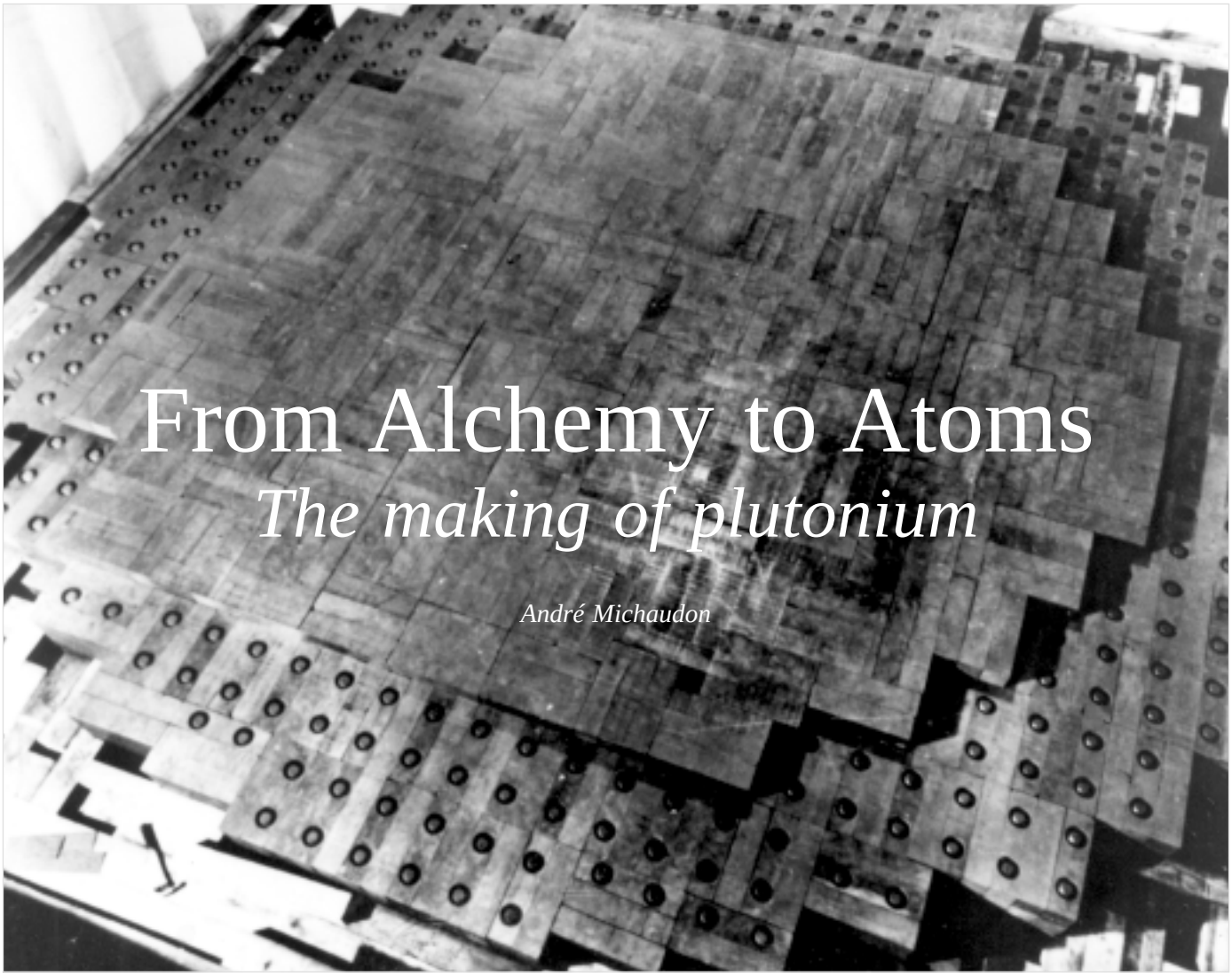


From Alchemy to Atoms

The making of plutonium

André Michaudon

To this day, plutonium—the famous isotope plutonium-239 in particular—continues to play a crucial role in nuclear weapons and peaceful nuclear energy. Having atomic number $Z = 94$, plutonium is the next element after neptunium in the periodic table and is two elements up from uranium. Yet until 1940, no elements beyond uranium were known to exist. They were not found because all transuranic elements are radioactive, with lifetimes that are short compared with geologic times. While large quantities may have existed very early in Earth's history, all natural accumulations have long since disappeared.[1]

