Hagemann described his work on the U233 decay products. There is evidence that the 30-day half-life alpha-particle emitter is due to actinium, which is followed by two alpha particles with a 5-minute half-life, then by a 40-minute half-life, then by a 40-minute alpha particle. The 5-minute alpha-particle half-life seems to be due to an emanation. Peterson described his work on the chemical fractionation of actinium and rare earths.

Stearns sent authorization to Hogness for the transfer of 2 mg of 50% U233 to the Area Engineer for shipment to Y-12. [9, pg.597]

1945-04-27 I sent a memo to Hogness stating that my U233 group has been operating almost exclusively as a service group since resumption of the U233 program last year. I point out the desirability of our keeping, over a period of time, a substantial fraction of the U233 supply to enable us to undertake a program of chemical research which has barely gotten underway. [9, pg.598]

1945-04-29 Mussolini has been killed in Milan, the birthplace of Italian fascism. [9, pg.600]

1945-04-30 Hitler committed suicide today. [9, pg.600]

Stoughton, who is in Chicago, and I conferred about the thorium carbonate cans. I learned that: (1) Only 3 out of 25 of our thorium carbonate cans passed the heat test of four days at 230°C. All 125 cans passed for 5-6 days at 175°C. These three plus three others went into the "Montreal" stringer (best position: center) about a month ago and are probably still in. (2) About 100 cans went in last week in a position about one-third of the maximum flux—a nearly average position. (3) The maximum flux is 1.5 times the average (according to the chemists), or 2.4 times the average (according to the physicists). [9, pg.601]

Captain J. H. McKinley, Area Engineer's Office, issued the following instructions for SED men today:

1. All enlisted men are advised that upon announcement of the capitulation of Germany, no celebration will be held at this installation and no time off will be granted therefore.

2. You are further advised that higher authority has directed that upon announcement of the capitulation of Germany, all military personnel are to remain off the streets of towns and cities, and out of public places to the extent possible. However all are expected to be on the job as usual. These restrictions to go into effect upon announcement of the capitulation of Germany and to remain in effect until contramanded by orders. Orders lifting restrictions will be issued immediately where circumstances warrant such action. This is necessary to avoid being involved unwillingly or otherwise in any situation which might result from celebrations which may occur among the civilian population.

3. All are cautioned that you are expected to set an example of sobriety and industry and to foster a "stay-on-the-job" attitude. Any willful deviation from the policy as set forth above will result in disciplinary action. [9, pg.601-602]

E Extracts from *History of Met Lab Section C-I, Volume 4*

1945-05-02 Katzin made a few introductory remarks on our recent "mass production" program for U233. In spite of the magnitude of the task involved, the deadline was met and the material delivered for physical measurements. **Katzin noted that the determination of eta by the physicists using this material has resulted in conflicting values. The earlier value of 2.5 has not been confirmed, and the present best estimate seems to be 2.35.** This is about the same value as was determined in the first rough measurement, and Katzin said that this illustrates again the action of the "Ghiorso principle," to wit, "the value obtained in a preliminary rough measurement will always turn out to be the best value." [10, pg.7]

1945-05-05 I talked at the afternoon meeting of group leaders in Room 251, Ryerson Laboratory, concluding the discussion of the effect of heavy isotopes on the operation and products of converter and breeder piles, which I had begun at the last Saturday's meeting. I considered two types of piles: (1) those operating on enriched U235, and (2) those operating on U233. With regard to the U235 pile, I pointed out that all the reactions occur that were described last week for the plutonium-fueled pile. In the later stages of the reactions, Pu238 is produced faster in a U235-U238 mixture than in a pile containing Pu239 and no U238; assuming that the pile is operated until the Pu240 produced is 20 percent of the Pu239, the alpha activity from the Pu238 is equal to about 10 percent of the alpha activity of the Pu239 and the alpha activity due to Pu240 is nearly equal to the alpha activity due to the Pu239. I mentioned that Pu238 would also be produced even faster in later stages of operation from the U236 formed by the n, γ reaction on U235. The formation of Pu236 was also discussed.

In considering the breeder pile operating on U233 to produce more U233 from thorium, I discussed the production of U234 from U233 and Pa233. I mentioned the importance of measuring the cross section for the reaction Pa233(n, γ)Pa234 in order to determine the efficiency loss in eta from the U234 decay product of Pa234. [10, pg.10-11]

1945-05-08 Today is V-E Day. Germany surrendered unconditionally today at 2:41 a.m. French time in the red Reims schoolhouse—General Eisenhower's headquarters. [10, pg.13]

1945-05-14 Katzin sent me a summary of the work of Group 9, Section C-I for my use at the Project Council Chemistry Information Meeting of May 16, 1945. It covers the following topics: (1) The neutron capture cross section of ionium by the reaction Th230(n, γ)Th231 was determined by irradiation of a sample of ionium (prepared at Berkeley) in the thermal column of the Argonne pile. (2) A redetermination of the radiations from Pa233 disproves the existence of a gamma-ray of 1.5 Mev making up as much as 2 percent of the radiation. Present evidence is that there is a maximum of 0.1 percent of any such hard radiation. (3) Measurements on a sample of 85 percent U235 disclosed a gamma-ray of 170 kev energy whose abundance is very high and most likely is due to the U234 present at 0.5 percent by weight. (4) A new procedure has been worked out for the recovery of Pa231 from uranium residues (using the precipitate of the carbonate neutralization step). The first step is treatment with strong caustic to remove the soluble silica which previously caused trouble. [10, pg.21]

1945-05-15 (4) Spedding said he has used U232-U238 mixtures and has obtained a 15 percent separation in one pass. (I remarked that he was probably separating uranium from the U232 descendants which are alpha-emitters and present in rather large amounts.) (5) Maurer discussed the heat transfer problem in the breeder and converter piles. (6) Soodak reported on considerations about the breeder piles and discussed the possibility of using very high energy neutrons to take advantage of the increase in eta. (10) Zinn discussed Langsdorf's method for measuring eta by its swing effect. [10, pg.23]

1945-05-16 I also discussed in some detail our U233 work, recalling that about six months ago a decay scheme was suggested which included a gaseous emanation of mass number 221 (after the first alpha emitters). I explained that we have returned to the study after interruptions caused by other work and have now found evidence that Ra225 emits beta rays (rather than alpha rays as previously postulated); the following chain of intermediates is now suggested between Ra225 and Bi213.

$$_{88}\mathrm{Ra}^{225} \xrightarrow[26d]{\beta} {}_{89}\mathrm{Ac}^{225} \xrightarrow[10d]{\alpha} 87^{221} \xrightarrow[5m]{\alpha} 85^{217} \xrightarrow[<1m]{\alpha} (_{83}\mathrm{Bi})^{213}$$

I mentioned that the identification of elements 87 and 85 in this decay series is made somewhat uncertain by the fact that the 5-minute alpha activity can be volatilized from the LaF3-covered counting plate by heating to red heat (this helped to mislead us earlier into believing that a gaseous emanation was formed). [10, pg.25]

I next introduced Jaffey who gave the results of a neutron bombardment of ionium. The sample, which was 25 percent Th230 and 75 percent Th232, was bombarded alongside a sample of ordinary thorium and two gold monitors. The cross section of ionium was calculated to be 88 barns. I suggested the possibility of measuring the ratio of U233 to Pa231 formed in bombarding such a sample, saying it would be desirable to put a milligram of this ionium-thorium mixture in the Clinton pile for an extended bombardment. [10, pg.27]

1945-05-19 I sent Joe Hamilton, at his request, a description of our best method for separating protactinium from thorium, uranium, and fission products. I also tell him that we have prepared a solution of about three micrograms of Pa231 for him which we will send as soon as we learn the mechanism for doing so. [10, pg.30-31]

1945-05-23 At 7:45 pm in Room 209, Eckhart Hall, I attended a meeting of the U233 Group. Others present were Ader, Asprey, Egan, Florin, Ghiorso, Gilbreath, Hagemann, Hindman, Hyde, Jaffey, Jones, Katzin, Larson, Manning, Morgan, Peterson, Phipps, Reinhardt, Simpson, Stewart, Studier, Roy Thompson, Van Winkle, and others. I opened the meeting and turned it over to Katzin to preside.

Studier, the first speaker, reported on absorption studies of the radiations from enriched U235 and from Pa233. In the U235 sample he has found a 170 keV gamma-ray of yield exceeding the number of alpha disintegrations of U235 present in the sample. It is suggested that the gamma-ray may originate with the U234 present which has a much higher specific activity.

The examination of the Pa233 sample did not show the 1.5 MeV component that had been reported by Stoughton. There seems to be at least seven components, in addition to the alpha-ray component, in the radiation spectrum of U233.

Hagemann discussed some further work on the decay products of U233, first reviewing the work of six months ago which resulted in a tentative scheme involving alpha decay of Ra225 to an alpha-emitting emanation. Work was discontinued to allow time for a new sample of U233 to decay and improve the carrying chemistry involved. The pressure of other more urgent assignments prevented resumption of the work until very recently. A few weeks ago word was received that the Montreal group had found Ra225 to be a beta-emitter. The resulting Ac225 decayed by successive alpha emissions through 87-221 and 85-213. At that time work was resumed here and resulted in confirmation of the following:

(paste decay chain picture here, page 34)

Hagemann then described the experimental procedure followed to isolate the decay products and measure the yields. [10, pg.33-34]

1945-05-28 I also received and read Farrington Daniels' response to Mulliken's request. He generally favors publication for reasons such as more rapid advances in nucleonics and showing the American public that there is an absolute necessity for the elimination of war. **He does say, however, that if an armament race appears inevitable, we should change our policies and keep secret details on how to build a pile or a weapon, use such secret information as a bargaining power to force international cooperation and control**, and recognize that we need, under such circumstances, a vigorous research program. [10, pg.37]

1945-05-29 As a result of Mulliken's request, Nickson wrote a memo to Jacobson today in which he states:

> Therefore it seems to me that one of the most important functions at hand is the education of the general public to the fact that the era of atomic power is at hand. Further, the implications of this fact must become general knowledge. If the public, as a whole, is not entirely conversant with the broad implications of the field of nuclear physics, it seems reasonably certain that the public will not accept any organization which attempts to take into account the implications of these facts. [10, pg.38]

1945-06-06 I brought up the subject of recommendations on the scientific, political, and social future of our project in atomic power, pointing out that I am on a Committee on Social and Political Implications. After summarizing the general statement prepared by the local committee about the political aspects of atomic power, I asked the opinions of the men about the following possible alternatives in the use of the atomic bomb: (1) not use the weapon in this war but simply announce at the end of the war that we have it; (2) tell the world immediately that we have it and give a demonstration at which representatives of all

countries, including Japan, are invited to see it; (3) make bombs as fast as possible and use them on Japan. I added that the committee's opinion is to follow the second procedure.

In the discussion which followed, one man said he would like to see no. 3 used on the basis that we should treat it as just another weapon. I asked if there was not a certain moral aspect that we must consider. **If we are the first to use the weapon and then want to build up a world organization that would never allow its use again, our position would be very much weakened.** I also mentioned that it might be important to give a demonstration from the point of view of keeping our allies' good will; this would be particularly important in the case of Russia who is our most likely future antagonist in view of Russia's great future potentialities as an industrial power.

The division of opinion of the people present was mostly between the possibilities of no. 2 and no. 3. Most people agreed that a strong international organization is ncesssary but that some precaution must be taken to prevent capturing the whole available supply of fissionable material by that organization. Some expressed pessimism to the possibility of forming a sufficiently strong organization to control the problem, but everyone agreed that it was essential to make the effort. [10, pg.51-52]

1945-06-07 Hagemann described Th229 and Th228 from the U233-U232 mixture. [10, pg.54]

1945-06-08 In fulfillment of my responsibilities as a member of the Program Committee to prepare recommendations for use for the scientific panel advising the Interim Committee, I prepared and sent to Zinn and Cole a ten-page summary of the chemical aspects of postwar research and industrial development. My proposed program encompasses eight areas (1) general investigation of the heavy elements, (2) pile investigations, (3) isotope separation, (4) military uses, (5) methods of analysis and instruments, (6) use of tracers, (7) applications of fissionable material to useful atomic power and other purposes, (8) fundamental chemistry.

With regard to (1), I propose preparation of all conceivable heavy isotopes by bombarding the following elements with positive ions such as protons, deuterons, etc.: Th230, Th232, Pa231, U233, U234, U235, U236, U238, Pu239, Pu240, 95-241, etc. I also suggest preparation of all conceivable heavy isotopes by intense neutron bombardment in piles of such elements as Pu239, Pu240, Pu241, Np237, 95-241, 96-241, 96-242. I call for determination of nuclear properties of individual isotopes and the basic chemical and physical properties of elements 89 to 96. I also ask for the development of natural sources—new and more efficient means for extraction of uranium and thorium from ores, the recovery of radium, protactinium, ionium, actinium, mesothorium, etc., from ores, and a search for new isotopes of transuranic elements in nature. Under "pile investigations," I cover construction and operation of piles, converter piles, breeder piles, and other new piles. Under "isotope separation," I propose chemical development connected with the electromagnetic method, the diffusion method, and the thermal diffusion method. I also suggest the isotopic separation of elements other than uranium using the electromagnetic method, and propose the investigation of new

methods for the separation of U235 including solvent extraction and adsorption methods, competing chemical reactions, fractional electrolysis, and centrifuge methods. [10, pg.55]

1945-06-11 The report of the Committee on Political and Social Problems [Franck (chairman), Hughes, Nickson, Rabinowitch, Seaborg, Stearns, and Szilard] is being issued today. It consists of I. Preamble, II. Prospects of Armament Race, III. Prospects of Agreement, and IV. Methods of International Control. I believe that it is a significant report, and I quote the Summary:

> The development of nuclear power not only constitutes an important addition to the technological and military power of the United States but also creates grave political and economic problems for the future of this country.
>
> **Nuclear bombs cannot possibly remain a "secret weapon" at the exclusive disposal of this country for more than a few years.** The scientific facts on which their construction is based are well known to scientists of other countries. Unless an effective international control of nuclear explosives is instituted, a race for nuclear armaments is certain to ensue following the first revelation of our possession of nuclear weapons to the world. Within ten years other countries may have bombs, each of which, weighing less than a ton, could destroy an urban area of more than ten square miles. In the war to such an armaments race is likely to lead, the United States, with its agglomeration of population and industry in comparatively few metropolitan districts, will be at a disadvantage compared to nations whose population and industry are scattered over large areas.
>
> **We believe that these considerations make the use of nuclear bombs for an early unannounced attack on Japan inadvisable.** If the United States were to be the first to release this new means of indiscriminate destruction upon mankind, she would sacrifice public support throughout the world, precipitate the race for armaments, and prejudice the possibility of reaching an international agreement on the future control of such weapons.
>
> Much more favorable conditions for the eventual achievement of such an agreement could be created if nuclear bombs were first revealed to the world by a demonstration in an appropriately selected uninhabited area.
>
> In case chances for the establishment of an effective international control of nuclear weapons should have to be considered slight at the present time, then not only the use of these weapons against Japan, but even their early demonstration, may be contrary to the interests of this country. A postponement of such a demonstration will have in this case the advantage of delaying the beginning of the nuclear armaments race as long as possible. If, during the time gained, ample support can be made available for further development of the field in this country, the postponement will substantially increase the lead which we have established during the present war, and our position in an armament race or in any later attempt at international agreement would thus be strengthened.

On the other hand, if no adequate public support for the development of nucleonics will be available without a demonstration, the postponement of the latter may be deemed inadvisable, because enough information might leak out to cause other nations to start the armament race, in which we would then be at a disadvantage. There is also the possibility that the distrust of other nations may be aroused if they know that we are conducting a development under cover of secrecy, and that this will make it more difficult eventually to reach an agreement with them.

If the government should decide in favor of an early demonstration of nuclear weapons, it will then have the possibility of taking into account the public opinion of this country and of the other nations before deciding whether these weapons should be used in the war against Japan. In this way, other nations may assume a share of responsibility for such a fateful decision.

To sum up, we urge that the use of nuclear bombs in this war be considered as a problem of long-range national policy rather than of military expediency, and that this policy be directed primarily to the achievement of an agreement permitting an effective international control of the means of nuclear warfare.

The vital importance of such a control for our country is obvious from the fact that the only effective alternative method of protecting this country appears to be a dispersal of our major cities and essential industries. [10, pg.57-59]

1945-06-13 I sent Ernest Lawrence at Berkeley my opinions and suggestions on the course to be taken for nuclear weapons in the immediate future and on the question of the postwar future for nucleonics. I indicate that the opinions I express are shared almost unanimously by the people associated with me in Section C-I of the Chemistry Division here. My major points are:

(1) The basic facts concerning the successful release of nuclear energy and its immense destructive possibilities should be made public very soon, both to the general public and to scientific channels. I propose withholding only information with respect to the actual detailed designs of the major manufacturing installations.

(2) With respect to use in the present war, we propose not to use the weapon on Japan without warning but to demonstrate the weapon in the presence of all leading countries including Japan.

(3) On postwar control, I indicate we favor free research in nucleonics throughout the world and control through an international organization. Probably the best method of control lies in the control of the raw materials. Perhaps the only method of maintaining these controls would involve world-wide pooling to form a stockpile of fissionable material to be used by the international organization for policing purposes. **I mention that, as suggested by Szilard, perhaps control could be effected by denaturization, *i.e.*, by mixing it with suitable isotopes**

to spoil its use for explosive purposes without interfering too much with its use for research purposes such as power pile developments.

(4) With respect to the organization of postwar research in nucleonics in this country, I state that it would be a good idea to establish, with government aid, about four large research laboratories at four of the major universities. These laboratories should form a sort of a foundation for the country's research program and should include men who are able and willing to advise outlying laboratories as to the research program. The outlying laboratories might consist of government laboratories working on the more practical aspects of the field, and also regular university and industrial laboratories supported by government contracts or grants-in-aid. [10, pg.62]

1945-06-16 I received and read the final report dated June 11 of the Research Program Committee (Daniels, Wigner, Seaborg, Jacobson, Cole, Howe, Zinn). The introduction reads:

"The field of nucleonics has reached the stage where it is fairly clear as to what should be the next immediate steps. The section of this report dealing with the social and political problems indicates several choices which face the nation concerning the future of this field. It is believed that irrespective of the choice made and irrespective of whether or not the point of view is one which envisages an armament race or is one which happily contemplates a peaceful development, the immediate problem is unaltered. The time scale, however, on which the work is carried out will be very much affected by the realities of international policies.

"The quantity production of fissionable isotope is now realized only at the expense of a raw material which is not at all abundant in our country and exists in rather limited quantities in the world as a whole. At the present rate of destruction of this material it cannot be imagined that a wide-spread use of fissionable isotope will be permitted. The wasteful utilization of this raw material is forced upon us by the necessities of war, but the advisability of continuing present methods of production far into the future even for military needs can be doubted. It follows that research directed towards a more efficient utilization of the primary material is the main problem of the nucleonics program.

"**If a sizeable percentage of the isotope uranium-238 could be burned and if, in addition, useful fissionable isotope could be realized from thorium, then the whole future of the development would be on much more secure grounds.** It is of paramount interest, therefore, that the fundamental research necessary to the design and construction of "breeder" piles be undertaken vigorously. It is only by demonstrating the practicability of the "breeder" principle that a sufficiently ample supply of fissionable material can be produced to permit the nucleonics program to proceed on the scale indicated by the benefits to be derived. It is also necessary that research and development leading to the useful utilization of the power from nuclear burning be vigorously supported. The ultimate goal

here would be to operate machines which produce useful power and which at the same time produce as much or more fissionable isotope than is consumed. Under these circumstances no real limit could exist to the application of nucleonics to all phases of our scientific and industrial life.

"The accomplishment of the goal set in the previous paragraph is not possible without an extensive program for basic research in physics, chemistry, and metallurgy. The present state of the art is such that new designs of reactor units invariably suggest situations about which no information is available. For the long term development of the field the research in the basic sciences will be of more importance than any immediate pile design or engineering. It is possible that the real future of atomic power does not involve the burning of uranium but rather other elements at the light end of the periodic table. Progress in this direction can only come if all possible freedom and support is given for investigations concerning the nucleus." [10, pg.65-66]

1945-06-19 At 9:00 am I attended a Chicago conference on new piles. The following talks were given: (1) Eugene Wigner on the fast neutron uranium-plutonium breeder, (2) Nordheim on the heavy water-thorium-U233 breeder, (3) Weinberg on resonance neutron uranium-Pu239 breeder, moderated by such coolants as fluorocarbons, (4) Fermi on the uranium-Pu239 fast breeder system, (5) Soodak on the uranium-Pu239 fast breeder system, (6) Daniels on a version of his high temperature pile. [10, pg.68-69]

1945-06-25 Stoughton is here from Clinton and will stay until June 29. [10, pg.75]

1945-06-27 I explained that we have to keep records on Section C-I's material. Katzin, who takes care of the U233 and thorium, should give Stewart a record of the U233; Hindman will inventory the neptunium; Gilbreath will handle uranium; Roy Thompson will keep track of the protactinium; Stewart will maintain records for the Q material (depleted uranium), U235, and special tracers. I added that Stewart will be authorized to send materials out for bombardment. [10, pg.77]

Stearns issued a memo to Division Directors and Section Chiefs stating that letters, reports, and documents containing the numerical values of nu and eta for U233 will be classified "secret" but will be handled in the same manner as if they were classified "top secret" when transmitted from one site to another. Numerical values of the nuclear physical constrants of thorium and U233 and their relative importance, as compared with other elements, are to be made available only to those persons who are directly concerned with their application. [10, pg.79]

1945-06-30 I prepared an account of the discovery and early study of U233 for the use of William L. Laurence of the New York Times who has been brought into the Project by General Groves to represent the news media. [10, pg.83]

1945-07-02 I received a letter dated June 27, from Hamilton in Berkeley. He enclosed a day-by-day log of the bombardments of uranium metal targets with helium ions.

He also told me the U238 as U3O8 went on that afternoon (June 27) and if everything goes well, the U235 target would be put on sometime Friday, June 29, and the thorium target on Monday morning, July 2. The three samples will then be put aboard the streamliner Monday afternoon (today), and are scheduled to arrive in Chicago on Wednesday morning. [10, pg.86]

1945-07-03 Ghiorso reported on the fission cross section for Th229. He told us that Hagemann has given him about 5,000 counts of Th229 and that the number of fissions in the sample corresponds to 5 c/m of U233 impurity. He indicated that, if there is no impurity, the fission cross section of Th229 is only 1/10 that of U233. [10, pg.90]

1945-07-07 I received a letter from Bill Watson in Montreal, who asked for additional information about the 10 mg sample of U233 we prepared for irradiation at Site X for Goldschmidt last January. I asked Katzin to look into the matter. [10, pg.98]

1945-07-09 Report CS-3072, Chemistry Division, "Summary Report for June, 1945" was issued today. The material on Section C-I has been covered in meetings held during the month and includes such work as reducing agents for the Redox process, the Berkeley helium ion bombardment of natural uranium, the alpha decay of Pu241, the chemistry of neptunium, of plutonium, of thorium, and of actinium. The report also includes a report on U233. [10, pg.100-101]

1945-07-10 Hyde reported that he is preparing to handle two targets that should arrive soon—one is the thorium plus deuteron bombardment and the other a target of U233 plus helium ions. In the U233 bombardment, he intends to look for such things as Np233, Np234, Pu234, Pu235, and Pu236.

Studier said that the bulk of the U233 has now been thoroughly purified and set aside for decay. [10, pg.102]

Katzin supplied me with some information to use in my reply to Bill Watson's questions. I say that the weight of isotope 23 in the sample, determined by radio-assay, was about 11.0 mg and we estimate the isotope purity to be 96 percent. The actual weight of the sample was 14.56 mg. I also say we claim no great accuracy. [10, pg.103]

1945-07-12 Katzin, Studier, Manning, Cunningham, and I met to discuss the U233 for the Koch-Dancoff Experiment. Studier will clean up approximately 30 mg just returned from Zinn sometime before August 10. On approximately August 10, he will take 80 mg from the 280 mg (set up for decay) and extract the Th229 and clean it up and give approximately 110 mg to Cunningham or Manning to have Scott or Britain make the plate for Koch. Manning will arrange to have that plate delivered to Koch or Dancoff. [10, pg.106-107]

1945-07-13 Doan called me from Clinton Labs to ask whether he should request our thorium carbonate cans. I told him to request "about 100 cans." He will try to put as many as possible in the pile. [10, pg.107]

I sent an estimate of our requirements for thorium to Daniels for the period from the present until December 1946. I ask for 500 pounds of metal and 100 pounds of oxide. This assumes that we will irradiate the metal at Site W to produce gram amounts of U233. [10, pg.108]

1945-07-16 At 5:30 a.m. mountain time, the first explosion of an atomic bomb occurred at Alamogordo, New Mexico, about 100 miles south of Albuquerque.

Later I read the account of this event that Joe Kennedy wrote in his diary:

> We got to the site around 3:00 a.m., and took our position roughly 20 miles north (?) of the spot. It had been raining, and was completely overcast, in spite of Jack Hubbard's prediction to Parsons (Sunday morning) that it positively would be clear overhead in the North and South directions. We didn't know whether they would fire; Bradbury who was to meet us was there, but didn't know he was to meet us; the radios didn't work except one in an MP car down the road. Finally we learned by MP radio that the shot would be delayed (due to weather) for perhaps one or two hours. Various ones tried to sleep on the ground, etc. CAT, Laurence, and I tried in the sedan, but L. snored awfully.
>
> Don aroused us at about 5:00 a.m., saying the shot would be within five minutes or not that day. Feynman had managed to get one plane, but not the ground station, on our radio. From it we finally guessed that 5:30 a.m. was the zero time. The plane gave signals at 10 minute and at 2 (?), also colored rockets were shot by soldiers at our camp but no one knew the code! A siren could be faintly heard, but again the code was not known.
>
> At zero, I fortunately was facing exactly in the right direction, and with my eyes well-shielded with the dark glass. A brilliant yellowish spurt of fire was first seen, about 1000 feet, in a direction just clockwise of exact vertical, where it mushroomed at the top. At the time of this mushrooming, the total brightness seemed greatest. The ball formed rapidly, grew (in a matter of seconds) to one roughly 1000-2000 feet in diameter, and became increasingly less brilliant. At this time, with the ball looking like flame from burning oil, I removed by dark glass. Before this, while the ball was forming and rising, I looked to the side around the dark glass and the ground and hill appeared as well illuminated as in mid-morning. Likewise, I viewed the sky above. The overcast of stratocumulus (?) clouds were pink on the underside and well illuminated, as at sunrise.
>
> The red and black fireball grew rapidly to several times its size, and rose continuously, reaching about 14,000 feet in about 100 seconds. As it grew, it changed quickly from red and black flames to a mass of white, billowy smoke interspersed and surrounded (to an extent of about 1000 feet beyond the borders of the smoke) by a purple-blue glow, like the low pressure discharge in air in color, and reminding me of the blue glow around hot Po samples but on a much larger scale. As tens of seconds went by, the blue glow diminished and the white smoke cloud became more the prominent feature, until, when the blast wave took me by surprise

at about 100 seconds, the blue glow was practically gone.

It was at this time well before sunrise, yet light enough for smoke to be easily visible. It formed a column, not entirely vertical, much narrower at the bottom than at the top, which was dome shaped (from the ball shape of the fireball). Soon after the blast and the end of the blue glow, the dome penetrated the thin cloud haze (at about 17,000 feet), and continued visible above the haze. It appeared to be drifting rapidly northeast (?) at the top. It was perhaps ten minutes later that we began to gather in the cars and buses, and everyone of the party was away shortly before 6:00 a.m.

Some high spots of the trip: (1) the rough "McKee workmen" appearance of the people in the party; (2) the rest stops along the road; (3) Teller, Bethe, and Thomas at about two minutes, applying sunburn lotion to their faces, with Teller saying "100 to 1 it's not needed, but what do we know?"; (4) the utter silence of the explosion until the sound wave reached us; (5) the quiet echoing of the sound in the mountains, audible for about five minutes; (6) the usual guess afterwards of 3-8 kilotons.

We got back to L.A. in the early afternoon, very much worn out. [10, pg.109-111]

At 7:45 p.m. Katzin spoke on the U233 problem at the Chemistry Seminar in Room 251, Ryerson Laboratory. [10, pg.112]

1945-07-17 The Chicago Sun, Tuesday, July 17, 1945, reports on page 3:

SHELL DUMP EXPLODES
Alamogordo, New Mexico, July 16 - (UP) - A magazine containing high explosives, gas and other shells blew up at the Army base here today with a pyrotechnic display visible 200 miles away. No one was injured. [10, pg.112]

Katzin suggested that I discuss the importance of measuring the cross section for the absorption of neutrons by Pa233 to form Pa234. This loss may block the U233 pile if the reaction has a cross section as high as about 500 barns. We hope to settle this question by putting a sample of 10 mg of thorium in a Hanford pile for 30 days, then work up the sample within five days. [10, pg.114]

I notified Hamilton that we are sending some U233 and ionium which we would like to have bombarded with helium ions—about 50 to 100 microampere-hours on each. I tell him that the U233 sample (GTS-104) contains 15.7 mg of U233 and has an isotopic purity of about 95 percent. The ionium sample consists of about 7.8 mg of Th230; the combined weight of the thorium and ionium dioxides is 35.5 mg. The samples are going by courier via Site Y and should arrive in Berkeley Thursday night. [10, pg.114-115]

I point out, in a memo to Daniels, that one of the secondary reactions in the thorium blanket of a breeder pile is the absorption of a neutron by Pa233 to form Pa234. The cross section for this reaction is unknown and, if it should be as large as 500 barns, a

breeder reactor of this type might not be feasible. I propose to measure this cross section by irradiating 10 g of thorium in one of the Hanford piles in a position of maximum neutron flux for 30 days. The sample should then be shipped to us as soon as possible so that the uranium fraction can be separated from the undecayed Pa233 at the earliest possible moment. This will keep the ratio U234/U233 at a maximum and make it possible to detect the U234 in the presence of U233.

Hilberry prepared a 24-page document of the suggestions to the Interim Committee for post-war organization. This document was the culmination of panel discussions with scientific personnel of the DSM projects. The report proposed the establishment of a Commission to be called the Commission on Atomic Energy, or possibly the National Nucleonics Commission, and would be a full-time working unit resembling an industry's executive committee. The Commission should consist of five or seven men appointed by the President for five or seven-year terms, with the appointments receiving Congressional approval. Of the five or seven men, two or three should be scientists, two or three should be a representative of the military authorities. The report also proposed an advisory panel of twelve members appointed by the President for six-year terms. Five would be scientists, three would be military, and four would be public representatives. The panel would advise the Commission on all matters of major policy, on budgets, on the appointment of a director, and on the appointment of divisional and special advisory panels. The report visualized the work of the Commission as being grouped in four divisions under a Director appointed by the Commission: (1) administration, (2) scientific and technical research and development, (3) social, political, and economic research, (4) industrial operations.

It was proposed that the research program be carried out by personnel at established universities or industrial laboratories through government grants or contracts. Regional cooperative laboratories should be encouraged, by government aid, for major tasks. The report recommended that industrial development be carried out in industrial laboratories and by the creation of two government-operated development laboratories. Military development should utilize the present ordnance laboratories. A laboratory under military and Commission control should be established. [10, pg.115-116]

1945-07-21 I read a letter I received today from Paneth in Montreal about the naming of the 4n + 1 series. He notes that I seem inclined to call the whole series after its longest living member so far known, the Neptunium series. He hopes that the name remains. [10, pg.120]

In other work a description is given of the preparation of thorium oxycarbonate with less than 0.01 ppm of uranium, starting with commercial thorium nitrate. Directions are given, using ceric oxidimetry for the determination of small amounts of uranium present in large amounts of aluminum powder. Details are given of the preparations for a long-irradiation experiment for the measurement of the neutron capture cross section of ionium. On the basis of interrupted studies, information and predictions are given for the properties of thoria slurries. [10, pg.122]

1945-07-25 In Oak Ridge. I again attended the meeting on breeder piles; Weinberg reported

on the Clinton pilot breeder, and Shapiro discussed the catalyst problems. Later reports included English's on the chemistry of reactor solutions, Nordheim's on controls, Coryell's on the physical chemistry of pile solutions, Wollan's on absorption coefficients, Ward's on circulation and cooling, Lyon's on removal of gaseous fission products, and Peterson's on the removal of non-gaseous fission products from U235. [10, pg.124]

1945-07-26 On Tuesday while I was in Oak Ridge, Groups 1 and 3 met at 8:28 am in the New Chemistry conference room. Asprey, Cunningham, Florin, Fried, Ghiorso, Hindman, Hopkins, Hyde, Jaffey, James, Katzin, Magnusson, Manning, O'Connor, Perlman, Peterson, Robinson, Scott, Simpson, Stewart, R. Thompson, S. Thompson, Van Winkle, Weissbourd and Westrum attended. Manning opened the meeting by announcing that three Berkeley deuteron bombardments arrived yesterday: thorium, U235, and U238. He also mentioned that Farmer's No. 3 has arrived, but the sample probably will not be worked on until next Monday. Perlman estimated that Farmer's No. 3 has 7 to 8 percent of Pu240 and should be good material for helium ion and deuteron bombardments. [10, pg.124-125]

Studier talked about the present status of work on the 4n+1 series. He noted that the half-life for Ac225 is expected to be as low as 10 days, perhaps even lower. He also talked about the half-life values for Bi213. The counts originally believed to be from emanation are counts arising from atomic recoil. [10, pg.124]

1945-07-27 The Allies issued an ultimatum to Japan for its unconditional surrender. [10, pg.128]

1945-07-30 Japanese Premier Suzuki scorned as unworthy of official notice the allied Potsdam surrender ultimatum, according to this morning's paper. [10, pg.131]

1945-08-01 Eugene P. Wigner left the Met Lab to join Princeton University. [10, pg.136]

I asked Daniels, by memo, to arrange for the neutron irradiation of 1 mg of Np237 in one of the Hanford piles in a position of maximum of neutron flux for a period of 90 days. Such a neutron irradiation is important because substantial quantities of the isotope are produced in any converter pile operating on U235 and in breeder piles operating on U233. It is, therefore, important to study all the isotopes formed under very strong neutron irradiation of Np237. The irradiation will give us a method to produce pure Pu238 for study of its fission properties. [10, pg.137]

1945-08-04 Perlman sent a memo to Daniels stating the production of about 10 grams of U233 in any of the Hanford units is entirely feasible and should not upset operations as currently carried out there. He says that the preparations required for the bombardment are the processing of over 80 kg of thorium metal free to the extent of 1 ppm of uranium, the determination of an adequate method for canning the thorium, and the carrying out of the operation.

The U.S. and Great Britain have agreed at Potsdam on new blows to speed the surrender of the Japanese in the Pacific, according to reports in this morning's paper. [10, pg.142]

1945-08-05 The top headline in today's Sun reads "B-29s Doom 12 More Cities." Fleets of superfortresses scattered "Evacuate or Die" leaflets over twelve Japanese cities early today.

MacArthur is set to head a huge invasion of Japan from Okinawa, the smaller Ryukyus, and the Philippines. [10, pg.142]

1945-08-06 At 11:00 a.m., Eastern time, ratio stations began broadcasting the following statement by the President of the United States:

> Sixteen hours ago an American airplane dropped one bomb on Hiroshima, an important Japanese army base. That bomb had more power than 20,000 tons of T.N.T. It had more than two thousand times the blast power of the British "Grand Slam" which is the largest bomb ever yet used in the history of warfare.
>
> The Japanese began the war from the air at Pearl Harbor. They have been repaid many fold. And the end is not yet. With this bomb we have now added a new and revolutionary increase in destruction to supplement the growing of our armed forces. In their present form these bombs are now in production and even more powerful forms are in development.
>
> It is an atomic bomb. It is a harnessing of the basic power of the universe. The force from which the sun draws its power has been loosed against those who brought war to the Far East.
>
> Before 1939, it was the accepted belief of scientists that it was theoretically possible to release atomic energy. But no one knew any practical method of doing it. By 1942, however, we knew that the Germans were working feverishly to find a new way to add atomic energy to the other engines of war with which they hoped to enslave the world. But they failed. We may be grateful to Providence that the Germans got the V-1's and the V-2's late and in limited quantities and even more grateful that they did not get the atomic bomb at all.
>
> The battle of the laboratories held fateful risks for us as well as the battles of the air, land, and sea, and we have now won the battle of the laboratories as we have won the other battles.
>
> Beginning in 1940, before Pearl Harbor, scientific knowledge useful in war was pooled between the United States and Great Britain, and many priceless helps to our victories have come from that arrangement. Under that general policy the research on the atomic bomb was begun. With American and British scientists working together we entered the race of discovery against the Germans.
>
> The United States had available the large number of scientists of distinction in the many needed areas of knowledge. It has the tremendous industrial and financial resources necessary for the project and they could be devoted to it without undue impairment of other vital war work. In the United States the laboratory work and the production plants, on which a substantial start had already been made, would be out of reach of enemy bombing, while at that time Britain was exposed to constant air attack and was still threatened with the possibility of invasion. For these reasons Prime Minister Churchill and President Roosevelt agreed that it was wise to carry on the project here. We now have two great plants and many

lesser works devoted to the production of atomic power. Employment during peak construction numbered 125,000 and over 65,000 individuals are even now engaged in operating the plants. Many have worked there for two and a half years. Few know what they have been producing. They see great quantities of material going in and they see nothing coming out of these plants, for the physical size of the explosive charge is exceedingly small. We have spent two billion dollars on the greatest scientific gamble in history—and won.

But the greatest marvel is not the size of the enterprise, its secrecy, nor its cost, but the achievement of scientific brains in putting together infinitely complex pieces of knowledge held by many men in different fields of science into a workable plan. And hardly less marvelous has been the capacity of industry to design, and of labor to operate, the machines and methods to do things never done before so that the brain child of many minds came forth in physical shape and performed as it was supposed to do. Both science and industry worked under the direction of the United States Army, which achieved a unique success in managing so diverse a problem in the advancement of knowledge in an amazingly short time. It is doubtful if such another combination could be got together in the world. What has been done is the greatest achievement of organized science in history. It was done under high pressure and without failure.

We are now prepared to obliterate more rapidly and completely every productive enterprise the Japanese have above ground in any city. We shall destroy their docks, their factories, and their communications. Let there be no mistake; we shall completely destroy Japan's power to make war.

It was to spare the Japanese people from utter destruction that the ultimatum of July 26 was issued at Potsdam. Their leaders promptly rejected that ultimatum. **If they do not now accept our terms they may expect a rain of ruin from the air, the like of which has never been seen on this earth.** Behind this air attack will follow sea and land forces in such numbers and power as they have not yet seen and with the fighting skill of which they are already well aware.

The Secretary of War, who has kept in personal touch with all phases of the project, will immediately make public a statement giving further details.

His statement will give facts concerning the sites at Oak Ridge near Knoxville, Tennessee, and at Richland near Pasco, Washington, and an installation near Santa Fe, New Mexico, Although the workers at the sites have been making materials to be used in producing the greatest destructive force in history they have not themselves been in danger beyond that of many other occupations, for the utmost care has been taken of their safety.

The fact that we can release atomic energy ushers in a new era in man's understanding of nature's forces. Atomic energy may in the future supplement the power that now comes from coal, oil, and falling water, but at present it cannot be produced on a basis to compete with them commercially. Before that comes there

must be a long period of intensive research.

It has never been the habit of the scientists of this country or the policy of this Government to withhold from the world scientific knowledge. Normally, therefore, everything about the work with atomic energy would be made public.

But under present circumstances it is not intended to divulge the technical processes of production or all the military applications, pending further examination of possible methods of protecting us and the rest of the world from the danger of sudden destruction.

I shall recommend that the Congress of the United States consider promptly the establishment of an appropriate commission to control the production and use of atomic power within the United States. I shall give further consideration and make further recommendations to the Congress as to how atomic power can become a powerful and forceful influence towards the maintenance of world peace.

During the afternoon a more detailed statement was released by the Secretary of War describing how the War Department had brought the bomb project to fruition and had made a start on planning for the future.

Enroute to Berkeley on the "City of San Francisco." The dropping of the atomic bomb was announced in newspaper headlines in papers that the passengers picked up at stops enroute. There was great excitement, and I had some difficulty keeping my counsel during the many conversations I heard in the club car and elsewhere, When the train stopped in Ogden, Utah, at 6:30 p.m., mountain time, I too, bought a newspaper announcing the event. [10, pg.143-145]

1945-08-07 At 8:50 a.m. I arrived in Berkeley on the "City of San Francisco."

During the day I had meetings with Lawrence and others. We discussed the great newspaper interest over the Hiroshima atomic bomb. The papers are replete with articles on the atomic bomb, how and where it was developed, Churchill made a statement on the importance of the bomb, indicating that the course of the war would have been altered had the Germans produced this weapon. [10, pg.145-146]

Under Secretary of War, Robert P. Patterson, issued the following statement to the men and women of the Manhattan District Project:

Today the whole world knows the secret which you have helped us keep for many months. I am pleased to be able to add that the warlords of Japan now know its effects better even than we ourselves. The atomic bomb which you have helped to develop with high devotion to patriotic duty is the most devastating military weapon that any country has ever been able to turn against its enemy. No one of you has worked on the entire project or known the whole story. Each of you has done his own job and kept his own secret, and so today I speak for a grateful nation when I say congratulations and thank you all. I hope you will continue to

keep the secrets you have kept so well. The need for security and for continued effort is fully as great now as it ever was. We are proud of every one of you. [10, pg.156]

1945-08-08 Today's headlines are still devoted to the Hiroshima bomb. Four square miles of the city were destroyed. [10, pg.146]

1945-08-09 A plutonium bomb was dropped on Nagasaki shortly before 11:00 a.m. Japanese time, August 9 (yesterday our time). The announcement was the top headline in this morning's paper here. [10, pg.146]

The Soviets have declared war on Japan and have made their first attack on the eastern border of Manchuria. [10, pg.147]

1945-08-10 President Truman warned the Japanese that they will suffer decimation and destruction if they do not surrender. Japanese radio claims that they have similar weapons and will retaliate. Smoke and dust is still covering Nagasaki, according to today's paper. [10, pg.147]

Daniels and Branch issued the following notice to all employees of the Metallurgical Laboratory.