The understanding of how to create plutonium and other transuranic elements was the result of several breakthroughs in nuclear physics in the 1930s, including the discovery of the neutron in 1932 and of artificial radioactivity in 1934. Those remarkable developments encouraged nuclear scientists, the modern-day alchemists, to pursue the lofty goal of reintroducing these elements to the Earth. But with the discovery of fission in 1938 came the potential to liberate huge amounts of nuclear energy. Once it was realized that plutonium-239 might undergo fission by slow neutrons, the erudite desire to simply create it was quickly superseded by another: to create enough material to build a weapon, one so powerful that it would change the affairs of man.

Chain Reactions

The year 1932 is considered the beginning of modern nuclear physics. In that *annus mirabilis*, the neutron was discovered by James Chadwick, the positron was identified by Carl Anderson, and the particle accelerator of John Cockroft and Ernest Walton was first used to perform artificial disintegrations of the atomic nucleus.

Before the discovery of the neutron,

[1]Minute amounts of transuranic elements are continuously produced on Earth. Through a process described in this article, uranium in uranium ore will absorb neutrons from natural radioactivity and get transmuted into neptunium and plutonium. Also, transuranic elements formed in violent cosmic explosions may spread into the cosmos and fall to Earth.

several groups had shown that bombarding boron and beryllium with α-particles resulted in the emission of a penetrating radiation—one that could knock out protons from absorbers containing hydrogen. Through numerous experiments, Chadwick was able to demonstrate that this radiation was actually a neutral particle with a mass nearly identical to that of the proton.

Dubbed a neutron, such a particle had already been envisaged by Ernest Rutherford as early as 1920, but as a proton-electron combination. Together with Royds, Rutherford had already proved (in 1908) that α-particles were helium nuclei. By 1911, Rutherford showed that essentially the entire mass of an atom was contained in a tiny, positively charged nucleus. After the discovery of the neutron as an uncharged elementary particle, the nucleus was understood to consist of neutrons and protons. The number of protons Z determines the electric charge, or elemental identity, of the nucleus, whereas the mass number A determines the isotopic identity (see Figure 1).

Chadwick's discovery also opened the door to neutron-induced nuclear transmutations. The alchemists' dream of transmuting the elements had already become a reality as early as 1919, when Rutherford demonstrated that bombarding nuclei with α-particles resulted in reaction products that were different from the target nuclei. For example, nitrogen-14 was transformed into oxygen-17 by the reaction

$$ {}^{14}_{7}\mathrm{N} + {}^{4}_{2}\mathrm{He} \rightarrow {}^{17}_{8}\mathrm{O} + p \ . \qquad (1) $$