> Despite the fact that the surrender terms of the Japanese may be accepted, it is necessary that all of us keep in mind that the Metallurgical Laboratory still has important and significant work to do.
>
> The Manhattan District has requested that all employees remain at work as usual.
>
> In the interest of future peace it is more important than ever that security regulations be maintained regarding the disclosure of scientific information.
>
> The Metallurgical Laboratory has not yet been authorized to release any information or publicity. Announcement of V-J Day must not be construed as authority to release such information. We will be notified officially when such releases are possible.
>
> In view of our solemn responsibilities and the fine spirit evidenced by the employees of the Metallurgical Laboratory there is no doubt of your full and wholehearted cooperation. [10, pg.157-158]

1945-08-11 Today's paper is full of hope for peace. It also reports that 30 percent of Nagasaki was destroyed. [10, pg.148]

1945-08-12 "Hirohito Can Stay On, But Only as Puppet" says this morning's headline. Peace in the Pacific grows closer as the Japanese were given a final chance of surrendering and keeping the Emperor, with occupation forces under an American supreme commander. [10, pg.148]

1945-08-13 The war is still going on. American and British pilots bombed Tokyo again this morning. [10, pg.148]

The British Information Services issued a 22-page news release entitled "Britain and the Atomic Bomb." The statement includes historical background, descriptions of the organization of the work in the UK and the scope of the research programs undertaken, the decision to transfer to Canada one section of the work which became a joint Anglo-CanadianU.S. enterprise, and the participation of British scientists in the work at Berkeley, Oak Ridge, and Los Alamos. [10, pg.158]

1945-08-14 Throughout the country crowds are in the streets to celebrate the end of the war. Thousands of people jammed into Lafayette Square in Washington, D.C., chanting, "We want Truman," until he and Mrs. Truman appeared on the White House steps and the President spoke to the crowd. [10, pg.148]

1945-08-15 "Great War Ends!" is today's banner headline. The Emperor of Japan said that the "atom bomb" caused them to give up. [10, pg.149]

The University of Chicago received the following telegram from General Groves today:

> Official declaration of cessation of hostilities with Japan does not in any way alter security limitations on release of information on the atomic bomb project. Security restrictions imposed in my telegram of six August continue to apply. The President in his broadcast of nine August emphasized the necessity in the interest of national safety for controlling release of information on this revolutionary development. Loose talk and idle speculation by persons now or formerly connected with the Project jeopardizes the security of the nation and must be controlled. I am again asking you personally to continue your complete cooperation through my office by your entire organization and by each of your subcontractors in the maintenance of national security. [10, pg.161-162]

1945-08-17 The University of Chicago issued a 12-page news release describing the University's role in the work which culminated in the release of atomic power. (Reference numbers in right hand margin of the release refer to chapter and paragraph of the official War Department report, The Smyth Report.) The Met Lab personnel identified by name were Compton, Fermi, Allison, Franck, Warren Johnson, Hogness, Seaborg, and Stone. [10, pg.163]

1945-08-18 The Japanese surrender delegation has been selected and will fly to Manila tomorrow. Also in today's paper appeared pictures of the bomb test at Alamogordo. [10, pg.149]

1945-08-20 Daniels requested for us the following Hanford irradiations from Compton: (1) neutron irradiation of 1 mg of Np237 for 90 days, (2) irradiation of 10 grams of thorium for 30 days in order to obtain material for measuring the neutron-capture cross section of Pa233, (3) production of gram amounts of U233 by irradiation of 80 kg of pure thorium metal to be obtained from Spedding at Ames. The latter irradiation would be for about three months. [10, pg.163-164]

1945-08-21 Manning asked about the plans for the ionium plus neutrons. Jaffey explained the idea was to determine the neutron capture cross sections of ionium and thorium relative to U238, but they are waiting for the Pa233 to decay. They plan to take a pulse analyzer curve to determine the ratios of alpha activities. [10, pg.149]

1945-08-22 The formal surrender of the Japanese will take place on August 31 and will be formally signed on a U.S. battleship in Tokyo. [10, pg.152]

The scientific program on "Power Possibilities" was held during the afternoon. The speakers were Metcalf (Patent Possibilities), Daniels (Preliminary Design for a High-Temperature Oxide Pile), Willard (Progress Report on High-Temperature Piles), Goldberger (Calculations of Critical Size of BeO High-Temperature Piles), Ohlinger (Engineering Features of Nuclear Power), and Gale Young (Turbine Power Plants).

The Policy Meeting of the Project Council was held at 9:20 a.m. in Room 209, Eckhart Hall. It was attended by Bartky, Chapman, Chipman, Compton, Daniels, Dempster, Doan, Hamilton, Harrell, Hilberry, Hogness, Howe, Jacobson, Jeffries, Johnson, Leverett, McKinley, Murphy, Nordheim, Spedding, Stone, Szilard, Thomas, Tracy, Warner, Watson, Whitaker, Wirth, and Zinn. Compton opened the meeting by saying he knew that all those present are glad to see that eight days after the first bomb was dropped, the war was at an end, indicating that, in spite of the great loss of life, it was a saving in the end.

There was a discussion of the statement from the President to General Groves to hold everything in publishing information. As a result those on the Project have been asked not to give any information at all. Warren Johnson asked if it is necessary to get permission from Washington to disclose anything that is in the Smyth Report and was told by Compton that technically it is not necessary, but we are likely to get into trouble if we do not. Compton then said that Truman has ordered no more information be disclosed. Zinn remarked, "It's more important than who gets the credit and who does not. If people are going to get the correct information they have to get it from the right people. Washington is really cutting off the right source of information which would educate the public."

Thomas, Watson, and Jeffries argued for patience, Thomas suggesting that if the situation is not clarified in two weeks they should take their troubles to the Panel through Compton. Jeffries felt the release of the Smyth Report was more than many of us had any right to expect from the standpoint of the amount of information that was given out. He went on to observe that the Smyth Report deals very little with future development. This led Compton to state there is a strong feeling in the group that the American public should have an opportunity to know as soon as possible from those who are informed on the scientific aspects about their own feelings concerning the future implications of this Project and to make this group free in expressing themselves as American citizens on the implications of the work in which we have been involved.

During the discussion of the release of scientific information Hogness said he felt impelled to speak for the chemists—there was not proper credit given to them in the Smyth Report.

Szilard said the group is not really clamoring for the release of scientific information and added that he thought they all feel the Smyth Report went far too far. [10, pg.166-167]

1945-08-25 Farrington Daniels announced that 150 copies of the Smyth Report have been shipped to the Metallurgical Project. Daniels states, "The statements in the Smyth Report continue to be the limit of disclosure which we are permitted to make." [10, pg.169]

1945-08-28 A meeting was held during the evening to discuss Rabinowitch's report on political implications and postwar plans. Rabinowitch listed eight possible comments on the atomic bomb crisis for discussion: (1) Solution is needed, and it ought to be the correct one. (2) Do not reveal the secret. (3) Let's corner the raw materials. (4) A defense is sure to be found. (6) Let's scatter our cities or dig underground. (7) If you smash our cities, we smash yours. (8) Let's prohibit atomic warfare. [10, pg.170]

1945-09-05 Perlman sent Captain Chapman information on the preparation for a special irradiation of thorium at Hanford. Spedding will prepare the thorium metal in the form of a billet; and Foote will supervise the extrusion into bar stock which can then be machined and canned. The canning will be done at Hanford. [10, pg.178]

1945-09-11 I also transmitted a proposed program of chemical research in connection with the production of U233, under the following headings, which will utilize the services of 115 men: (1) Small-scale Production in Present Type Natural Uranium Piles, highest priority, 15 men; (2) Large-scale Production in Converter Piles Using Enriched U235 or Plutonium as Fuels, highest priority, 100 men; (3) U233 Breeder Piles, medium priority. This problem will follow No. 2 above and the men can be shifted as necessary. [10, pg.184]

Hamilton and I then talked about the fact that the yield of U232 from thorium plus deuterons bombardment is about twice as great at 22 MeV as at 20 MeV. [10, pg.185]

1945-09-12 I met with A. T. Cape of Coast Reductions, Inc., 2 W. 45th Street, New York, New York, in New Chem. Cape told me that one of the men in his company found the thorium-uranium mineral in which we are interested in 1937 in the black sand of the beach at Monterey, California. The mineral consists of 60 percent ThO_2, 20 percent U3O8, and 20 percent SiO_2; the deposit extends from Monterey north to San Francisco. Cape gave me a sample that has a density of 7.4. He said that his company owns 10,000 feet of beach right at Monterey Bay from which they have taken out 50,000 tons of this black sand for roofing paper. Cape said that he works at Ohio State University. His home address is 40 W. Dominion Blvd., Columbus, Ohio; Jefferson 2166. [10, pg.186]

1945-09-15 I phoned E.O. Lawrence at Berkeley and told about the thorium-uranium ore deposit at Monterey Bay. We talked about my impending move to the Radiation Laboratory at Berkeley. He mentioned he will stop in Chicago on a United Airlines flight due to arrive at 11:13 am Tuesday morning, and I said that I would meet him at the airport. We agreed this would give us time to talk about the move of my colleagues and me to Berkeley. He will then take a train to Washington or New York about 3:30 pm. [10, pg.190]

1945-09-25 I telephoned Captain Chapman in the Area Engineer's Office to give him all the essential information on the source of thorium at Monterey Bay as described to me by Cape. [10, pg.206]

1945-10-01 A. T. Cape called me from Columbus, Ohio. He told me that they have analyzed the thorium ore from the Monterey Bay area with the following results: ThO_2 - 70.40 percent, U3O8 - 11.18 percent, SiO_2 - 18.22 percent. Cape wants to know whether we have any interest in this ore. People are considering purchasing the surrounding land, and this may make it difficult to buy the critical area later on. [10, pg.211]

1945-10-02 I then asked Hamilton to tell Lawrence that Cape has called to inquire whether we might have any possible interest in the thorium ore from Monterey Bay. [10, pg.216]

1945-10-03 President Truman sent the following message to the Senate today:

> Almost two months have passed since the atomic bomb was used against Japan. That bomb did not win the war, but it certainly shortened the war. We know that it saved lives of untold thousands of American and Allied soldiers who would otherwise have been killed in battle.
>
> The discovery of the means of releasing the atomic energy began a new era in the history of civilization. The scientific and industrial knowledge on which this discovery rests does not relate merely to another weapon. It may some day prove to be more revolutionary in the development of human society than the invention of the wheel, use of metals, or the steam or internal combustion engine.
>
> Never in history has society been confronted with a power so full of potential danger and at the same time so full of promise for the peace of the world. I think I express the faith of the American people when I say that we can use the knowledge we have won, not for the devastation of war, but for the future welfare of humanity.
>
> To accomplish that objective we must proceed along two fronts—the domestic and the international.
>
> The first and most urgent step is the determination of our domestic policy for the control, use, and development of atomic energy within the United States.
>
> We cannot postpone decisions in this field. The enormous investment which we made to produce the bomb has given us the two vast industrial plants in Washington and Tennessee, and the many associated works throughout the country. It has brought together a vast organization of scientists, executives, industrial engineers, and skilled workers—a national asset of inestimable value.
>
> The powers which the Congress wisely gave to the Government to wage war were adequate to permit the creation and development of this enterprise as a war project. Now that our enemies have surrendered, we should take immediate action to provide for the future use of this huge investment in brains and plant. I

am informed that many of the people on whom depend the continued successful operation of the plants and the further development of atomic knowledge, are getting ready to return to their normal pursuits. In many cases these people are considering leaving the project largely because of uncertainty concerning future national policy in this field. Prompt action to establish national policy will go a long way toward keeping a strong organization intact.

It is equally necessary to direct future research and to establish control of the basic raw materials essential to the development of this power whether it is to be used for purposes of peace or war. Atomic force in ignorant or evil hands could inflict untold disaster upon the Nation and the world. Society cannot hope even to protect itself—much less to realize the benefits of the discovery—unless prompt action is taken to guard against the hazards of misuse.

I therefore urge, as a first measure in a program of utilizing our knowledge for the benefit of society, that the Congress enact legislation to fix a policy with respect to our existing plants, and to control all sources of atomic energy and all activities connected with its development and use in the United States.

The legislation should give jurisdiction for these purposes to an Atomic Energy Commission with members appointed by the President with the advise and consent of the Senate.

The Congress should lay down the basic principles for all the activities of the Commission, the objectives of which should be the promotion of the national welfare, securing the national defense, safeguarding world peace, and the acquisition of further knowledge concerning atomic energy.

The people of the United States know that the overwhelming power we have developed in this war is due in large measure to American science and American industry consisting of management and labor. We believe that our science and industry owe their strength to the spirit of free inquiry and the spirit of free enterprise that characterize our country. The Commission, therefore, in carrying out its functions should interfere as little as possible with private research and private enterprise, and should use, as much as possible, existing institutions and agencies. The observance of this policy is our best guaranty of maintaining the preeminence in science and industry upon which our national well-being depends.

All land and mineral deposits owned by the United States which constitute sources of atomic energy, and all stockpiles of materials from which such energy may be derived, and all plants or other property of the United States connected with its development and use should be transferred to the supervision and control of the Commission.

The Commission should be authorized to acquire at a fair price, by purchase or by condemnation, any minerals or other materials, which are not already owned by the United States.

The power to purchase should include real and personal property outside the limits

of the United States.

The Commission should also be authorized to conduct all necessary research, experimentation, and operations for the further development and use of atomic energy for military, industrial, scientific, or medical purposes. In these activities it should, of course, use existing private and public institutions and agencies to the fullest practicable extent.

Under appropriate safeguards, the Commission should also be permitted to license any property available to the Commission for research, development, and exploitation in the field of atomic energy. Among other things such licensing should be conditioned, of course, upon a policy of widespread distribution of peacetime products on equitable terms which will prevent monopoly.

In order to establish effective control and security, it should be declared unlawful to produce or use the substances comprising the sources of atomic energy or to import or export them except under conditions prescribed by the Commission.

Finally, the Commission should be authorized to establish security regulations governing the handling of all information, material, and equipment under its jurisdiction. Suitable penalties should be prescribed for violating the security regulations of the Commission or any of the other terms of the act.

The measures which I have suggested may seem drastic and far-reaching. But the discovery with which we are dealing involves forces of nature too dangerous to fit into any of our usual concepts.

The other phase of the problem is the question of the international control and development of this newly-discovered energy.

In international relations as in domestic affairs, the release of atomic energy constitutes a new force too revolutionary to consider in the framework of old ideas. We can no longer rely on the slow progress of time to develop a program of control among nations. Civilization demands that we shall reach at the earliest possible date a satisfactory arrangement for the control of this discovery in order that it may become a powerful and forceful influence toward the maintenance of world peace instead of an instrument of destruction.

Scientific opinion appears to be practically unanimous that the essential theoretical knowledge upon which the discovery is based is already widely known. There is also substantial agreement that foreign research can come abreast of our present theoretical knowledge in time.

The hope of civilization lies in international arrangements looking, if possible, to the renunciation of the use and development of the atomic bomb, and directing and encouraging the use of atomic energy and all future scientific information toward peaceful and humanitarian ends. The difficulties in working out such arrangements are great. The alternative to overcoming these difficulties, however, may be a desperate armament race which might well end in disaster. Discussion of the international problem cannot be safely delayed until the United Nations

organization is functioning and in a position adequately to deal with it.

I therefore propose to initiate discussion, first with our associates in this discovery, Great Britain and Canada, and then with other nations, in an effort to effect agreement on the conditions under which cooperation might replace rivalry in the field of atomic power.

I desire to emphasize that these discussions will not be concerned with disclosures relating to the manufacturing process leading to the production of the atomic bomb itself. They will constitute an effort to work arrangements covering the terms under which international collaboration and exchange of scientific information might safely proceed.

The outcome of the discussions will be reported to the Congress as soon as possible and any resulting agreements requiring congressional action will be submitted to the Congress.

But regardless of the course of discussion in the international field, I believe it is essential that legislation along the lines I have indicated be adopted as promptly as possible to insure the necessary research in, and development and control of, the production and use of atomic energy. [10, pg.216-220]

1945-10-12 I answered the letter I received from Latimer yesterday, enclosing a copy of a statement on the history of U233 which I prepared earlier for another purpose. I tell him the basic patents on the use and on the preparation of U233 have been prepared and applied for in the names of Seaborg, Gofman, and Stoughton. [10, pg.227]

1945-10-16 I suggested that Katzin, Studier, Hyde, and Hagemann begin thinking about the apparatus and other preparations needed for handling the thorium that will be shipped from Hanford between November 6 and November 13. I then asked about plans for fluoride work. Florin said he would like to try to prepare protactinium fluoride after first making trial runs with tantalum. The program will be to study PuF_6, fluorides of protactinium, and fluorides of neptunium.

Peterson announced that he has sent the sample of radium to Clinton for neutron irradiation. I pointed out that this is a feasible way of making pure actinium. Pile bombardment of ionium-thorium mixtures should be an easy method of preparing pure protactinium.

The first session of the two-day meeting on new piles began at 9:00 am in Room 209, Eckhart Hall. I arrived late after the adjournment of our meeting. Others present were Aebersold, Allen, Brown, Burton, Captain Chapman, Cole, Connick, Dancoff, Daniels, Dempster, English, Foote, Franck, F. Friedman, Goldberger, Hogness, Huffman, Hughes, Hutchison, Jesse, W. Johnson, Lapp, Leverett, Lauletta, Manning, Ohlinger, Perlman, Rabinowitch, Rubinson, Soodak, Spedding, Urey, Way, Willard, Wollan, G. Young, Zener, Zinn, and Zirkle. Soodak of Clinton began the meeting with the results of further pile calculations on their pile which is to consist of U235 salt dissolved in heavy water, surrounded by a heavy water reflector, by a row of thorium rods, and finally by a cylindrical graphite reflector. Critical experiments are now being planned.

English spoke on the composition of homogeneous pile solutions at 90 degC. $NaHCO_3$ solutions are favored at present.

Burton discussed bubbling problems in homogeneous piles—they want no bubbles. The answer seems to be in the use of catalysts and operation at a higher temperature—80 or 90 deg C.

Leverett reported on the present state of homogeneous pile technology, saying that the catalyst problem is holding up planning. They have found that sodium silicate suppresses corrosion.

M.D. Peterson then talked about the semiworks experiments on the treatment of pile solutions. They are considering hexone for recovery of the uranium. Stripping with water or evaporation (if a volatile solvent is used) are other possibilities for removal of the uranium from the organic extract. Little work has been done on recovery of U233 from thorium. [10, pg.232-234]

1945-10-17 The thorium plus helium ions target (Berkeley bombardment 10a) arrived.

The morning session of the New Piles Meeting devoted to the Argonne Program was held at 9:00 am in Room 209, Eckhart Hall. Dancoff spoke on the use of sodium as coolant in a fast pile. The problem of power dissipation appears tremendous but not impossible, according to Dancoff. The pile will be equivalent to five Hanford piles in the rate of fission and to eight Hanford piles in the rate of production of new plutonium.

Hughes talked about cross sections for fast neutrons; this is a new method based on the utilization of fission neutrons.

M. L. Goldberger gave some calculations of resonance piles; these are at a less precise stage than those of Soodak for fast piles. For Pu239, the value of eta, which is 2.05 for thermal neutrons, rises to as much as 2.95 for neutrons of several keV energy. [10, pg.234-235]

1945-10-22 I received a letter from Ray Stoughton in which he describes a number of samples of ours now in the Clinton pile. Stoughton says the people in charge of the pile want to know what we want done with them. [10, pg.239]

1945-10-23 I mentioned the letter I received from Ray Stoughton yesterday; we decided to request the plutonium fluoride, the U oxide, and the thorium carbonate. [10, pg.240]

I sent a proposal to Daniels that the 30-year alpha-emitting U232 would be admirably suited for use in connection with the trigger mechanism of the bomb, in place of the short-lived (140 day) polonium. I discuss the feasibility of its production by neutron bombardment of Pa231 that could be obtained as a by-product by extraction and recovery from the uranium during the processing of uranium ores. I say that Pa also could be produced by neutron bombardment of Th230, also found in uranium ores, but in an abundance 60 times that of Pa231.

Finally I point out that the production of large amounts of Np237 would permit the production of another useful alpha emitter, Pu238, through neutron bombardment of the Np237,

and that, in the future high energy piles, the transplutonium isotopes, especially isotopes of element 96, will be produced in such amounts that they might be used for this and other purposes. [10, pg.242]

1945-10-29 Last Wednesday (October 24) Manning answered the letter I received last Monday from Stoughton. Manning suggested, at Stoughton's convenience, that the following items be removed from the Clinton pile and sent to us: (1) 46 mg product fluoride, (2) 5 mg 23 oxide plus 5 g natural uranium as oxide, and (3) 50 g $ThOCO_3$.

Manning wrote to Hamilton on Saturday to tell him that we are sending a sample of U233 for a short bombardment with deuterons (Berkeley bombardment 8b) before Hamilton changes over to helium ions. A courier is scheduled to leave here by plane Tuesday and should deliver the sample by Wednesday, October 31. Manning indicates the material for bombardment 12a (protactinium plus helium ions) will probably be delivered at the same time.

Also on Saturday Farrington Daniels issued a memorandum to academic personnel. He quoted the following statement from Robert P. Patterson, Secretary of War.

> Public discussion of great issues such as the dissemination and regulation of knowledge of atomic science is one of the basic principles on which democratic government is founded. In it all citizens have a right to participate. American scientists in particular, because of their knowledge of the technical matters involved and because of their comprehension of the full social significance of the achievement, can contribute powerfully to it. Security, of course, still requires that nothing beyond the specific subject matter contained in the Smyth Report be brought into discussion, and the use of due care that matters outside the content of this report and still under security regulations be not inadvertently encroached upon. With this sole restriction, however, which applies to all citizens, our scientists should feel that it is proper for them as citizens to join actively in public consideration of this question.

Daniels infers from this recent statement that it is not necessary to submit for review those manuscripts which are concerned exclusively with political and social discussions.

Daniels also says that the organization known as the "Atomic Scientists of Chicago" has done important service in educating the public to the implications of the atomic bomb and the need for international cooperation and control. He would like to make an informal suggestion to the effect that use of the name of this association in press releases should be used sparingly, particularly when non-technical information is involved, in order that the influence of the organization may remain effective.

He then states the Metallurgical Laboratory has important scientific work to do for the welfare of the nation and meetings, which are not directly connected with the Metallurgical Laboratory should, in general, be carried out in the evenings and on Sundays. [10, pg.246-247]

1945-11-05 Daniels sent Colonel Nichols a request for the loan of 25-40 kilograms of enriched U235 and the authorization to build a Chemistry Division proposed experimental high-temperature pile at Argonne. He refers to the initial research on two other pile types (the homogeneous, heavy-water pile proposed by Clinton, and the all-metal, fast-fission pile proposed by Argonne) and suggests that work be pushed vigorously on these two types in addition to our proposed high-temperature pile. Our pile will use beryllium oxide and enriched uranium oxide (containing 20 percent of U235). Daniels makes the point that this design appears promising for a quick demonstration of the practical utilization of power with the minimum investment, while at the same time giving experience in the conversion of thorium into U233. Daniels expresses the need for new reactors as follows:

> "We are now in competition with other nations in the field of nucleonics, and we must develop actively all possible piles, particularly enriched piles. Tests of breeder piles are necessary from a military standpoint, and there is a public demand for power piles and peacetime applications of nuclear energy. To obtain full public support we believe that a power pile should be demonstrated at the earliest possible moment." [10, pg.262]

1945-11-10 A reply arrived from Stoughton about our request (Manning to Stoughton, October 24) for removal of our samples from the Clinton pile. Stoughton says the samples will be sent in about a week. He includes irradiation data on each sample. [10, pg.270]

1945-11-15 I attended a meeting on Pile Evaluation held at the Metallurgical Laboratory today. **Nordheim spoke about the importance of breeder reactors in meeting the future pile requirements of the U.S. Fermi also underlined the future significance of breeder reactors. He said that he thinks many 1,000 kW reactors would be better to have than a few very large reactors. He thinks that thermal reactors will be better for the production of electric power while fast breeders may be best for the production of plutonium.**

Zinn gave a description of power reactors and breeder reactors. He included among the latter, fast breeder reactors with metal coolants such as bismuth, mercury, and lead. Zinn pointed out that these have the highest fast neutron absorption cross section and indicated that perhaps sodium or sodium potassium alloy might be the best possibilities for use as a coolant. He sketched a 2,000 kW unit that might start with U235 but eventually switch over to Pu239.

Willard described a high temperature pile in which beryllium oxide would be mixed with fissionable material in a steam-cooled system surrounded by thorium rods.

Weinberg talked about the heavy-water cooled reactor working on the thorium-U233 cycle.

Fermi indicated that he thinks that the decisions as to the best course of action should be delayed. **He said that the possibility of generating electricity by nuclear power looks good and that breeding is clearly a possibility.** He believes that the production of radioactive substances will be one of the most important things; this will be so even if nuclear power becomes an important source of energy. He pointed out that biological tracers such as C14

will be very important and should have free distribution. He considers this as a duty of this project to science and this nation. He suggested that an announcement should be made very soon that such isotopes will be available. [10, pg.281-282]

1945-11-20 I attended an 8:28 am meeting of Groups 1 and 3 of Section C-I in the New Chemistry conference room. Also present were Asprey, Britain, Cunningham, Florin, Ghiorso, Hagemann, Hindman, Hopkins, Hyde, Jaffey, James, Katzin, Kohman, Magnusson, Manning, O'Connor, Osborne, Peterson, Robinson, Sedlet, Simpson, Stewart, R. Thompson, S. Thompson, Van Winkle, and Westrum. I reported that ten grams of thorium were bombarded with neutrons in the Hanford pile for 16 days and then flown to Chicago. The purpose was to extract the uranium as soon as possible after the pile exposure and determine the ratio of U233 to U234 from which the capture cross section of Pa233 can be calculated. Hagemann said the thorium metal was dissolved in HNO_3 and the solution salted with $Ca(NO_3)2$ from which the uranium was extracted with ether. The yield was 310 micrograms if it is assumed to be pure U233. Samples of this material will be made to run pulse analysis, fission, and mass spectrometry measurements. [10, pg.286-287]

1945-12-15 Helen and I had lunch at the Wrigley Building with John Dunn, the Chicago announcer of the Watson Davis "Adventures in Science" radio program. At 1:15 pm, following a short rehearsal, I was introduced to the CBS radio audience as the guest scientist on the program. Following comments by Watson Davis by wire from Washington, I was interviewed on the direct broadcast from Chicago.

Announcer: You certainly have been discovering a lot of the fundamental stuff of the universe, Dr. Seaborg, and the writers of chemistry texts are going to have quite a time keeping up with you.

Seaborg: Here is something else. There has always been a question as to what kind of family these heavy elements were going to form. We think we have pretty good evidence that the elements with atomic numbers larger than 88 actually form a new series like the rare earths that the chemists know about. And thus the elements actinium, thorium, and protactinium of atomic numbers 89, 90, and 91 are the first elements in this new rare-earth-like series which corresponds very much to the series of elements that begins with lanthanum, which is farther down in the scale of atomic numbers. [10, pg.331-334]

1945-12-18 Perlman and I visited the Ottawa Laboratory of the National Research Council during the morning. We had meetings and discussions with Goldschmidt, Mungen, Cook, Steacie, Gueron, and Mills. George Weil, the U.S. representative to the Canadian Nuclear Energy Program, also attended. We primarily discussed the proposed process change for the 23 separation process involving the removal of the MnO_2 precipitation step and a change in the extraction solvent.

Mungen described his work on the separation of U233 and Pa233 from thorium and later showed us the separation process plant equipment set up in Ottawa. [10, pg.337-338]

1945-12-19 Stewart received a call from Rodin about Stewart's request on December 13, that five cans of thorium carbonate (1, 2, 3, 4, and 5) be removed from the pile. Stewart had said that we had assumed that the cans had already been removed, but he recently learned the cans were still in the pile. Rodin reassured Stewart that the cans had been removed in October and will be shipped to us as soon as possible.

A thank-you letter arrived for me from Watson Davis for my radio performance on his program. He would like a "pre-release" statement about the naming of elements 95 and 96. [10, pg.341]

1945-12-29 I sent Daniels an additional memo (MUC-GTS-2115) about the use of U232 and other material in weapons components (see memo MUC-GTS-2038, October 23, 1945). The District Engineer's Office pointed out that the neutron background arising from the Be(gamma,n) reaction is 50 times higher than the present initiator and might cause predetonation. In addition, the gas phase of thoron in the decay chain might cause problems. I suggest to Daniels that another material other than beryllium, such as boron, lithium, C-13, or fluorine, might be considered for use with U232. I feel that the gas phase problem could be taken care of through proper gadget design.

I stress that the point that the potentialities for the use of Pa231 as a source of U232 appear sufficiently great so that steps should be taken to arrange for its recovery in the uranium processing procedures. It is equally important to recover Th230. [10, pg.351]

I answered a letter dated December 18 from Walter Yust of Encyclopedia Britannica. He asks that I prepare articles on uranium, thorium, artificial radioactivity, protactinium, plutonium, neptunium, and actinium. I agreed to do this and say that the fall of 1946 would be a practical deadline. [10, pg.352]

1946-01-03 I also called Farrington Daniels to discuss the results of Daniels' telephone conversation with Hood Worthington about the provision of materials and their irradiations in the Hanford reactors. Daniels said that they talked about the problems associated with loading Hanford reactors with thorium to product U233. Worthington told him that fuel slugs will be available later which should have 800 g of plutonium per ton. We must supply shipping containers for these slugs. Daniels said that Worthington also discussed the progress on samples being irradiated for us. These include sample CW-4 and a number of milligrams of Np237 and of U233. [10, pg.358]

1946-01-12 I wired P. Gerald Kruger suggesting that he consider Stoughton, Friedlander, and Duffield as candidates for the radiochemistry position at the University of Illinois. [10, pg.375]

1946-01-15 Groups 1 and 3 met at 8:28 am in New Chem conference room. Ray Stoughton from Clinton Laboratories and Eugene Huffman from Berkeley, who are visiting the Met Lab, attended the meeting in addition to the following individuals from Section C-I: Ames, Anderson, Britain, Florin, Ghiorso, Hagemann, Hindman, Hyde, Jaffey, James, Katzin, Magnusson, Manning, Osborne, Peterson, Scott, Sedlet, Simpson, Stewart, Studier, Templeton,

R. Thompson, S. Thompson, Van Winkle, and Westrum. Stewart reported on the shipment of materials. He said that Osborne should receive the neptunium from the Hanford pile irradiation around January 20. The Pa231 sample from Clinton should also arrive about the same time; and R. Thompson, Van Winkle, and Osborne will handle the sample. The purpose of the work is to obtain U232 for fission measurements. The U233 and U238 targets from Berkeley should be here this week. [10, pg.380]

Hagemann announced that the isolation of U233 from thorium slugs is almost complete. Although the purity of the product is still not known, he estimates that 8.5 mg of U233 should be obtained per slug.

Hyde said he has started experiments to determine the neutron absorption cross section of ionium. A mixture of Th230 and Th232 was irradiated in the Clinton pile. After a period of time to permit the Pa233 to decay to non-detectable levels, the ratio of Pa231 to U233 will be measured. Uranium-232 is also present from the second order reaction on Pa231 so that a value for the capture cross section for Pa231 will also be obtainable. Jaffey pointed out that the half-life for U232 is still uncertain and will lead to uncertainties in the cross section values. [10, pg.381]

1946-01-29 Hagemann talked about the recent neutron-induced fission measurements carried out on pure U233 and a sample of U233 which contains some U234. He stated that previous measurements on these two samples resulted in a lower fission rate for the pure U233 sample than was observed for the U233-U234 mixture. The most recent measurements made last week, however, show that the fission rate for the pure U233 sample is now somewhat larger than the fission rate for the U233-U234 mixuture. Katzin conjectured that this latest result would probably correspond to 0.5 percent to 1 percent of U234 in U233 and would give a cross section for neutron capture by Pa233 of 100 barns $\pm$ 50 percent. The actual calculations have not been made. [10, pg.412]

1946-02-05 James said that he has nothing new on the half-life of U232 from the experiment in which it was milked from Pu236. He has prepared a sample to look for U232 in a uranium fraction separated without U233 tracer. The ratio of U232 to U233 in the previous sample indicated a half-life for U232 of 65 years. However, a small amount of U232 coming through the chemistry from the original cyclotron-bombarded U235 could cause a big effect. [10, pg.417]

1946-02-22 Yesterday while I was in Peoria, Stewart sent Chapman a memo saying that he is sending Chapman, Hagemann's design for lead shields to be used for the shipment of 60-80 kg samples of thorium irradiated in the Hanford pile. Hood Worthington asked for the design of these shields. [10, pg.464]

1946-03-07 In Chicago the extensive report, "Feasibility and Methods of Inspection of World for Plutonium and Other Power Reactors, and for Chemical Processing Plants Associated with Reactors," which L. I. Katzin has been editing is being submitted today.

This report was prepared for the Committee on Technical Phases of Inspection and Control of Atomic Energy by L. B. Borst, H. S. Brown, C. V. Cannon, W. E. Cohn, C. D. Coryell, S. G. English, J. R. Gilbreath, H. Hyman, A. H. Jaffey, L. I. Katzin, D. E. Koshland, Jr., R. N. Lyon, D. R. Miller, K. Z. Morgan, M. D. Peterson, D. Saxon, R. Scalettar, G. T. Seaborg, H. Soodak, R. W. Stoughton, A. Wattenberg, and A. Weinberg. The appendices were prepared by W. Blaedel, J. R. Gilbreath, H. Hyman, A. H. Jaffey, and L. I. Katzin. [10, pg.491]

In the section on "Problems of fissionable material control at the pile" (Section IIB), the report states: We inquire into the possibilities of denaturing fissionable species. There are two ways: One is based on the fact that a uranium bomb is impossible to construct unless the light isotope is present to at least 15 percent. The other is based on the fact that a plutonium bomb is very difficult to make if the Pu content exceeds, say 5 percent. We can therefore tentatively establish the following code:

1. **It is unlawful to possess uranium in which the U235 or U233 content exceeds, say 25 percent.** (The figure 25 percent is deemed safe since 15 percent would give an infinitely large bomb—25 percent would still require a bomb of impractical size.)
2. It is unlawful to possess plutonium in which the Pu240 content is less than 5 percent or in which the spontaneous neutron background is less than that corresponding to this amount of Pu .

How could this code be implemented, and what bearing would its enactment have on the future development of nuclear power? [10, pg.496]

1946-03-25 A letter arrived from Iz this morning asking about Berkeley needs for 23 for six months. He suggests 100 mg. He then goes on to mention the present Hanford irradiation of thorium, saying that if it is bombarded for 100 days, it will produce U233 at somewhere between 400-900 gt level. He would like to consider whether this high a level is desirable for the various uses to which the U233 is to be put; particularly, he questions the sizable amount of U232 alpha activity that would be present. Iz then asks about the amount of neutron-bombarded thorium we should request. [10, pg.526]

1946-03-29 In the Chicago Times today there was a report by Howard W. Blakeslee on the AAAS meeting in St. Louis. He says in part,

> The big fact on which scientists rely to prevent surprise atomic attacks—namely, that all atomic energy is limited strictly to two natural metals, without even a slight prospect of extension—came out here today at the meeting of the American Association for the Advancement of Science.
>
> The metals are uranium and thorium, both scarce and controllable at mines and in power houses, so that scientists believe no nation will be able to prepare for an atomic attack without automatically giving the world advance warning of 18 months to several years.

These facts came from a plan for international control of atomic energy issued simultaneously last night in Washington and St. Louis. Here the plan was explained by Dr. Charles A. Thomas, Vice President of the Monsanto Chemical Company, himself a member of the plan committee and an atomic bomb expert.

Uranium makes both bombs and power. It also makes a new metal, plutonium, for bombs and power. Uranium converts thorium into a new form of explosive uranium. Uranium alone is the base and no other chemical in creation now appears to yield atomic energy.

Important also is denaturation that renders the metals non-explosive, but leaves them useful for power. Denaturation is what happens when an egg is hard boiled. The metals are not boiled; they are denatured by adding a secret chemical element. But, Dr. Thomas said, there is no doubt that "boiling" them will take a long time and large plants.

The proposal is to give an international atomic development authority three controls over: (1) all mining of uranium and thorium, (2) all plants making the metals, (3) inspection of large peacetime power plants (because these could be secretly modified to make bomb materials)... [10, pg.541-542]

1946-04-09 The State Department released the following report of our meeting at the War Department on the problem of denaturing:

The possibility of denaturing atomic explosives has been brought to public attention in a recent report released by the State Department on the international control of atomic energy. Because, for security reasons, the technical facts could not be made public, there has been some public misunderstanding of what denaturing is, and of the degree of safety that it could afford. We have thought it desirable to add a few comments on these points.

The report released by the State Department proposes that all dangerous activities in the field of atomic energy be carried out by an international authority and that operations which by nature of the plant, the material, the ease of inspection and control, are safe, be licensed for private or national exploitation. The report points out that the possibility of denaturing explosive materials so that they "do not readily lend themselves to the making of atomic explosives" may contribute to the range of licensable activities, and to the overall flexibility of the proposed control. The report does not contend nor is it in fact true that a system of control based solely on denaturing could provide adequate safety.

As the report states, all atomic explosives are based on the raw materials, uranium and thorium. In every case the usefulness of the material as an atomic explosive depends to some extent on different properties than those which determine its usefulness for peacetime application. The existence of these differences makes denaturing possible. In every case denaturing is accomplished by adding to the explosive an isotope, which has the same chemical properties. These isotopes

cannot be separated by ordinary chemical means. The separation requires plants of the same general type as the plants at Oak Ridge, Tennessee, though not of the same magnitude. The construction of such plants and the use of such plants to process enough material for a significant number of atomic bombs would probably require not less than one nor more than three years. Even if such plants are in existence and ready to operate some months must elapse before bomb production is significant, but unless there is reasonable assurance that such plants do not exist it would be unwise to rely on denaturing to insure an interval as much as a year.

For the various atomic explosives the denaturing has a different effect on the explosive properties of the material. In some cases, denaturing will not completely preclude making atomic weapons but will reduce their effectiveness by a large factor. The effect of the denaturant is also different in the peaceful application of the material. Further technical information will be required, as will also a much more complete experience of the peacetime uses of atomic energy and its economics, before precise estimates of the value of denaturing can be formulated. But it seems to us most probably that within the framework of the proposal advanced in the State Department report denaturing will play a helpful part.

In conclusion we desire to emphasize two points, both of which have been challenged in public discussion. (1) Without uranium as a raw material there is no foreseeable method of releasing atomic energy. With uranium, thorium can also be used. (2) Denaturing, though valuable in adding to the flexibility of a system of controls, cannot of itself eliminate the dangers of atomic warfare.

L. W. Alvarez
A. H. Compton
G. T. Seaborg
R. F. Bacher
Farrington Daniels
F. H. Spedding
M. Benedict
J. R. Oppenheimer
C. A. Thomas
H. A. Bethe
J. R. Ruhoff
W. H. Zinn

[10, pg.547-548]

1946-05-03 Today the Chicago Sun carried its big supplement, "The Atomic Future," by Howard W. Blakeslee. It is 25 pages long and shows where uranium and thorium are mined, gives a glossary of the languages, and discusses the future peaceful uses of atomic energy. Blakeslee says, "Harnessing the atom ranks with the greatest discoveries of man." He writes of the risks

to their careers taken by scientists who worked on the atomic bomb and of the risks President Roosevelt took in this matter. [10, pg.593]

1946-05-15 Manning said that there are also some 80 pounds of thorium being irradiated in the Hanford pile to produce large quantities of U233 and we may want to work up some of these slugs here. He then said that any further expression of opinions should be turned in within the next few days to permit him tentatively to schedule manpower distribution. He noted that now is the time to make suggestions for special provisions to be made in the new chemical building at Argonne since plans for its construction are still in the formative stage. [10, pg.607]

F MUC-LAO-17: Notes on Meeting of April 26, 1944

9:00 - 10:30, 209-Eckhart

Present: Fermi, Allison, Szilard, Wigner, Weinberg, Seitz, Morrison, Cooper, Vernon, Tolman, Watson, Ohlinger

The first speaker in today's meeting was Mr. Fermi. His remarks follow.

It was assumed for today's discussion that the aim of the chain reaction would be the production of power.

The first type of pile assumed for this purpose was a permanent large pile of about the Hanford size (but not the Hanford type necessarily) for production of energy in the neighborhood of 10^6 kilowatts. The arrangement suggested was one in which one large mother plant would produce 49 for consumption in a series of smaller plants. In the mother plant, the energy produced would be used to reduce the cost of the 49 produced. (Mr. Fermi mentioned that he viewed the use of this power for the heating of cities with sympathy). There may be non-technical objections to this arrangement, for example, the shipment of 49 to the smaller consuming plants offers the serious hazard of its falling into the wrong hands, but these were to be omitted from this discussion.

The fundamental aim in the mother plant would be to get the maximum possible yield, with full utilization of the metal as the goal. If a solution to such a proposal can be found, then the schemes for isotope separation are not of great interest. If such a solution is not possible, then the schemes for isotope separation should undoubtedly be investigated further.

In the following discussion of full metal utilization, the isotopes 28 and 49 will be referred to as 8 and 9, respectively. In the reaction cycle suppose that one fission of 9 and ψ fissions of 8 take place in a single cycle of generation. Then the production of neutrons will be $\nu_9 + \psi\nu_8$. Some neutrons are lost in the moderator, coolant, etc. Let L = the number lost and α = the number used in producing 40-10. Then the excess of neutrons available for absorption by 8 to produce 9 will be

$$(1 - L)(\nu_9 + \psi\nu_8)$$

and the production of 9 per cycle will be

$$(1 - L)(\nu_9 + \psi\nu_8) - 1 - \alpha - \psi$$

The term $1 + \alpha$ represents the destruction of 9. Therefore, the ratio of production of 9 to its destruction, which we will call γ, will be

$$\gamma = \frac{P}{1+\alpha} = (1 - L)\left(\frac{\nu_9}{1+\alpha} + \psi\frac{\nu_8}{1+\alpha}\right) - 1 - \frac{\psi}{1+\alpha}$$

To utilize all the metal, γ obviously must be greater than 1. If γ is only very little greater than 1, the chain reaction would keep going with maximum economy of fissionable materials and would continue to go on until all the metal were used, but the value of such a pile would not be great and it would only be good for, say, hardening materials (the Wigner effect) or possibly (though less desirable) heating cities. The effective ν_9, is around 2.1 to 2.2.