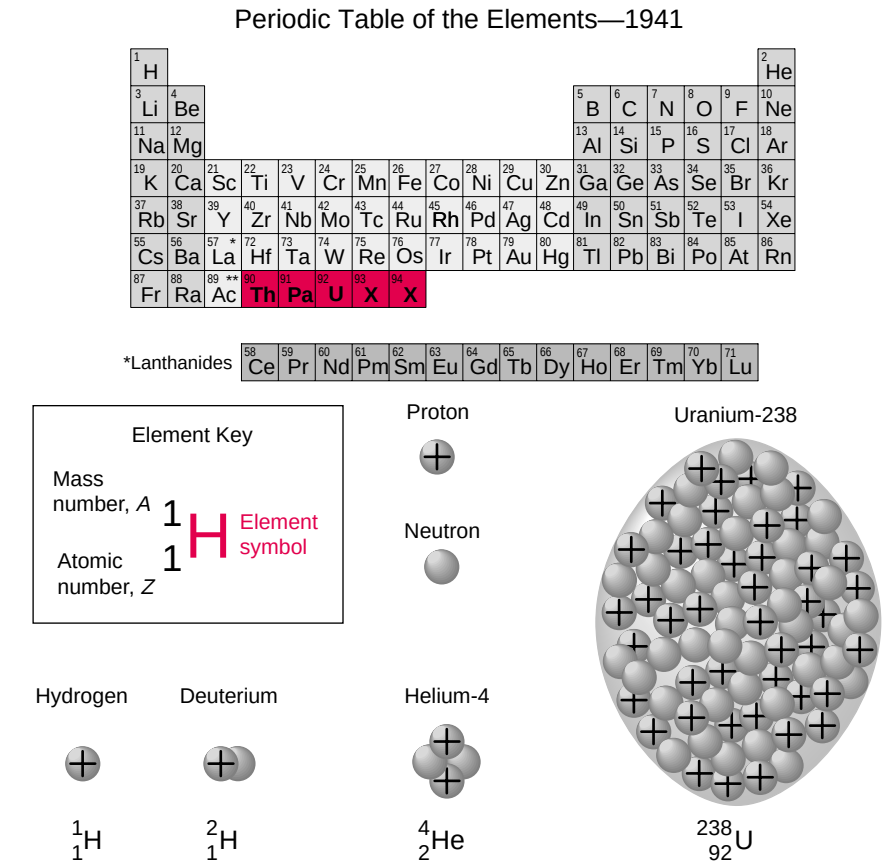


Figure 1. Elements and Nuclei

The top portion of the figure shows the periodic table in 1941. Each column contains elements with similar chemical properties. Each row contains elements arranged in order of increasing atomic number *Z*, the number of protons in the atomic nucleus. In 1941, no elements beyond uranium had been identified, and thorium, protactinium, and uranium were thought to be transition metals. James Chadwick discovered the neutron, a neutral particle with nearly the same mass as the positively charged proton. His discovery led to a concise model of the nucleus. Each has *Z* protons, *N* neutrons, and a mass number *A* = *Z* + *N*. Hydrogen, the first element, has the simplest nucleus (a lone proton). Deuterium is an isotope of hydrogen. (Isotopes of the same element have the same number of protons but different numbers of neutrons.) The nucleus of the next element, helium, is also known as an α-particle. Nuclei increase in size roughly as $A^{1/3}$. Thus, uranium-238 (with 238 nucleons—protons plus neutrons) is a huge nucleus, and it is distorted like a football. Uranium has two common isotopes, uranium-235 and uranium-238.

James Chadwick

The α-particle (helium-4 nucleus) is absorbed by the nitrogen to create a fleeting, unstable nuclear state, which rapidly decays to oxygen-17 by emitting a proton. This reaction and the others known at the time were induced with positively charged projectiles—namely, protons or α-particles—whose energy had to be sufficient to overcome the Coulomb repulsion from the target nucleus. But neutrons have no electric charge and, consequently, no Coulomb barrier to overcome. Therefore, they can penetrate matter and be absorbed by nuclei more easily than charged particles.

The first to realize that the neutron was also the key to releasing the energy embedded in the nucleus was Leo Szilard, a Hungarian physicist. On September 12, 1933, Szilard envisioned neutron-mediated chain reactions. A neutron could induce a nuclear reaction that would emit two or more neutrons and thus act as a neutron multiplier. This nuclear reaction would also liberate energy. Because they are uncharged, emitted neutrons would not lose energy