Assume first a Hanford type pile with an equivalent amount of 49 substituted for the 25, i.e., in the early stages, 25 would be burned to produce 49 which would gradually improve its condition.

The earlier estimate of 1.9 for the ratio of the fission cross section of 49 to that of 25 has been more recently estimated by Y as 1.4. The ratio of absorption cross section for 49 to that of 25 is around 1.5. With these conditions, ν_9 is about 10% higher than it was previously thought to be. (The actual values of ν and ν-effective are not really known so the discussion can only show ranges). The situation then in a pile of Hanford design and lattice would be for a ν-effective (which will be referred to hereafter as μ) of 2 to 2.2, γ will be from 0.8 to 0.98. In the latter case, the pile is close to a balanced situation but not quite there. To adjust such a pile without drastic changes of design, large diameter slugs or more metal could be used to improve the thermal utilization and increase ψ. However, over-sized lumps increase the difficulty of cooling since the annular type cooling is badly limited in power production by the metal temperature.

The second type pile considered for the production of power was the P-9 moderated pile. For a μ of 2 to 2.2, γ would be 0.93 to 1.13. These values do not necessarily represent the optimum but are merely indicative of what can be done with P-9 piles and one with such a γ of 10 to 15% may or may not be an operable plant. The practical difference between continuous and discontinuous P-9 plants is not large in this respect since the loss by absorption for the coolant and its tubes practically compensates for the less efficient reproduction in slurry piles. One might hope to improve the situation by capturing the escaping neutrons in a reflector but the absorption in the pile container is an Important problem.

Another type of pile to consider is one with very little or no moderator (fast chain reacting type). From the nuclear point of view this is very desirable and is simple in principle but, practically, it involves serious problems in removing the heat. Ignoring the cooling, and considering only the nuclear point of view, this type pile may be of either one of two forms:

In Fig. A, a small spherical core of 49, say, 10 cm in diameter, would be surrounded by a sphere of 28 or normal tubealloy about 40 to 60 cm in diameter. This arrangement is good from a γ standpoint and one might expect a γ of 1.3 to 1.4, because L can be made small since the fast neutrons from the 49 get into the 28 readily. (Mr. Allison pointed out that if 25 is not considered for the surroundings here, thorium might be used). The pile shown in Fig. A only requires a few kilograms of 49. To utilize more 49 it would be possible to construct units like A with multiple 49 cores spherical or cylindrical in shape.

Fig. B represents a homogeneous sphere of 28 with 49 uniformly distributed throughout the mass, the whole surrounded by a reflector of pure 28 to catch the leakage neutrons. In this arrangement about 70% of the neutrons get into 28 immediately to produce fast fission. Assuming a mixture of 49 and 28 in which X represents the percentage of 49, critical conditions (i.e., where the chain reaction continues if the pile is of infinite size) would be reached with about 5% of 49 in the mixture (X = 0.049). For values of μ of 2 to 2.2, γ would be 1.37 to 1.57. As the pile size is decreased, the following results would be obtained. They are calculated without reflector.

Adding a reflector would decrease the critical radius of the active sphere by about 10 cm and improve very considerably the value of γ since the reflector would utilize the neutrons escaping from the active core. Taking the case of the 70 cm sphere above, this represents about 1.5 m3 or say 30 tons of the mixture. Therefore, 6% or about 2 tons of 49 would be required to keep this machine running. Thus a plant of this type requires a large quantity of 49 for operation although this is not sufficient reason for discarding this type of pile as a possibility.

Table 1:

Critical Radius of Sphere	X (fraction of 49)	γ $\mu = 2$	$\mu = 2.2$
100 cm	0.054	1.23	1.43
70 cm	0.060	1.10	1.30
50 cm	0.067	0.98	1.18

The serious objection to these fast chain piles is the removal of the heat. Since practically all the heat is produced in the 49 (about 70 to 80%), piles like those in Fig. A are harder to cool since it is mainly the tiny core which must be cooled while in Fig. B the whole mass is to be cooled.

As another possibility, a compromise enriched pile might be designed which would have enough moderator to reduce the percentage of enrichment required to keep the chain reaction going. But not as large an amount would be required for the conventional optimum conditions.

Mr. Fermi suggested that at a later meeting he would consider the question of how to use the 49.

Mr. Szilard was the second speaker and proposed approaching the problem from a different viewpoint,—that of assuming more optimistic values of the constants so as to indicate other potentialities. He pointed out that the fast reaction is preferable to the slow chain reaction for producing 49 from tubealloy and that this is probably more true if we assume more pessimistic values for ν or μ. Before discussing these values of the constants, sketches of a possible design were distributed and described briefly. These sketches are attached hereto.

The sketches show two different arrangements. In sketch A, the enriched tubealloy,(enriched to where the chain reaction will go) and natural tubealloy would be distributed in the form of rods in a cylindrical pile, in which the enriched material would be in the center portion of the rods lying within a circular area in the center of the pile. Part of the rods, located within three circular areas around the center (as indicated in Fig. 1) would be arranged so the cylindrical bundles could each be rotated about its axis. In each of the rotating bundles, part of the rods would be natural tubealloy and the balance of natural tubealloy with the center section enriched.

In the beginning, the enriched material in the three:bundles would all face the center of the pile and lie within a cylinder whose axis would coincide with the axis of the pile and whose cylindrical surface would pass through the three axes of the revolving bundles. By means of this arrangement, as the multiplication factor increased with the continued operation of the pile, the enriched material could be rotated away from the center of the pile and the natural tubealloy brought towards the center where it in turn would be enriched. In the center of the pile would be a single tube for introducing mercury, liquid bimuth, or some other absorbing or slowing material for controlling the pile. The coolant for this type pile would be a bismuth-lead alloy and would flow downward through the pile between the static and rotating rods. The possibility of using liquid sodium in place of bismuth-lead should also be looked into. The volumetric heat cabacity of the liquid sodium is about the same as that of the bismuth-lead alloy but its density would be 10 times less, so that the pressure drop would be about 1/10 that for the bismuth-lead alloy or the velocity about 3 times larger for equal pressure drop. In the scheme just described, the following approximate conditions

would obtain: (1) the bismuth-lead alloy would occupy about 1/3 of the enriched core and would pass through the pile at a velocity of about 15 meters per sec; (2) with 1/2 cm diameter rods raised to 700°C metal temperature at the center of the central rod and with 150°C temperature increase in the coolant, about 250,000 kW will be removed. The pumping power for the coolant will consume about 5% of the power produced.

In the alternative scheme B, control of the pile would be obtained by means of a nest of tubes for the mercury or other controlling medium arranged as in Figs. 3A and 3B and 4A and 4B. The metal rods would all be stationary and vertical (nos. 12, 13 and 14 in Fig. 3A) and would be about 1/2 to 1 cm in diameter by about 2 meters long.

In both designs, the enriched core would be about 1/2 to 1 meter in diameter by about the same height. The balance of the material around the core would be ordinary tubealloy of the same rod size. The total diameter and the height of the pile would be about 2 meters.

The objective of such a pile must be to produce as much extra 49 as invested. It is assumed that the production will be double the original investment. For every atom of 49 disintegrated, two atoms of 49 could be produced. Part of these will be produced in the enriched core and part in the surrounding natural tubealloy. Some of the production in the core will tend to leak out into the natural tubealloy and this leakage must be kept within certain limits. Then k will increase over a period of time. As the chain-reaction goes on, the multiplication factor k will then increase so that the controls must provide for this as well as the normal operating control of the pile.

In the slow chain reaction, 49 captures neutrons in radiative not fission capture to produce a new element which we will call super plutonium or 40-10. It is assumed there is a 50% chance that this new element will be fissionable. If it is not fissionable, it is assumed there is 50% chance that it will be formed only in negligible quantity in the capture of fast neutrons. Thus, there is a 75% chance in a fast chain reaction that we may use ν and not μ in getting the production balance ($\mu = 2.2$ neutrons per neutron absorbed, $\nu_{25} = 2.2 \times 1.175 = 2.6$ neutrons produced per neutron absorbed). As the energy of the neutrons increases from thermal to fission energies, it is assumed there is no decrease in ν. The main argument in favor of the fast chain reaction is that if a fission neutron is released in tubealloy, it causes fission in the 28 to produce 1.2 neutrons (fast effect). If all the neutrons are captured, the overall balance would be that for every atom of 49 destroyed, two atoms of 49 would be produced. One goes back into the chain reaction, the other replaces the 49 destroyed, providing a net gain in 49.

In experiments in which a Ra-B neutron source was surrounded by 28, measurements indicated a 5.3% increase in the number of neutrons and that 63% of the neutrons remained above the fission threshold. This means that the increase in the number of neutrons for an infinite sphere would be 5.3/(1 - 0.63) or 19.5%. If the fission cross section is taken at 0.35 and the inelastic cross section at 2.7 for a ν_{28} of 2.2 to 2.6 ϵ will vary from 1.18 to 1.245.

Referring to the value above of ν_{25} of 2.6, if we were to use the more optimistic results reported by Y (that ν_{49} is 20% larger than ν_{25}) then ν_{49} equals 3.1 neutrons produced per neutron absorbed. If we are less optimistic and assume ν_{49}-effective = 2.5 but use the 19.5% increase indicated by the experiment mentioned above, we have three neutrons produced in a mixture of 28 and 49 for one atom of 49 destroyed.

It has been suggested that one of the subjects for one of the meetings soon to be held would be

a review of the availability of the metal producing ores and other sources of tubealloy. This is to be given by Mr. P. Morrison.

G MUC-LAO-18: Notes on Meeting of April 28, 1944

9:00 - 10:30, 209-Eckhart

Present: Fermi, Allison, Wigner, Smyth, Szilard, Morrison, Watson, Feld, Hogness, Young, Gleinberg, Creutz, Cooper, Vernon, Ohlinger

The first speaker was Mr. P. Morrison; his comments follow: If we view the chain reaction merely as a source of unspecialized energy it goes directly into competition with many existing large scale prime movers and fuel sources. Economics are then paramount. It is Mr. Morrison's opinion that we should not move in that direction for some time to come, but should attempt to exploit the particular properties of the chain reaction as a concentrated source of a highly special form of energy. Power simply as kW hours should appear at most as a by-product.

The following data was gathered from the Bureau of Mines bulletins and other sources and correlated by Mr. Morrison.

In 1941 the average yearly power produced by the major sources was as follows:

Water	25×10^6 kW
Gas and Petroleum Products	340×10^6 kW
Coal	470×10^6 kW

The first item covers mostly government ana private utilities and private industrial power developments. Of the power produced from the second fuel source about 40% was from gasoline in all its uses. Of the power produced from coal, about 19% was used in public utilities, about 10% for coke and chemical uses, about 20% for railroad transportation, and the balance, of about 60% for other miscellaneous uses including domestic, and industrial fuels. Of the above total figures for the world power production, the U.S. consumed about 35% of the coal-produced power and about 66% of the petroleum-produced power.

Cost figures for the above power, while not on a strictly comparative basis, are interesting. The figures below are all per megawatt hour.

Government hydropower	1.20
Private hydropower	2.00 to 2.50
Most efficient coal-steam plants	0.80
City consumer	30.00
Large industrial consumer (firm power)	4.00
Rjukan	0.40
(Energy, not mechanical power)	
TNT	200.00
Hi-octane gas	4.50
Motor fuel	2.00

The first item is based on the cost of the power at Boulder Dam at the bus-bar face on a guaranteed consumption continuously throughout the 24 hour day and includes the cost of the original equipment, maintenance, etc. The figure for the coal-steam plant is based on the fuel cost

only and does not include the equipment, maintenance, etc. As Mr. Szilard pointed out, these two cannot be compared and, as Mr. Fermi observed, it is the 80 ¢figure of the coal-steam plants with which we must compete in the production of power.

As Mr. Morrison noted in a recent memorandum, the approximate abundance of tubealloy and thorium in the top crust (5 to 8 km thick) of the earth is as follows:

Tubealloy: 4 ± 2 ppm (found mostly in granite and does not include sands, etc.)
Thorium: 12 ± 4 ppm (found mostly in sedimentary rocks)

The most efficient gold extraction plant yet constructed has been able to dig the rock, crash it, treat it, etc. to recover the gold for about $7.00 per ton. Crushing rock requires about 100 kW hours per ton when the rock is reduced to particles of about 1 mm3 in size. Therefore, Mr. Morrison estimates that we could afford to work the rock for power (based on using the 25, not the 28), if necessary, providing the abundance of the tubealloy was not less than 5 to 10 ppm. It has been estimated that there are about 10^{14} - 10^{15} tons of natural tubealloy to be found in rock in the earth's crust and about 10^{8} tons in sea water (the concentration in sea water is about 10^{-9} gms of tubealloy per gm of water).

The richest occurences of natural tubealloy in the world are at the following locations. All figures are very rough estimates based on incomplete and often reluctant surveys by the producers. All tonnage figures are tons of tubealloy and not ore.

U.S.	(mainly Colorado vanadium ores or Carnotite, having a content of about 1% tubealloy. Estimated at 3,000 tons of tubealloy available in 1925.)	about 5,000 tons in sight.
Czech	(also about 1% content)	about 1,000 tons remaining.
Congo	(of five sites total, one was estimated in 1925 at not less than 5,000 tons of ore having 20% tubealloy content)	about 5,000 tons in sight.
Canada	(although other deposits undoubtedly exist, the known deposits containing 20 to 50% tubealloy content ore have)	about 5,000 tons.
Russia	(1 to 2% tubealloy content Carnotite vanadium ore deposits)	not less than 3,000 tons in sight.
		Total about 20,000 tons of tubealloy in sight.

A similar table of the distribution of thorium throughout the world follows. This is found almost entirely in monazite sand having about 5 to 11% thorium content:

If we were to use the entire U.S. and Canadian resources of tubealloy (10,000 tons) using a γ of 0.9 and utilizing only the 25 and not the 28, we would have an equivalent replacement factor of only nine months for the total power consumption of the U.S. or about ten years for the hydropower.

US:	(mainly in North Carolina with a small amount in Idaho and Florida)	about 1,000 tons.
Brazil	(on the coast)	about 80000 tons.
British India		about 150,000 tons.
Netherland East Indies		may have about 5,000 tons.

The second speaker was Mr. Szilard who continued his discussion from the previous meeting. He recapped first the three possibilities as he saw them:

1. Unseparated tubealloy —> 49 production
2. Enriched tubealloy —> 49 production
   (a) slow chain reaction
   (b) fast chain reaction
3. Enriched tubealloy —> no 49 production

On the basis of Morrison's report, Mr. Szilard felt that the tubealloy should be utilized more efficiently, *i.e.*, using the 28 and not just the 25. However, since the power production indicated in item (3) above is a long term proposition, he did not intend to discuss this phase at great length at this time.

In item (1) above, heat is only a byproduct and not the primary object. Concerning item (2), Szilard proposed answering the question if an amount A of 49 were invested, how long would it be until 2A of 49 were obtained. In a fast chain reaction, if two tons of enriched ore containing 10% of 49 were used in the core surrounded by 28 at the rate of 125,000 kW, then 2A of 49 would be produced in 4-1/2 years. In order to have any practical significance, this time should not be very much larger and the readjustment of the material should be easy during the time of operation.

Considering first the slow chain reaction: assume a η_{49} (neutrons emitted in fission/neutrons absorbed) equal to 2.0 - 2.2. Szilard struck out the latter figure when Fermi stated that the η_{49} is probably lower than η_{25}. With $\eta = 2$, just as much 49 is being produced as is being destroyed. In a slow chain reaction this might be improved by using the fission of 28 mixed with 49 but this is not very effective in a slow chain reaction because even from large lumps embedded for instance, in graphite, many neutrons escape, are slowed down, and do not produce fission, Since the super-plutonium (40-10) formed might be fissionable, there is, say, a fifty-fifty chance that we can improve η to a $\eta_{\text{eff}} = 2.5$.

Considering the fast chain reaction, the situation is more favorable. With a low concentration of 49 in the mixture with 28, experiments have shown that ϵ might be raised to 1.2. In addition, there is a high energy tail producing an (n, 2n) reaction which may give a 2.5% increase in η (based on observations of Turkevich).

(The value η has been defined as the number of neutrons produced/number absorbed in 49. Szilard uses ν_0 defined as the number of neutrons produced/number of fissionable atoms used up.).

In a fast chain reaction, even if 40-10 is not fissionable, Szilard felt that it is probably true that the branching ratio for 40-10 moves in a favorable direction, or that ν_0 may be taken as $\nu_0 = 2.5$. He felt strongly that there is a very good chance that Pu-240 is either fissionable in the thermal region or at least that the branching patio can be counted upon to decrease by a factor of 3 as one goes from thermal energies to, say, 1 MeV. (Fermi pointed out that the branching ratio of 49 is greater than that for 25.)

The arguments for this belief is in part based on the rule of thumb (ΔM/M) - (2ΔZ/Z) (see also Morrison in Project Handbook, Chapter IV B 1.1) which gives a rough indication of the fission threshold and is partly based on the belief that, with increasing neutron energy, the time required for fission decreases whereas the time required for radiative capture remains constant. Szilard therefore assures that, in a mixture of 238 and Plutonium, $\epsilon\nu_0 = 1.2 \times 2.5 = 3.0$ neutrons emitted per thermally fissionable atom destroyed aad this would mean that there is a net gain of one thermally fissionable atom per similar atom destroyed.

Referring to item (3), Szilard emphasized one possibility, the burning of Plutonium in a slow reaction and absorbing the neutrons by bismuth to give Polonium. Of the heat dissipated when Plutonium is destroyed to give Polonium, only about 3% would be stored in the Polonium. However, this energy will be available for use free of γ radiation and could be used for driving airplanes, etc.

In the discussion following, Fermi questioned the estimated value of $\nu_0 = 2.5$ on the ground that it might be too optimistic and pointed out that there is a long range future in developing the full utilization of 28 and thorium.

Wigner questioned the feasibility of the rotating disc arrangement described at the previous meeting on the ground of poisoning and questioned the 4-1/2 year investment return. He felt this would probably be more newly 10 to 20 years by which time, as Mr. Morrison suggested, we may be burning water.

After discussion, Szilard expressed his view that item 2a is of more immediate concern than 2b.

Mr. Morrison suggested that more work should be done on the nuclear development of thorium because of its greater availability and also suggested experiments to obtain ν_{23} and other useful constants.

Mr. Ohlinger explained very briefly an outlined program, of which a copy is attached hereto. This program for consideration of future trends in the laboratory work, is a compilation of the independent results of the "homework" requested at an earlier meeting. It has been assembled to give a majority concensus of the classification of possibilities.

In general, the outline is first divided into three main groups based on the use to which a pile would be put. Thereafter, the potentialities are subdivided for easy consideration. An open discussion of this outline will occur at the next meeting on Friday, May 5th.

I. POWER PRODUCTION (49, 25, or 23 consumed)

A. Purpose of power (in order to determine following)

1. Physical size and mobility
   a. Large stationary piles (slow chain reacting or group of fast chain reacting) such as for central stations
   b. Medium mobile (or stationary) piles (slow chain reacting or group of fast chain reacting) such as for boats or locomotives
   c. Small mobile piles (fast chain reacting) such as for planes or cars
2. Power output range, *e.g.*
   a. over 10^6 kW
   b. between 10^3 - 10^6 kW
   c. less than 10^3 kW

B. Utilization of Energy

1. Direct.
   a. Electrical removal (including thermocouple type)
   b. Working fluid absorbing heat directly
      - Direct liquid vaporization in pile to operate turbine, etc.
      - Gas cooling with or without gas turbine
      - Endothermic chemical reactions, *e.g.*, $2H_2O \longrightarrow O_2 + 2H_2$
2. Indirect Energy removed by circulating
   a. the metal
   b. the moderator (if any)
   c. the coolant

II. ISOTOPE PRODUCTION (49 or 23 produced)

(Since its purpose is well defined and there are no limitations on size, lack of mobility and power produced, except by the operating potentialities of the design chosen, these factors are not outlined here.)

A. 49 Production

1. Slow chain reaction (thermal neutrons) using normal U
   a. Homogeneous piles moderated with
      - P-9
      - P-9 + H_2O
   b. Heterogeneous piles moderated with
      - Graphite - improvement in operation and utilization of present piles (X and W) and design new improved piles

- P-9 - also require a separation process to recover P-9
- P-9 + H_2O - also require a separation process to recover P-9
- Be
- BeO

c. Cooling accomplished by moving
- the metal (UF_6, molten U, solutions)
- the moderator (P-9, P-9 + H_2O)
- a separate coolant (H_2O, P-9, liquid Bi or Bi-Pb alloy, diphenyl)

2. Enriched piles (enriched with 49 or 25)
   a. Slow chain reaction as outlined above
   b. Fast chain reaction as outlined below
3. Fast chain reactions (no moderator)
   a. Homogeneous piles
   b. Heterogeneous piles
   c. Cooling accomplished by
      - Liquid Bi as coolant
      - Liquid Na as coolant
      - Using U molten and moving it

B. 23 Production

1. Reflectors

Comment: If power produced in making 49 can be used practically, this constitutes another case, – a combination of I and II.

III. RADIATION SOURCE (High levels of intensity)

(Since the purpose is well defined and size, mobility and power produced are not criteria, they are not outlined here. However, such a pile must be flexible with the ability to reach high levels of intensity and expose materials or take measurements with ease. Flux and not total power important here. Most suitable of piles defined in II to be chosen as experimental machine. Routine metal removal is not important.)

PROBLEMS COMMON TO ALL PILLS (or a large number of them)

1. Shielding
2. Reflectors

H MUC-LAO-19: Notes on Meeting of May 5, 1944

9:00 - 10:30, 209-Eckhart

Present: Allison, Szilard, Wigner, Weinberg, Morrison, Creutz, Vernon; Young, Watson, Ohlinger

Please note a correction in the notes of the last meeting, Friday, April 28th. On page 2 at the top, the cost of the energy from TNT should read $200.00 per megawatt hour instead of $2.00.

Mr. Morrison started the discussion by noting the above correction and adding, at the request of Mr. Wigner, a rough figure for the cost of energy obtained from tubealloy on a comparative basis. Assuming a cost of $2.00 per pound for tubealloy and assuming that all the 25 is used, the cost of energy (not mechanical power) would be about $0.02 per megawatt hour. Mr. Morrison noted that the least expensive horsepower (based on first cost) for a prime mover is an airplane engine, and the most expensive is the old fashioned steam locomotive. Mr. Morrison also transmitted the following information from Mr. Z. Jeffries. At the request of the WPB but with only limited resources, the U.S. Geological Survey has been carrying on a research into the abundance of various materials found in pegmatites. Among the many materials to be surveyed were tubealloy and thorium. This survey is to be published in one or two months and copies will undoubtedly be obtained by the laboratory. By word of mouth, Mr. Jeffries did obtain the following advance information based on the preliminary results, — a large number of granite bodies were found to contain up to 100 ppm of tubealloy and a small number up to 1,000 ppm.

Mr. Ohlinger carried on the discussion from this point with a review of the outline appended to the notes for the last meeting. A general discussion followed, of which the highlights follow.

Mr. Szilard pointed out that in our discussions we must not overlook the peace time uses of this power. We can only hold the advantage we have obtained in our development of this process in America if its peace time use is well developed. Following this vein of thought; Mr. Wigner said that very little has been said about this phase of the subject but that he had one or two suggestions to offer. By subjecting the tubealloy bearing granite mentioned by Mr. Jeffries to a bombardment of neutrons, it might be possible to obtain a mechanical dissolution or porosity such that nitric acid could readily penetrate the granite to dissolve out more tubealloy. Another possibility is a polymerization of hydrocarbons to produce synthetic rubber.

Mr. Allison suggested that in projects where the transporation of fuel is a major problem, such as an exploration of the South Pole or other distant objectives, a small unit would be very useful. He also repeated the previous suggestion for heating entire cities since it would also eliminate the terrific nuisance of the usual smoke pall.

Mr. Wigner observed that the age of technical problems is past. The only obvious needs are probably large scale heating and fuels. Mr. Szilard felt that if there are no present needs, then new needs should be created. Mr. Morrison suggested stellar travel.

Mr. Ohlinger brought the discussion back to the outline with the suggestion that the subject of the direct utilization of energy by electrical removal (IB1a in the outline) should be dismissed from our thinking for the most part. He mentioned that little has been forthcoming on this subject outside of the suggestions which accompanied Mr. Wigner's "homework". One of these suggestions was to use the active material in the form of an extremely thin wire surrounded by a metallic tube, with the interspaces evacuated. The fission products would assume a considerable charge before leaving the

thin wire and by virtue of their kinetic energy would he able to reach the tube surrounding the wire even if it had a considerable positive charge. For a reasonable utilization, the potential difference between the tube and wire would have to be of the order of magnitude of 5,000,000 volts and one could obtain a current of about 50 amperes if the system were run at 500,000 kW. The wire and pipe would have to be cooled and, although, this is, in a way, the most direct utilization of the energy of fission, it obviously gets into great technical difficulties, is not very efficient, and does not furnish the power in a very suitable form, so it is noted only as a curiosity. The second suggestion was Mr. R. Williamson's idea to extract the heat from a pile by using the Peltier effect to convert the energy directly to electrical energy. However, the thermoelectric constants of tubealloy are not known and one can estimate that the amount of power obtainable from a pile like that at W would only be of the order of 5,000 kW. One arrangement for accomplishing this would be to have every tubealloy rod be one electrode of the thermocouple and the graphite the other. The pile would be run at low power level with reasonably good efficiency despite the heating loss. Another arrangement would be to subdivide the tubealloy rod into short sections, cooling one end of each. The cooled end would then form the cold junction, the hot end, the hot junction. However, utilization of power in such a system is much poorer. Mr. Szilard asked for the efficiency of such a pile. Mr. Watson suggested that it would probably not exceed 1% although Mr. Wigner was of the opinion that this figure was too low. Mr. Szilard thought the subject could bear further investigation, but Mr. Wigner pointed out that by high temperature operation (700°C) it is possible to obtain efficiencies up to around 65% and so, as long as we would just be making kilowatt hours, we should abandon all "crazy schemes" and think seriously about high temperature operation. "Goldbergs" are not attractive for power production and Mr. Allison thought it would be much easier to develop the high temperature operation.

Items IB1b (2) and (3) in which the working fluid absorbs the heat of reaction for direct utilization of the energy offered more interesting possibilities according to Messrs. Wigner, Morrison and Vernon. Mr. Wigner noted one difficulty with the endothermic chemical reactions, — the gases which have these reactions usually react chemically with the pile materials.

Mr. Szilard stated that when the supply of petroleum is exhausted, it might be possible to break down the CO_2 molecule and hydrogenate the carbon atom to make synthetic hydrocarbons for gasoline, etc. Mr. Morrison suggested an alternate of breaking down the H_2O molecule and carbonizing the hydrogen atom.

Mr. Ohlinger suggested that in order to speed up the discussions at these meetings and avoid wasting the time of the entire group with detailed discussions of schemes which do not hold much promise for next year's program of research work for the laboratory, assignments should be made of individual pile types for investigation and group discussion outside of the regular meetings. The individual assigned any particular type would make it a point to investigate the advantages and disadvantages of that type as thoroughly as possible and discuss these and any design details with other members of the group who would be likely to have constructive information on the subject. Thereby, the problem would be well investigated prior to the meeting at which it would either be presented in concise form for discussion as a likely problem for investigation, theoretically or experimentally or both, or else indicated as an unpromising project which should not be considered at this time. Problems common to several designs could then be correlated for an experimental

program and the more promising of the designs brought forth by these investigations outside of the regular scheduled meetings could be discussed by the group as a whole before being turned over to the various divisions for their detailed development. This proposed policy was accepted by all and the following assignments made.

Mr. Wigner will lead the investigation into the possibility of power producing piles utilizing the energy directly by endothermic chemical reactions [Item IB1b (3)]. Mr. Vernon will do the same for gas cooling [Item IB1b (2)]. In order to give them time to prepare these assignments, it was suggested that Mr. Young speak at the next meeting on Wednesday, May 10th on "Suggested Improvements for a Hanford Type Pile" and on Friday, May 12th, Mr. Weinberg on "Conversion Units".

I MUC-LAO-30: Notes on Meeting of July 6, 1944

9:00 - 10:30, Eckhart-209

Present: Messrs. Seitz, Hogness, Allison, Szilard, Young, Vernon, Weinberg, Smyth, Cooper, Creutz and Ohlinger

Mr. Seitz presented a very interesting report on his visit to the General Electric Company at Schenectady to discuss turbine systems and in particular, the new mercury turbine and mercury-vapor processes.

This subject is of interest to us because, first, the mercury "topping" system is the most modern and efficient power production system in use today and, second, it presents something new and different in power production—the use of liquid metal at high temperatures. The latter is of particular interest to us because of its application to certain of the potential pile designs involving the use of liquid metal at high temperatures.

The mercury-vapor process is based on the fact that to increase thermodynamic efficiency, one should obtain the maximum possible working temperature or rather the maximum possible differential temperature. The mercury system which is a "topping" system would operate between two temperatures T_1 and T_2 to produce about 1/3 to 1/2 of the total power while the steam system would operate between temperatures T_2 and T_3 to produce the balance of the power, about 2/3 to 1/2. Most of the mercury-vapor systems in use now, have the temperature T_2 established according to the top temperature available in their existing steam system, with the mercury system added to bring the top temperature up to T_1. The engineers at Schenectady would much prefer to lower the value of T_2 and recommend that new systems which are designed provide for a lower T_2 in order to give a better overall efficiency.

The mercury-vapor process is a binary system for producing power from fuel with greater thermal economy than is possible with the steam cycle alone. The mercury cycle can also be considered as a steam producer in which, for a given amount of fuel, nearly as much steam is produced as in a steam cycle and, in addition, the by-product power from the mercury turbine generator is obtained at nearly the mechanical equivalent of the thermal energy. The advantage of the mercury over the water system is that one obtains as high temperature with mercury vapor as with water vapor at only 1/10 the pressure. One disadvantage of the mercury system is that the weight and volume of mercury required is larger than that of the water required in a water system. In a water system operated at about 950°F, the entropy per pound is 1.546 BTU/°F/lb and the volume of vapor per pound is 0.3538, while in a mercury system operating at about 1050 to 1100°F, the entropy is 0.1193 and the volume per pound is 0.3998. From this we see that a mercury system requires about ten times the working volume of a water system.

The present mercury units are designed to.produce about 20,000 kW in the mercury system alone and one unit uses about 400,000 pounds of mercury. At the present market cost of mercury of around $2.00 per pound, this means an outlay of nearly $1,000,000 for mercury for one unit. The known world deposits of mercury are large and with a production no greater than the present top yearly production, as much as 1,000,000 kW in combined mercury-steam plants could be installed each year. Whereas these binary units might be installed on large ships, at present they do not look very practical for smaller power requirements of a mobile nature such as trains, etc.

Although the figure above shows the utilization of 20 pounds of mercury per kW, the engineers at Schenectady believe that a minima of 8.25 pounds of mercury/kW can ultimately be achieved. They have set up as a basis of comparison of efficiencies a figure based on the total overall efficiency from the heat contained in the coal to the power at the busbar. On this basis, the maximum theoretical efficiency which can be obtained according to the first law of thermodynamics is 3,413 BTU/hr/kW. The best overall efficiency obtained to date in any of the mercury-steam combination units is 9,175 BTU/hr/kW. The best overall efficiency obtained from an all steam unit is 10,039 BTU/hr/kW. This means the all steam power plant has an efficiency of about 33% compared to the first law efficiency noted above while the binary system has an efficiency of about 37% or 4% gain over the all steam plant. The operating temperatures for a typical binary unit are: 1050°F for the mercury or T_1 (this is saturation temperature) and 925°F for the water or T_2 (this included the superheat). Thus the thermodynamic efficiency computed from the temperatures or the second law efficiency would be about 65%.

The drawing attached illustrates diagrammatically the mercury-vapor process for the production of power. Referring to the drawing, mercury is vaporized in a boiler at comparatively low pressure and passed through a mercury turbine which drives a generator. The vapor from the turbine is exhausted to a condenser boiler where its latent heat is transferred to water which vaporizes at any desired pressure. The steam formed in the condenser boiler is superheated in coils located in the gas passages of the mercury boiler and is then used in steam turbines or for process work.

The top limit to the temperature T_1 at present is set by the tubes in the fire box. These are actually located in the hot coal gases immediately above the fire bed and have temperatures on the outside surface of around 1200°F and on the inside of around 1075°F. Under these operating conditions, it is obvious that the tubes are easily attacked by the gases and slags in the fire box and must have high creep strength as well as stainless properties. The best alloys found to date for these tubes are the Sicromo series having about 1% each of silicon, chromium and molybdenum with the balance iron. When better alloys are found, T_1 can be raised above 1075°F and the efficiency improved. However, in our type of power production units, we do not require a furnace and so we can better the value of T_1.

Mercury, as the material for the liquid metal portion of this binary system, is a good material for this purpose because it dissolves very few metals used in commercial high temperature practices. In fact, the solubility of all metals in mercury is less than one part in 10^4. One problem, however, with the use of mercury was the difficulty in obtaining good wetting of the boiler tubes by the mercury. This is especially necessary in the fire box and condenser boiler to obtain good heat transfer. It was finally found that by adding 0.5 ppm of titanium and 10 ppm of magnesium as wetting agents, good heat transfer coefficients could be realized. These wetting agents are probably effective because they are both good oxygen getters, taking it away from the mercury and the iron. Mercury probably tends to wear away the iron oxide while the titanium and magnesium, in taking the oxygen away from the mercury and iron, help the mercury in cleaning off the scale and keep the tubes clean and easy to wet. When starting up a unit, it takes about ten minutes before wetting occurs but from that time on, no further trouble is encountered if about one pound per month of titanium and possibly a very small amount per year of magnesium is added. Unfortunately, a small amount of air (about 1 cu ft per hr) leaks into the mercury turbine at the condensing or low pressure end. This probably

accounts for the necessity for the extra titanium.

In an all steam power plant, an entire corps of chemists is required for constant water analysis and treating to avoid scaling while the mercury system is practically self-maintaining except for the small addition of titanium. Therefore, in addition to the increased efficiency, the mercury system offers a tremendous reduction in maintenance costs.

One great worry in a mercury system is the possibility of leaks in view of the high mercury costs. In the many operating years credited to mercury-vapor binary systems, only one leak has occurred. This was brought about as a result of the following process: powdered coal was fed into the top of the fire box while ashes produced therefrom fell into the bottom of the furnace and were swept out by streams of water. In the case of the single leak, water splashed up and caused steam which attacked the tubes with a resulting loss of about one ton of mercury through the leak.

Despite all premonitions, the mercury turbines have caused no trouble and have operated very successfully without pitting or erosion. The careful design of the turbine to produce impact incidence of the mercury with the turbine blades at a safe angle instead of perpendicular appears to be the secret for the successful erosion resistance of these turbines. They are made with high carbon steel parts instead of stainless steel as employed in steam turbines. However, at present, the steam turbine is more efficient than the mercury turbine based on the overall transfer of kinetic energy in the gas to mechanical energy from the turbine. For a steam turbine the efficiency is about 85% as compared to only 73% for the mercury turbine. The latter figure is lower probably because of the tendency of the mercury to condense in the turbine.

Of the heat produced in the furnace, only about 15% is lost to the stack. The operating pressure for the mercury system is around 125 pounds per sq.in and for the water system around 1,250 pounds per sq. in.

To consider the application of this information to our problem of designing a new high temperature pile, we must consider the possibility of a metallic alloy of uranium or plutonium which can be used molten through a pile which would replace the fire box in the above process. There are only two systems giving a eutectic with uranium (and presumably plutonium) with low enough melting points to be practical. These are the iron-uranium (or plutonium) alloys and the nickel-uranium (or plutonium) alloys. The latter eutectic melts below 750°C and has about 40 atomic percent nickel. The difficulty with the first named eutectic is that iron dissolves everything which can possibly be used for the tubes.

Very little is known about the mercury-uranium alloys which are pyrophoric. More data should be obtained on the phase diagrams of this alloy and further information obtained on the nickel-uranium alloys. Of course, the pile need not be operated with the active metal in molten form circulating through the pile but may have another metal in liquid form as the coolant with the uranium or enriched material stationary within the moderator. An example of this type is Mr. Szilard's bismuth cooled pile.

Another problem in using a uranium eutectic is that the fission products will be contaminating the liquid metal constantly. At Hanford, this may take only a few hours. While most of the fission products are good elements and might help as wetting agents, iodine and the alkaline metals might interfere with the cleansing action.

In the discussion that followed, Mr. Allison pointed out the possibilities of a bismuth-uranium

(or plutonium) eutectic instead of mercury-uranium. The question was also brought up as to the relation between wetting and heat transfer and it was pointed out that wetting was very important on a rough surface and less important on a smooth surface. Mr. Ohlinger questioned the supposed reduction in maintenance in the binary system and Mr. Seitz agreed that there was no reduction in maintenance in the present units where the mercury system has been added simply as a "topping" unit on an existing steam plant but that any newly designed unit where the temperature T_2 can be reduced, it is to be expected that maintenance will be reduced accordingly.

Mr. Vernon concluded the discussion period with a brief discourse on a high temperature gas cooled pile in which the hot gases would replace the coal gases in the furnace in the binary system, the active metal would be located throughout the moderator in lumps either of uranium carbide or molten uranium contained in crucibles, say, of beryllium oxide. The furnace in the binary system would then become essentially a heat exchanger for absorbing the heat carried off by the cooling gas through the pile. Mr. Cooper's objection to this scheme was the high gas pressure that would probably be necessary in order to get sufficient heat capacity in the cooling gas (probably around 10 atmospheres). Mr. Weinberg's objection was that such a pile would be exceedingly large because of the high temperature operation and the use of the carbide. In fact, he felt that it would be much larger even than the original helium-cooled graphite-moderated plant. Mr. Hogness objected to the large amount of power that would be used up in recirculating the gas because of the low heat capacity of helium.

At subsequent meetings, Mr. Wigner will discuss his "pulsating" pile and those utilizing endothermic chemical reactions and lir. Szilard will discuss his seed piles.

J MUC-WHZ-FF-169: Importance of 24 for Thermal Breeders, April 15, 1945

The recent experiments on the properties of 23 indicate capture of neutrons in 23 leading to the formation 24. That is to say, $\alpha_{23} \neq 0$. It is therefore desirable to consider the effect of 24 on the problems of a 23 breeder. If the 24 and 25 which is produced are kept in the reacting unit indefinitely while the 23 is replenished from the production in thorium, the amount of 23 produced in thorium per destruction of 23 in the reactor is (in equilibrium) not the usually quoted result:

(1a) $\eta_{23} - 1-$losses to moderators and structural materials, but this result plus

$$\frac{\alpha_{23}}{1+\alpha_{23}}(\eta_{24}-1) + \frac{\alpha_{23}}{1+\alpha_{23}}\left(1-\frac{\sigma_{f24}}{\sigma_{a24}}\right)(\eta_{25}-1)$$

The relative number of 24 to 23 present is

$$N_{24}/N_{23} = \frac{\sigma_{a23}}{\sigma_{a24}}\left(\frac{\alpha_{23}}{1+\alpha_{23}}\right)$$

and the 25 is present as

$$N_{25}/N_{23} = \frac{\sigma_{a23}}{\sigma_{a25}}\left(\frac{\alpha_{23}}{1+\alpha_{23}}\right)\left(1-\frac{\sigma_{f24}}{\sigma_{a24}}\right)$$

The lower η_{23} proves to be, the more important for the breeder the terms in (1b) become. It is quite possible, on the basis of our present knowledge, that they may increase the rate at which the 23 supply may be accumulated by a factor of 1-1/2 to 2 times that predicted from (1a) alone.

The great significance for 23 breeders of the terms (1b) makes it appear advisable to add to the program of measurements at the Argonne Laboratory a series of determinations of the properties of 24 — $\sigma_{f24}, \sigma_{a24}, \eta_{24}, \nu_{24}$. For this purpose it is necessary to obtain samples containing 24 in moderately high concentration, the higher the better.

Another possibility is that 24 may be considered as a valuable isotope for fast reactions. In this case instead of accumulating pure 23 it would be possible to accumulate a mixture of 23, 24 and 25. The pure 23 produced in the thorium would be fed back into the breeder, and the gain taken out from the reactor region where 23, 24 and 25 would be extracted. In this system (at equilibrium) the total amount of 23, 24 and 25 per destruction of 23 is also $\eta_{23} - 1 -$ losses $+ \frac{\alpha_{23}}{1+\alpha_{23}} f(23,24,25)$ where $f(23,24,25) \approx 1$ and depends on the properties and 23, 24, 25. Whether $f(23) = \eta_{24} - 1 + \left(1-\frac{\sigma_{f24}}{\sigma_{a24}}\right)(\eta_{25}-1)$, which is the corresponding function taken from (1b), is bigger or smaller than f(23,24,25) depends on the details of the properties of all the elements. For example, however, if $\sigma_{a24} = \sigma_{f24}$ and $\eta_{24} = 1.5$, f(23,24,25) is very close to 1 but f(23) = 0.5. That is to say, in this case the possible 23 + 24 production is significantly greater than the possible 23 production. The possibility of mixed 23 and 24 production makes it again important to find out about the properties of 24 not only in the thermal region but also for fast reaction.

The measurements of the properties of 24 which are suggested here are exceedingly difficult with the amounts and concentrations currently available. Probably only the fission cross-section

can be known until there are larger quantities and higher concentrations. In measuring the absorption cross-section at the present time the experimental inaccuracies are of the same magnitude as the result to be expected. Other measurements seem to be even less promising. The main purpose of this note is to point out the importance of obtaining good samples of 24 for future work. η_{23} does not tell the whole story of the thermal breeder. Eventually the thermal breeder may stand or fall according to the properties of 24.

F.L. Friedman

K MUC-EPW-134: Preliminary Calculations on a Breeder with Circulating Uranium, May 17, 1945

E.P. Wigner, A.M. Weinberg and G. Young

The following constitutes our present ideas on the breeder which, no need to say, are very preliminary and which we hope will undergo considerable changes before they will crystallize. Under ordinary conditions we would not report on work which is still as much in flux as the present one is. However, we had to shift our attention, at least temporarily, to another subject and we want to record for this reason the results obtained so far.