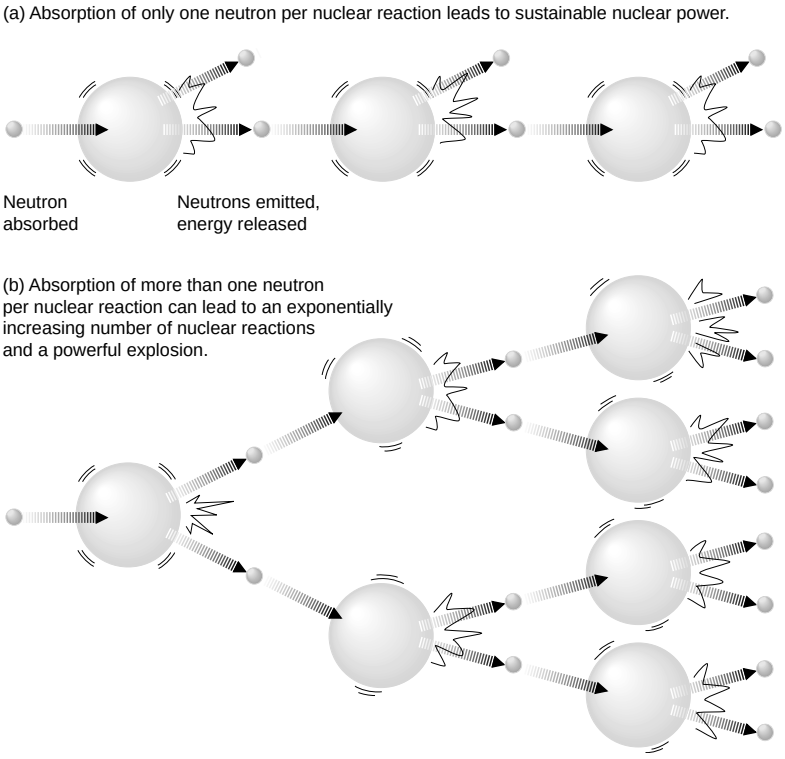


Figure 2. A Nuclear Chain Reaction

Leo Szilard realized that a self-sustaining chain reaction could occur if absorption of one neutron causes a nucleus to emit several others. Each reaction releases energy, and the amount of energy released per unit time depends on the rate at which emitted neutrons are reabsorbed, inducing more reactions. (a) If one neutron is reabsorbed per reaction on average, the chain proceeds in a linear fashion. This is the concept of a nuclear reactor, wherein the number of neutrons is purposely limited to keep the chain reaction under control. (b) If more than one neutron is reabsorbed, on average, the number of nuclear reactions increases geometrically. Without controls, the chain can grow so quickly and release so much energy that a massive explosion occurs.

Leo Szilard

to electrons in matter and would therefore race through a material until they collide with other nuclei. These neutrons could then induce similar reactions, again followed by neutron multiplication. The same process would continue with more and more reactions. If two neutrons were emitted, one reaction would be followed by two, which would be followed by four, then eight, sixteen, thirty-two, sixty-four, and so on. The number of reactions would increase geometrically, as would the total energy released. A self-sustaining chain reaction would be established, resulting in an enormous release of nuclear energy (see Figure 2). If the neutrons were fast, each generation of the chain reaction would occur in a short time, and there would be many generations before the energy liberated by the process blew the material apart. A massive explosion would occur with a force millions of times greater than anything man had previously unleashed.

Harnessing nuclear energy was therefore inextricably linked to creating a nuclear bomb, and Szilard accepted this woeful connection as he began looking for elements that could act as neutron multipliers when bombarded by neutrons. He thought that beryllium, for example, might emit two neutrons when it absorbed one. Szilard also realized that neutrons had to induce neutron-multiplying reactions before diffusing out of the material. Along those lines, Szilard introduced the concept of critical mass (of still unknown elements), or the minimum amount of material needed to sustain a chain reaction. Although Rutherford had stated that energy could not possibly be released from atomic nuclei, by 1934 Szilard had filed a patent on this subject.