A. General Considerations.—The breeder is an arrangement in which 23 undergoes fission and the neutrons produced thereby are absorbed by thorium. When this thorium decays it produces again 23 via Pa. Since the η of 23 is, according to Anderson and May, 2.37 the net increase in the k number of neutrons per 23 destroyed is 1.37. If all these neutrons ware absorbed in thorium, we would obtain 1.37 23 atoms per 23 destroyed. It is well to remember, however, that if only as many as 15% of the 2.37 neutrons emitted are lost by escape from the system or parasitic absorption by other materials or otherwise, the efficiency of the breeder goes down to 1 and no increase in the amount of 23 results.

Another point that is important is the power to product ratio of such a breeder. Let us assume that the total losses in neutrons amount to only 7-1/2%. In this case 1.19 atoms of 23 are produced per 23 atom destroyed. Thus the net gain in 23 is 0.19 atoms per 23 atoms destroyed or per 0.89 atoms 23 undergoing fission. As a result, the power output per net production is about five times greater than in the conventional piles.

During wartime it might possibly be more important to operate the systems to be described as converters of 49 to 23. In this case the above considerations do not hold necessarily and the efficiency does not have the paramount importance which it will be given in the following pages. If we disregard this fact, the main importance of the breeders is that they eventually permit the utilization of all the natural thorium. If one wants to make use of the full amount of energy one will have to construct a system in which the power generated by the breeder is utilized, *i.e.*, in which the breeder acts not only as a producer of 23 but also as a power source. This will demand high temperature operation of the breeder. If only the energy of the 23 produced by the breeder is utilized, the energy of at least 1/1.19 $\approx$ 84% of the Th is wasted if we adopt the numbers of the preceding example.

An enumeration of the losses follows:

1. Delayed neutrons which may be released outside the pile.
2. Absorption by materials present in the chain reacting part, such as the moderator and anion in case of solution, Bi and tubing in case of a second system to be described below.
3. Absorption by fission and corrosion products in pile.
4. Absorption by tank.

5. Escape in wrong direction.
6. Absorption by 24 formed.
7. Absorption by Pa.
8. Absorption by fission and corrosion products in reflector.
9. Absorption by moderator and coolant in reflector.
10. Escape from reflectors, particularly of neutrons formed by fission in reflector.
11. Losses in chemical separations.

It may be well to introduce, at this point, the notion of the efficiency of the breeder. This is the net gain in the number of 23 atoms per 23 atoms destroyed. In the ideal case, it is 37%, in the example discussed above only 19%.

It will be seen in the next section that the arrangements which we think are most promising consist of essentially two parts: a chain reacting unit containing the 23 (the "pile") and a reflector containing all or most of the thorium. If all the thorium is contained in the reflector only 1/2.37 = 42% of the neutrons will be absorbed in the pile and 58% will be absorbed in the reflector. Hence a relative neutron loss ℓ inside the pile entails the loss of the fraction 0.42ℓ of all the neutrons. A relative neutron loss in the reflector gives a loss of the fraction 0.58ℓ of all the neutrons. The loss in "efficiency" caused by some impurity which absorbs in the pile 0.01 times as many neutrons as the 23 is 1%, the loss in efficiency caused by an impurity which absorbs in the reflector 1% of all the escape neutrons is 1.37%. A loss of 1% of all the neutrons entails a loss of 2.37% in the efficiency. All losses must be subtracted from the original 37% efficiency.

The present report assumes that the active material in the pile is 23 because only this is known to have an considerably in excess of 2. It is worth noting, however, that 49 is an equally or even more suitable material for the same purpose if 40 is slow neutron fissionable. According to Miss K. Way's calculations, this should be the case. Assuming that the properties of 40 are the same as those of 49—which is perhaps a somewhat optimistic assumption—the number of fissionable atoms formed per 49 consumed is $\eta + (\alpha - 1)/\alpha = 2.05 + 0.33 = 2.38$, *i.e.*, just the same as for 23. In spite of this, under the above assumption, 49 would be a somewhat more suitable material for the breeder than 23 because of its larger total cross section which would reduce the amounts needed and the concentrations necessary by the ratio of the cross sections, *i.e.* 630/1050 = 0.6. Furthermore, a loss of p percent of all the neutrons would reduce the efficiency in the case of Pu only to $\eta(1-p) - 1 - 1/\alpha = 0.35 - 2.05p$ while the same loss would give, in the case of 23, an efficiency of $\eta(1-p) - 2 = 0.37 - 2.37p$. This calculation assumes that 24 is not slow neutron fissionable. The possibility of using 49 is presented also because there may be a certain experimental error in the η of 23, as determined by Anderson and May.

B. Brief Description of the Arrangements Considered.—The general arrangement which at present appears most attractive consists of a relatively small pile proper which contains heavy water as

moderator and all the 23 that is added to the system. The heavy water and the 23 are circulated to the outside of the system, cooled, stripped of the fission and decomposition gases and freed of the fission products outside. The chain reacting part is surrounded by the reflector. This also consists essentially of heavy water and contains most or all of the thorium. To give an idea we include a sketch at this point, although the detailed calculations will follow later on. The chain reacting unit has a volume of about 400 liters and contains of the order of 3 kg of 23. The thickness of the reflector is about 50 cm and contains about equal weights of thorium and heavy water. The pile is to be operated at 100,000 to 200,000 kW.

The first possibility (Fig. 1) envisages the uranium dissolved in the heavy water in the form of a salt, such as the nitrate, sulphate or fluoride. This solution is pumped into the pile through the neck which is on the bottom side and leaves the pile through the neck on the top side. The circulation rate is about 500 liters per second. The solution enters a heat exchanger and a degasser after it left the pile, whence it re-enters the pile again. The pile is surrounded by the thorium which is in the form of a slurry containing about one gram of ThO_2 per cm^3 of heavy water. If the wall of the tank separating the uranium solution from the thorium slurry is of a low absorbing material such as Be, Pb, Bi, or perhaps Cb, the neutron absorption of the tank may be sufficiently low. If, however, the tank has to be made out of a more absorbing material it may be necessary to back this up with a higher concentration of thorium, possibly in the form of small spheres ("beebees"), in order to reduce the thermal neutron intensity in that region.

The second arrangement shown (Fig. 2) contains the uranium dissolved in Bi, about 1 gm to 25 cm^3. This bismuth is circulated in suitable tubes and is cooled and freed of fission gases outside the pile. The shape of the pile is so chosen that the neutron escape at the ends shall be about 1% of the total neutron escape because this part of the neutrons will be lost. It may be possible to achieve the same purpose by another shape of the pile more similar to that in Fig. 1. This would involve, however, a more complicated inner structure of the pile, The tubes containing the bismuth are surrounded by heavy water which also has to be recirculated, degassed and cooled outside the pile.

The interest in UF_6 as a pile material has been raised on this project by H. Brown and we are indebted to him for a discussion on both this and other problems. UF_6 could be used instead of metallic U in the pile of Fig. 2. The Bi would be replaced in this case by a fluorocarbon.

The first arrangement is much simpler than the second one and has a very much greater production rate, but the power which it furnishes cannot be well utilized because the temperature at which the pile can be operated is quite restricted. The greatest unsolved problem is that of the degassing of the heavy water without increasing the holdup outside the pile unduly. The second system is much more complicated and involves many serious problems among which we believe that a suitable choice of the pipes and the separation of the fission products from the Bi solution is most serious. Its production rate is much lower than that of the first pile but it furnishes the energy at a sufficiently high temperature for purposes of power production. If UF_6 is used in a fluorocarbon instead of U in Bi, the complications are much decreased but so would be the temperature of operation. The problems of chemical separation would be greatly alleviated but one would have to cope with the effects of radiation on the fluorocarbons. According

to arguments presented by Burton and Franck, one may expect a somewhat more favorable situation in case of unsaturated ringlike structures, if these are sufficiently stable chemically, than in case of saturated fluorocarbons.

C. Reasons for the Arrangements Discussed.—The piles as described above assume pumping of the uranium solution by some suitable pump. Instead of this the solution can be pushed alternately into and out of the pile and cooled and degassed while it is outside (pulsating pile).

The most definite reason for using the 23 in solution is the need for eliminating the Xe-135. If we assume that altogether 5 kg of 23 are in the system and that it operates at 100,000 kW, 100 gm of 23 undergoes fission every day, the amount of Xe produced per day is 3.3 gm. If this were permitted to remain in the system as long as one day its cross section would be five times greater than that of all 23 present, In order to reduce this cross section to 1% of the cross section of the 23, one has to eliminate the Xe formed every 5 minutes. This is possible only if the Xe can be flushed out of the system at frequent intervals and eliminates the possibility of using the 23 in a solid form. If the Xe is flushed out at very frequent intervals one gains an additional substantial elimination of other fission products which might otherwise poison the pile.

The above conclusion that the 23 must be used in a non-solid form is not absolute. The maximum loss of efficiency if all the Xe-135 is left in the pile is 5.8% and although the permissible loss must be well below 37% a loss of 5.8% could be tolerated if all the other losses can be kept at a minimum. However, circulating the 23 has many other advantages; particularly from the point of view of heat transfer and it is not evident that using it in a solid form will facilitate the elimination of the other losses. We feel, therefore, that the conclusion to use the 23 in a non-solid form is a valid one. The only other possibility to eliminate the Xe-135 loss which we realized is to operate in such a way that practically all neutrons be absorbed at higher energies (by resonance absorption) before they reach such energies at which the Xe-135 cross section is large.

The Xe-135 could be removed from the 23 inside the pile by letting a gas bubble through the liquid which contains the 23. This might be a practical possibility if the 23 is dissolved in Bi. We felt, however, that such an arrangement would introduce unnecessary complications and since it has great advantages to remove the 23 from the pile periodically for cooling and for separating the other fission products, we decided against it. This fixed one of the most important properties of the piles which were described above, that of circulating the 23 to the outside of the pile. Such a circulation of course increases the amount of 23 necessary for the pile but greatly facilitates the energy transfer from the 23 to the outside.

It will be seen that considerable difficulty is caused by the need of inserting a tank between pile proper and reflector because the tank is likely to absorb neutrons. It would be advantageous, therefore to place the thorium, or at least most of it, to the inside of the pile. This would result in the use of a synthetic "natural uranium" which would give a pile with a very small multiplication factor, an arrangement which would greatly help in decreasing the neutron losses. There is no question that there is great merit in this point of view. If we reduce the multipli-

cation constant (which is nearly 2.37 in the pile as described above) to 1.05, only 5% of all the neutrons escape from the pile proper and only these have a chance of being absorbed by the tank. However, the diameter of the pile will be increased in this way to about 3 m and its volume to 14 m3. The amount of 23 required for the pile proper would be of the order of 70 kg under these conditions. In order to keep the amount of 23 needed for the pile at a relatively low levels we have decided therefore to separate the chain reacting part from the reflector where the production of the 23 takes place. It might be advisable to add some thorium to the pile proper but this should be kept at such a level that the multiplication constant does not decrease below about 1.5. If this is done the absorption by the thorium within the pile proper will be about 60% of the absorption of the 23 and its amount in the pile will be about 60 times the amount of the 23. Such an arrangement has the advantage that the number of neutrons which have to leave the pile is reduced by a factor of almost 3 and the losses in the tank and outside will be diminished by the same factor. If the pile proper contains the 23 in a solution in heavy water we have decided against this possibility because the thorium could be present only in the form of a slurry which would aggravate the pumping problems and the problems of chemical separation.

The next question is the question of the moderator. In this respect we believe that the choice of heavy water is rather unique. If we want the number of neutrons absorbed in the moderator to be only 1% of the neutrons absorbed by the 23, the volume g of the different moderators associated with 1 gm of 23 is given in the following table:

Table 2:

	σ_a/molecule	Density	Volume associated with 1 gm to give loss of 1% (g)	Heat capacity of this volume in joule/°C (Cs)	Age
C	0.0045	1.6	44.5 cm^3	65	320 cm^2
D_2O	0.004	1.1	120 cm^3	500	115 cm^2
D_2O + 1/2 H_2O	0.007	1.1	70 cm^3	290	110 cm^2
H_2O	0.6	1	0.8 cm^3	3.3	33 cm^2
Be	0.01	1.8	13.3 cm^3	42.5	200 cm^2
Bi	0.017	9.8	33 cm^3	48	—
$(CF_2)_n$	0.023n	1.8	32 cm^3	~70	

Among these C is undesirable because the radius of the pile would amount to about 60 cm, assuming a multiplication constant of 2.37 and a migration area corresponding to the age alone (*i.e.* assuming zero diffusion length for the thermals). Such a pile would contain far too much 23. Water is undesirable because the size of the pile would become too small so that one would run into heat extraction difficulties. If the 23 is in the pile as short a time as 1/10 sec and if one wants a power output of 100,000 kW for 5 kg 23, the temperature increase is the water would amount to 20,000 x 1/10/33 = 600°C. Be in itself is not much more suitable than graphite and Bi cannot be seriously considered as a moderator. This leaves D_2O as a rather unique choice. On the other hand, it is possible for the pile to contain substantial amounts of Bi and Be, in addition to the D_2O which has to do the bulk of the moderation.

Heavy water as a moderator gives considerable leeway for the concentration in which 23 can be used. The importance of this is the following: If we use a high concentration of 23 only a relatively small fraction of all the neutrons will reach thermal energies and many of them will be absorbed at resonance above thermal energies. There is not much reason to believe that the η is absolutely constant even in the very small energy region which extends from ordinary thermal energies to about 1 volt and if it should turn out that η increases with increasing energy it may constitute a considerable advantage to go to high concentrations and high resonance absorption. On the other hand, if it should turn out that η decreases with increasing energy of the neutrons, it would be best to keep the concentration of 23 in heavy water as low as possible. This question will be taken up again in section D.

It is well known that if the uranium is dissolved in water the fission causes a very large amount of disintegration. We estimate from data of Burton and Allen (Γ = 0.25) that such a pile operating at 100,000 kW would give 220 liters NTP hydrogen oxygen mixture per second. While the gases have the desirable effect that they carry with themselves the Xe and some other fission products when they leave the liquid, the disengagement of such amounts of gas causes considerable trouble even if it is done at relatively high pressures. There is considerable advantage therefore to have the 23 separated from the heavy water which can be done most simply by dissolving it in Bi and keeping the Bi separated from the heavy water by tubes of a non-absorbing material or thorium. This leads us to the two systems which we have considered most seriously and which will be described more closely in the next section.

There is one other remark that ought to be made in this connection. At the concentrations or 23 which reduce the loss due to absorption by the moderator to the order of 1% a considerable number of neutrons will be absorbed before they reach thermal energies. If both the moderator and the 23 obeyed the 1/v law this would not change the ratio of the neutrons absorbed by the moderator. There is considerable indication, however, that the absorption of the 23 at resonance energies is considerably greater than would correspond to the tail of the 1/v absorption extrapolated from thermal energies. Mr. Dancoff estimates on the basis of experiments carried out in the Argonne Laboratory that the $\int \sigma_a dE/E$ is at least twice greater than it would be if 23 followed the 1/v law. As a result, the fraction of neutrons which are absorbed by the moderator will be about half as great for the neutrons absorbed at resonance than for the neutrons absorbed at thermal energies. Another effect which this resonance absorption has comes in in the calculation of the Laplacian which is increased by this effect. Since the ratio of the actual $\int \sigma dE/E$ to the value which is obtained by extrapolating the 1/v law is not known the calculations will be carried out under three different assumptions, viz, that that ratio is α = 1, 2, 4.

D. Critical Size.—The critical sizes for pure fissionable material have been calculated by Christy and Wheeler (CP-499), by Weinberg, and more recently these calculations have been extended by Nordheim.

It turns out that the two group theory and the three group theory both give results which are substantially below those given by Fermi's theory. The results of the three group theory are just about midway between those of the two group theory and the Fermi theory. Plass calculated

the corrections to the Fermi theory and in the case of heavy water they turned out to be quite negligible, as was already indicated by Weinberg's work.

Our calculation includes the effect of the resonance absorption for which the $\int \sigma dE/E$ was estimated in the following way. The value of σ_a was taken to be $(98\alpha)/\sqrt{E}$ which gives for $\alpha = 1$ the correct value for thermal energy. However, three values of α were used, viz., $\alpha = 1, 2, 4$, as explained before, The integration was extended from $2kT$ to ∞ giving 900α. Evidently these assumptions are quite arbitrary but should give the right order of magnitude. The average energy of the neutrons in the pile will no doubt be considerably higher than 0.05 eV, but the slowing down just above the thermal region will be considerably slower than the formula for the resonance absorption implies. These two effects give corrections of opposite sign, The following table gives the probability for escaping resonance for g = 100, 200, 300, 400 cm^3 heavy water per gram 23.

Table 3: Number of Neutrons Reaching Thermal Energy

g	α = 1	α = 2	α = 4
100	0.875	0.769	0.592
200	0.936	0.875	0.769
300	0.957	0.916	0.840
400	0.968	0.936	0.875

The Laplacians L were then calculated by Mr. Wilkins on the basis of the formula developed by Messrs. Friedman and Weinberg

$$k_s \left(\frac{\chi_{t^2}}{\chi_{t^2} - L} \right) + k_f = \exp(-L\tau)$$

and his calculation will be given in more detail elsewhere. In this, $k_s + k_f = k$ is the multiplication constant; k_s is the number of neutrons produced per slow neutron absorbed, multiplied with the fraction p of neutrons which are thermal when absorbed; k_f is the number of neutrons produced per fast neutron absorbed, multiplied with the probability $1 - p$ of resonance absorption. In our case $k_s = pf\eta_s \approx p\eta$; $k_f = (1 - p)\eta_f$.The results with Fermi's theory are given in

Table 4:

	10^3 L cm^2			R in cm			Volume in liters			23 in kg.		
g	α = 1	2	4	1	2	4	1	2	4	1	2	4
100	5.63	5.83	6.19	41.8	41.1	39.9	162	152	136	1.62	1.52	1.36
200	4.53	4.66	4.95	46.6	45.9	44.6	241	228	216	1.20	1.14	1.08
300	3.79	3.90	4.12	50.9	50.3	48.9	332	317	291	1.11	1.06	0.97
400	3.30	3.39	3.57	54.8	54.1	52.6	430	411	372	1.07	1.03	0.93

It was assumed in the calculation that the age τ of fission neutrons in heavy water is 115 cm^2, the diffusion mean free path 2.4 cm. This gave 50, 100, 150 and 200 cm^2 for the square of the thermal diffusion length $1/\chi_{t^2}$ for the four different g. The radii R of a bare critical sphere are also given in Table III. According to calculations of Mrs. Monk and Mr. Friedman, which also will be reported elsewhere, it seems reasonable to subtract 8 cm from the radius in case of a ThO_2 slurry reflector. This gives the amounts of 23 necessary for the pile which are given in Table III in kg.

The advantage of using a high concentration lies in the smaller losses of neutrons to the moderator, etc. On the other hand, a low concentration of 23 would give increased bulk to the system and thus facilitates a higher production rate. If α should be large there is an increased advantage in using high concentrations because the neutron losses to the moderator are further reduced by the resonance absorption. It is seen from Table III that the holdup of 23 within the pile is quite small. There is of course a holdup outside the pile.

If it should be necessary to use the cylindrical arrangement given in Fig. 2, the height of the cylinder will have to be chosen in such a way that the escape toward the two ends of the cylinder be about 1%. This gives the following equation for the height H of the cylinder in terms of its radius R

$$\frac{\pi^2}{H^2} = \frac{1}{100}\left(\frac{2.405}{R}\right)^2$$

Calculating the volume from this equation it appeari that it is 4.3 times greater than the volume of the sphere considered in Table III. This would necessitate a four times greater holdup in 23 which can be reduced, however, by reducing the diameter of the tubes containing the 23 solution toward the two ends of the pile.

The lattice spacing is not a critical quantity in the second arrangement. Of course, the total amounts of heavy water, beryllium, and solvent must be kept in such limits as can be obtained from Table I. In addition, one will not want the neutron density in the moderator to exceed substantially the neutron density in the solution. If $\chi r_0 < 1$, the neutron density at the tube will be less than 10% higher than the average neutron density in the tube. Since $\chi = 0.2$ for both the Bi and the fluorocarbon solution of Table I, this only gives $r_0 < 5$ cm. The ratio of the neutron density difference between surface of cell and surface of tube, divided by the density at the tube is

$$\frac{N\sigma_{ao} r_0^2}{2D}\left(\frac{r_s^2}{r_s^2 - r_0^2}\ln\frac{r_s}{r_0} - \frac{1}{2}\right)$$

In this $N\sigma_{ao} = 0.048$/cm for the solutions of Table I, D = 0.8 cm is one third of the diffusion mean free path, r_s is the cell radius. The above ratio will be under the conditions of Table I, smaller than 0.1 if r_0 < 3.5 cm.

E. Holdup Outside the Pile.—The holdup outside the pile depends to a large extent on the diameter of the tubes in which the solution passes through the heat exchanger, the temperature difference between coolant and the temperature of the 23 solution, holdup in the pumps and, particularly in case of the arrangement of Fig. 1, on the time necessary for degassing. No calculations are available on these points at present but one can estimate that the solution will spend about 2 sec outside the pile before it re-enters it. Under this assumption the power output depends solely on the holdup and the temperature difference by which the solution is cooled down. It is given by

$$P = \frac{C_s T}{t} \frac{\text{kW}}{\text{kgr holdup}}$$

In this C_s is the heat capacity of the solvent per gram of 23 dissolved in it (cf. Table I), t is the time of holdup outside the pile, and T the temperature difference between the solution as it leaves and re-enters the pile. In case of pile of Fig. 1, t = 2 sec, T = 50°C, g = 100, this gives P = 10,000 kW/kg, g = 200, P = 20,000 kW/kg.

One may consider flashing the liquid in order to achieve some cooling by evaporation as well as elimination of the fission gases. Messrs. Newel and Ginns proposed in some other connection to let the solution flow down on the inside of a cooled tube in a thin sheet and one could maintain sufficiently low pressure inside the tubes to permit evaporation and release of the bulk of the decomposition gases. These would carry the Xe and some other fission products with themselves and also a considerable amount of heat in the form of heat of vaporization. The advantage of this arrangement is that it unites the elimination of the decomposition gases with some cooling and also that the pressure drop in such a system is quite small. Considerations have not progressed sufficiently to know how large the saving in the holdup would be if this arrangement is used.

A possibility which might be applied to the arrangement of Fig. 2 is that of pulsating it. This avoids the holdup in the pump and also the need of pumping the radioactive solution. Figures for the theoretical efficiency of this cooling are given in Table IV in kW per gm holdup outside the pile. It was assumed that the temperature difference between coolant and 23 solution of the pile is 50° and that the temperature difference between the coolant and the tube of the heat exchanger is made negligible by vigorous circulation of the coolant and finning of the heat exchanger tubes. The pressure difference between the active pile and the end of the tubes alternates between 0.6 and -0.6 atmospheres in such a way as to give a velocity of 700 cm/sec in the tubes. The holdup in the degasser is not taken into account in the figures in Table IV.

The figures given for the power assume that the pressure adjustment at the end of the tubes can be carried out instantaneously. The bracketed figures assume an adjustment time of 0.1 sec. It was further assumed that the tubes can be entirely emptied on the "in" cycle. Assuming that the meniscus has to stay at least 10 cm away from the pile tank further reduces the power holdup by 20% to 5% for tube lengths of 50 cm to 200 cm. The advantage of longer tubes is, obviously, that they permit a greater total production, although with a somewhat decreasing efficiency. The advantage of wide tubes is the same and a greater mechanical simplicity.

Table 5:

Radius of tubes (cm)	Length of tubes (cm)	Length of cycle (sec)	kW/gr g = 100	kW/gr g = 200
1	100	0.45	37.5 (26)	75 (52)
1	200	0.90	31 (25)	62 (50)
0.5	50	0.23	82 (44)	164 (88)
0.5	100	0.45	65.5 (44)	131 (88)
0.5	200	0.90	45 (37)	90 (74)

F. Estimate of the Losses—It will be seen that, on the whole, the losses from the different sources enumerated under A become smaller as the production rate of the pile is diminished. It appears quite natural that the losses in a pile with a high power output are greater than if all attention can be given to a greater efficiency. We proceed with the discussion of the eleven separate points given under A.

(1) The number of delayed neutrons is about 0.8% in 25, Although its amount is not known for 23, Anderson found indications that it is greater for this element. As pointed out under section A, a loss of 1% of neutrons decreases the efficiency by 2.37%. Since all the delayed periods, except perhaps the first one, are long compared with the time of uninterrupted sojourn of the 23 within the pile, the number of delayed neutrons emitted outside the pile will be equal to the holdup outside the pile divided by the total amount of 23 present. From this point of view it is indeed advantageous to have a large pile and a very small holdup outside.

The flash cooling system may form an exception under this rule be cause it may be possible to arrange it in such a way that most of the neutrons still would be utilized. Similarly it may be possible to achieve this at least partially in the pulsating system. In spite of this there will be a loss of efficiency from delayed neutrons in every system in which the uranium is circulated at the outside. This constitutes a rather serious drawback of these systems since the loss in efficiency from this source is not very much smaller than that caused by the Xe-135 would be.

(2) The absorption by the moderator has been discussed in detail before. It will amount to about 1% for the pile of Fig. 1 and one will be quite lucky if it is as little as 2% in the second case. However, in the first case the absorption by the anion also has to be taken into consideration. If the uranium is used in the form of a nitrate the nitrogen of the nitrate alone will give a loss of 1/2%. In addition to this it appears likely that further nitric acid has to be added to the system in order to keep the uranium in solution in spite of the presence of the hydrogen peroxide. The situation would be very much better if N-15 were available for use both in the nitrate and the nitric acid.

The reason that we gave the nitrate precedence over other salts is that we expect it to reduce the corrosion of the walls and also because it does not give products under radiation which

may gum the system. The sulfate which has negligible absorption may precipitate sulphur although it is not clear that this cannot be avoided by the addition of peroxide. The parasitic absorption of the fluoride, the use of which was proposed by Allen, is also negligible but there is a strong opinion (Hiskey) that it will be difficult to avoid precipitation of the peroxide at such concentrations of the hydrofluoric acid which are tolerable from the point of view of corrosion. If the uranium is used in the form of a solution in Bi or fluorocarbons it may be necessary to eliminate the products of nuclear reactions of these substances along with the fission products.

(3) The estimate of the poison by the fission products is probably the most difficult at present. According to Miss Way only the Sm is a serious poison among the stable fission products. Inasmuch as the yield of this is probably 1.6% its elimination is very important and may demand an average interval of one day between purifications. In order that this be permissible, it is necessary that the purification be carried out with a loss of about 0.1% of the 23.

The above applies to the stable fission products. There may be, however, numerous radioactive fission products (in addition to the Xe-135) which have large cross sections. Their effect has been estimated by Miss Way on the basis of a statistics of the cross sections and she finds that those with an odd number of neutrons are most dangerous. All elements may give substantial contributions, the lifetime of which is an hour or more Only those with a lifetime of more than a day have been surveyed to date and Miss Way estimates that they may cause a loss in efficiency of 3.5% if the purification is carried out once a day. If this is done it is unlikely that the corrosion products will contribute much to the poisoning.

Probably the most efficient way of eliminating the fission products is by the solvent extraction method. We are much indebted to Mr. Tepe for discussing this system with us. The solvent extraction method would permit a continuous operation and cause a holdup of only about one hour in the column. As a result it would appear that such a purification increases the amount of 23 required by only about 4%. However, it is likely that it will be impossible to use the solution of uranium in heavy water directly in the column because the deuterium will exchange with the hydrogen of the solvent. For this reason it will be necessary to separate first the uranium salt from the heavy water which may be a more time consuming operation than the operation of the column itself and consequently increase the holdup more than the fission product separation itself does.

If the uranium is used in a solution in Bi the problem of chemical separation is even graver. Although many methods were discussed no really promising one turned up so far. One must remember that after the chemical separation has been carried out the uranium must presumably be reduced again to the metallic form. It is unlikely that this operation can be carried out with as small a loss as would be necessary for the successful operation of the breeder. This is the most serious drawback of the Bi system that has turned up yet. On the other hand; if the uranium is dissolved as a hexafluoride in fluorocarbons the problem of separation may be much smaller. As pointed out by Brown and Anderson, distillation may lead to the goal and the extraction column also could be used because reconversion to the fluoride can probably be handled with relatively great efficiency.

The Xe-135 occupies a particular position. Let us first consider the case in which the uranium is dissolved in heavy water. In this case there is a considerable amount of water decomposition and the hydrogen and oxygen generated must be eliminated from the system every second. Even so, the equilibrium pressure of H_2 and O_2 will amount to a dozen atmospheres. During the degassing the hydrogen and oxygen will leave the water either in form of bubbles or through the surface. In both cases the fraction of Xe eliminated will be just about as great as the fraction of hydrogen and oxygen eliminated because the solubility of Xe is only insignificantly higher than the solubility of hydrogen and oxygen and because the diffusion of Xe to the surfaces will not be much slower than the diffusion of the oxygen. As a result, the amount of Xe will be reduced by a factor of the order of 1/2 every second and the amount present will be that generated during a few seconds only. Since it is necessary to eliminate the Xe only about every couple of minutes, it is evident that the need of elimination of the hydrogen and oxygen will cause in this system an about 100 times better elimination of the Xe than is necessary. Even if it should prove feasible to reduce the gas evolution to 1/10 or even 1/30 by introducing some suitable catalyzer into the system which recombines most of the hydrogen and oxygen in the water the elimination of the Xe would be quite satisfactory.

For a quantitative assessment of the problem let us assume that the mean life of the 23 in the pile is $\bar{t}$. This is the amount of 23 present, divided by the consumption rate and is for 100,000 kW power rate of the order of 6 kg/100 gm/day = 60 days. Assuming a fission yield of 5% for the Xe-135, the production rate of Xe-135 will be 0.05 $N/\bar{t}$ where N is the number of 23 atoms. The total number of Xe-135 atoms in the system will be 0.05 $Nt_e/\bar{t}$ where t_e is the average time after which the Xe is eliminated. The poisoning due to this amount is $Nt_e\sigma_{Xe}/\bar{t}N\sigma_u$ and this will be less than 1% if

$$t_e < 0.2\frac{\sigma_u}{\sigma_{Xe}}\bar{t} = 4 \times 10^{-5}\bar{t}$$

For $\bar{t}$ = 60 days this gives $t_e \approx 3$ min.

The situation is somewhat different if the uranium is dissolved in bismuth or a fluorocarbon. In the former case at least it will be necessary to sweep out the Xe by bubbling some gas through the bismuth periodically. This has to be done, as pointed out before, every five minutes at least and will cause some additional holdup, the magnitude of which will depend on the size of the bubbles sweeping through the liquid and the area over which the solution is spread out during this operation. It does not appear, however, that the increase in holdup necessitated by this operation will go beyond a few percent.

(4) The absorption by the tank is principally determined by the material of the tank. If this absorption is low it will be quite possible to have the slurry of thorium which is outside the tank in immediate contact with the tank, Mr. Friedman and Mrs. Monk have calculated this effect. In the region of interest, their results can be approximated by the formula

$$\frac{\sigma_{\text{at}}\ell_t}{\sigma_{\text{as}}\sqrt{\tau}}$$

for the fraction of neutrons entering the reflector which are absorbed by the tank. In this, σ_{at} is the absorption cross section of the tank per cm^3, ℓ_t the thickness of the tank, σ_{as} the absorption cross section of the slurry per cm^3, r= 115 cm^2 is the "age" in the reflectors. We assume a slurry which contains 1 gr Th per cm^3 which gives σ_{as} = 0.016.

The loss of efficiency is obtained by multiplying the above expression by 1.37. It is only 1% for 1.5 mm thick Al tank (σ_{at} = 0.014) and correspondingly less for a tank out of lead or beryllium or for some backing of the aluminum made out of beryllium.

If the tank has to be made out of a strongly absorbing material, it will be necessary to back it up with a layer of thorium in a more highly concentrated form (cf. Fig. 3). Beebees of metallic thorium which are cooled by heavy water circulated through them may be a suitable form. This arrangement has two effects: first the thorium reduces the density of the thermal neutrons in the neighborhood of the tank to such a level that the ratio of the thermal neutrons which are absorbed by the tank to the number of thermal neutrons which leave the pile toward the reflector is given by the formula

$$\sqrt{\frac{3\sigma_b}{\sigma_{ab}}}\sigma_{at}\ell_t$$

In this σ_b is the scattering cross section in the backing per cm^3 of the backing, σ_{ab} is the absorption cross section of it, again per cm^3. One calculates that if the backing consists of 80% metallic Th and 20 volume percent of heavy water (σ_{ab} = 0.13, σ_b = 0.33) the absorption of thermal neutrons by a 1.2 mm steel tank (σ_{at} = 0.22) would give a loss in efficiency of about 1.5% if one assumes that 1/2 of all neutrons escape as thermal. A reinforcing of the tank by a few mm aluminum would hardly add anything to this. Behind the backing there must be of course further heavy water containing thorium to slow down and absorb the neutrons which leave the tank at higher energies. It is easy to see, along the same lines as used in the argument by Friedman and Monk, that the fraction

$$\frac{\chi_b\sigma_{at}\ell_t\sqrt{\tau}}{\chi_b\sigma_{as}\mathrm{Ch}\chi_b\ell_b + \chi_s\sigma_{ab}\mathrm{Sh}\chi_b\ell_b}$$

of these neutrons will be absorbed by the tank. In this formula, as before, s and b refer to slurry and backing, σ_a, χ, and ℓ are absorption cross section per cm^3, reciprocal diffusion length and thickness of the corresponding region. For the above backing, containing 80 volume percent Th, 20 D_2O, χ_b ~0.35, and the above slurry, containing 1 gr/cm^3 Th χ_s = 0.14, As a result, if the backing has a thickness of about 5 or 6 cm the absorption of these neutrons is also quite small (< 1/2%). Evidently the introduction of the backing will somewhat reduce the efficiency of the reflector. However, the 8 cm thickness which was used in the previous calculations already takes this effect into consideration. Another point that might be mentioned is that the resonance absorption given in Table II also will serve to reduce the losses due to the tank by the factors given in that Table.

(5) The shape of the tank as given in Fig. 1 or 2 is already designed to reduce the escape from the tank in the directions in which there is no reflector and it is sufficient to repeat that the

extremely elongated shape given in Fig. 2 may not be necessary if the tubes toward the two ends of the tank are made thinner than at the center, thus reducing the multiplication constant in that region. This will, of course, cause a slight increase in the diameter of the tank but will still very substantially reduce the amount of uranium in the pile,

(6) The effect of the absorption by the 24 can be considered under the assumption that the 24 itself is not fissionable. In this case every atom 24 will eventually be transformed into 25 so that in equilibrium there appears to be a loss of 12% in efficiency due to the absorption by the 24 because in equilibrium, 0.12 as many neutrons are absorbed by 24 than by the 23. Of course if the cross section of the 24 is very small this equilibrium will be reached only after a very long time and until this happens the loss in efficiency is correspondingly smaller. However, even when equilibrium has been reached the loss in efficiency will be very much smaller than the 12% quoted because the absorption leads to a fissionable element 25 and the loss in efficiency is caused only by the fact that the 25 has a smaller η than the 23. When the absorption of 25 has come to an equilibrium one can consider the total effect in the following way. A 25 is formed by the absorption of two neutrons from the 24 and gives η_{25} new neutrons. If we denote, as in Section A, the fraction of neutrons which survive all losses by 1 - p, the loss due to the absorption of 24 will be given at equilibrium by (2)(0.12) - (0.12)η_{25}(1 - p) = (0.25p) - 0.01. One sees that this absorption will not cause a considerable decrease in efficiency. Strictly speaking, one should further consider the formation of 26 from the 25, but this evidently will not cause any major effect.

(7) The absorption by the Pa is a more serious effect. This element is, according to Miss Way, probably not fissionable and will therefore cause a loss in two ways. First of all, by the neutrons which it absorbs and second by forming an element of a relatively short half-life which decays into 24 instead of the fissionable 23. For this reason it will be necessay to keep the absorption by the Pa at a minimum. This can be done by extracting the Pa at sufficiently frequent intervals so as to reduce the absorption of neutrons by this element to about 1/2% of the absorption by the thorium. This will be relatively easy if the absorption cross section of Pa is of the same order as the absorption of the thorium and if all the thorium can be used in the form of a slurry, since for a power output of 100,000 kW only about 120 gms of Pa are formed per day and since the mean life of Pa is 40 days, there will be, in steady state, 4.8 kg Pa in the reflector. Since the total amount of Th in the reflector is more than a ton, it will be sufficient to extract the Pa only n times a month if its cross section is n times that of the thorium. However, if it is necessary to use part of the thorium in form of a backing of the tank, a considerable fraction (about 25%) of all of the neutrons will be absorbed by the first few cm of this backing. As a result, the Pa accumulates in this part of the thorium much more rapidly than in the bulk of it and the purification would have to be undertaken more frequently. This is especially disconcerting because the removal and replacement of this backing is a somewhat awkward problem.

(8) Relatively frequent purification of the thorium seems also indicated by the accumulation of fission and corrosion products. The frequency of the need of this purification can be estimated in the following way: The absorption cross section of thorium is just about 100

times smaller than the absorption cross section of the 23. Therefore, if we divide the amount of thorium in the reflector by 100 times the amount of 23 in the pile, we obtain the number by which the cross section of the thorium in the reflector is larger than the cross section of 23 in the pile. This number is at least 4 to 5. On the other hand, the amount of fission in the reflector will be less than 1% of the amount of fission in the pile if all the thorium is distributed uniformly in the reflector. The relative poisoning of the reflector by the fission products will be about 400 times smaller than the poisoning of the pile and a correspondingly less frequent purification would be quite sufficient for the elimination of the fission products. It is seen from this that the need for purifying the thorium of the reflector will arise mainly from the absorption by the Pa, the absorption by the corrosion products, if any, and to reduce the holdup of 23 within the reflector. This last factor will demand a purification of at least once a month because after this time there will be about 3 kg of 23 in the reflector. Of course a more frequent purification maybe demanded either because of the absorption of the Pa or the absorption by the corrosion products but we shall see that this does not seem to be the case. The above discussion assumes that the thorium is uniformly distributed within the reflector but the situation is not very different if a backing of the tank is needed.

(9) As mentioned before, the absorption cross section of thorium is just about 100 times smaller than the absorption cross section of 23. Therefore, if we want to reduce the losses due to the moderator in the reflector to 1% of all the neutrons absorbed in the reflector—this will give a loss of 1.37% in efficiency—the volumes associated with 1 gm of thorium are 100 times smaller than the figures given in the fourth column of Table I. It appears from this that only D_2O can serve as a carrier of the thorium in the reflector. Using the thorium in form of a slurry has the further advantage that a frequent mixing of the whole amount of thorium can be achieved and it will not be necessary to remove parts of the thorium at more frequent intervals.

(10) There are three causes for losses of neutrons from the reflector: The first one is the escape of neutrons toward the ends of the pile in case the arrangement of Fig. 2 is used. This, however, will be a very small quantity if the loss from the pile in the same directions is also very small. The escape of neutrons perpendicular to the main surface of the pile is governed by the tail of the slowing down density of neutrons. If one could adopt for this tail the Gaussian expression it would be sufficient to use such a thickness ℓ_s that $\ell_s^2/4\tau = 4$ which would give $\ell_s = 4\sqrt{\tau} = 45$ cm. Actually it may be safer to surround the reflector by a second reflector which may be out of graphite. In this case one can reduce the thickness of the reflector which contains the thorium to about 38 cm.

This figure is arrived at in the following way. The number of neutrons absorbed in the reflector divided by the number of neutrons entering the graphite reflector is approximately

$$\frac{4}{\sqrt{3}}\sqrt{\frac{\sigma_{\mathrm{ag}}}{\sigma_{\mathrm{g}}}}$$

In this, $\sigma_{\mathrm{ag}}/\sigma_{\mathrm{g}}$ is the ratio of absorption and scattering cross section in C. This formula as-

sumes that all the neutrons entering the graphite are thermal which is a slightly pessimistic assumption. The fraction of neutrons entering the graphite reflector is given by

$$\frac{3\sqrt{\tau}\sigma'_{\rm s}}{2\sqrt{\pi}}\exp(-\ell_{s^2}/4\tau) - \frac{3\ell_{\rm s}\sigma'_{\rm s}}{2\sqrt{\pi}}\int_{\ell_s/2\sqrt{\tau}}^{\infty}\exp(-y^2dy) + \frac{\exp(-\ell_{s^2}/4\tau)}{4\sqrt{\pi}\tau\sigma_{\rm as}}$$

$\sigma'_{\rm s} \approx 0.4$/cm is an average fast neutron cross section of the slurry per cm^3. The first two terms refer to the fast neutrons entering the graphite, the last term to the slow neutrons entering it. The length, 38 cm, is so chosen as to reduce the total number of neutrons escaping from the system to less than 0.5% of all neutrons entering the thorium reflector. The above calculation should be somewhat improved to take into account the neutrons generated by fission in thorium. However, the number of these neutrons is quite small, less than 1% of all the neutrons, and the probability of their escape remains small even if they are found somewhat to the inside of the thorium reflector.

(11) The losses in chemical separation should really be considered in conjunction with the poisoning problem. Evidently the smaller the losses the chemical separation involves, the more frequently it is worth while to carry out such a separation. The figures given under (3) were not calculated but estimated on the assumption that the loss of 23 during a chemical cycle amounts to 0.1%

The total loss in efficiency therefore can be estimated as follows:

Table 6:

	System I (no power)	System II (some power)
1. delayed neutrons	2	2
2. parasitic absorption in pile	1.5	2.5
3 and 11. fission and corrosion products, chemical separation	3.5	5
4. tank	1	1
5. parasitic escape	0.5	1
6. 24 absorption	1.5	1.5
7 and 8. 13 absorption	1.5	1.5
9. parasitic absorption in reflector	1	1
10. parasitic escape	1	1.5
	13.5%	17%

It is unnecessary to remark that these figures give, in the best case, an orientation and that the chances are that they are optimistic. The losses which were enumerated are probably all present and it is unlikely that they are greatly overestimated in the systems considered. However, there may be other losses which we failed to recognize. In spite of this, it appears, at present, that

the efficiency losses can well be held below the 37% figure which is the margin if Anderson and May's η for 23 proves accurate.