Nuclear Transmutations

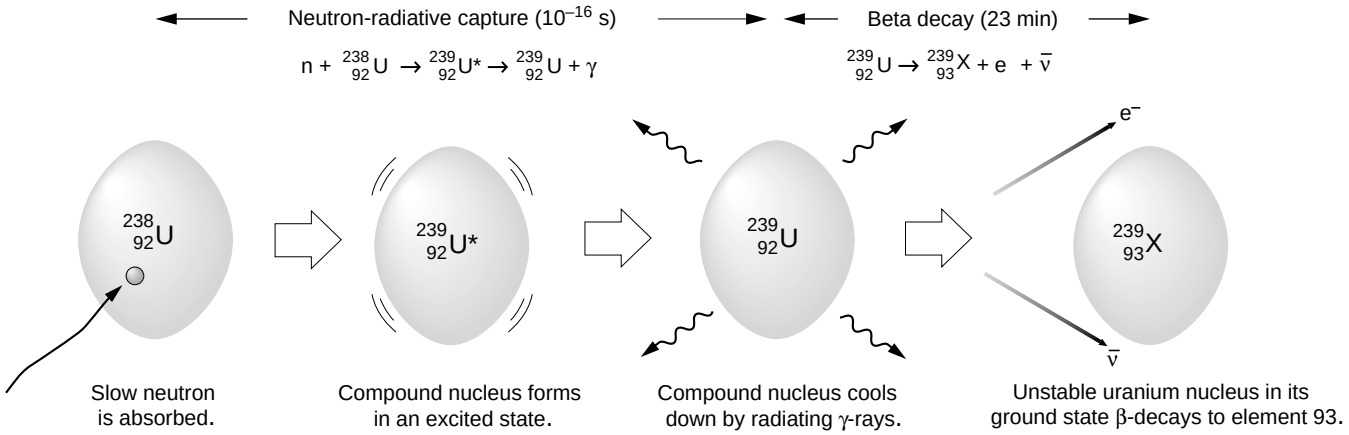
Before Szilard began thinking about chain reactions, nuclear transmutations had been achieved but were initially thought to produce stable nuclei like the oxygen-17 of Reaction (1). In further studies, positrons were observed after light elements such as boron, aluminum, or magnesium had been bombarded with α-particles. Positrons (e^+) are the antimatter counterparts of electrons. They are positively charged and have a mass equal to that of the electron. Frederic Joliot and Irène Curie observed that positron emission continued after α-ray irradiation had been stopped. Furthermore, the number of emitted positrons decreased exponentially with time. The positron signal was therefore similar to what would be expected from the decay of a radioactive element.

Using a chemical precipitation method, Joliot and Curie separated the source of this persistent positron emission from an irradiated aluminum target and showed that the source was an unstable isotope of phosphorus that subsequently decayed into silicon-30 by positron emission:

$$^{27}_{13}\mathrm{Al} + {}^{4}_{2}\mathrm{He} \rightarrow {}^{30}_{15}\mathrm{P} + n \text{ , and} \qquad (2)$$

$$^{30}_{15}\mathrm{P} \rightarrow {}^{30}_{14}\mathrm{Si} + e^{+} + \nu \text{ .} \qquad (3)$$

Joliot and Curie had thus discovered artificial radioactivity (1934). Whereas




Enrico Fermi

Figure 3. Creating Transuranic Elements

Enrico Fermi discovered that irradiating elements with low-energy (slow) neutrons greatly increased the probability of neutron capture. Often, the result was the creation of an unstable nucleus that would radioactively decay. If uranium-238 captured a slow neutron, it would become the compound nucleus uranium-239* (an excited state of uranium-239), which would cool down to the ground state by emitting γ-rays. It was expected that uranium-239 would then undergo β-decay, wherein a neutron in uranium-239 decays into a proton, an electron, and an electron antineutrino, thus creating the first transuranic element, X-239, with atomic number *Z* = 93.

natural radioactivity had been observed in many of the heaviest elements (from thallium to uranium), artificially induced nuclear transmutations could now create new nuclei across the periodic table. Furthermore, Joliot-Curie had rigorously proved their result by chemically isolating their product. In this way, they established a precedent for identifying transmuted nuclei.

Spurred on by these remarkable events, several groups pursued using neutrons, rather than charged particles, to induce nuclear transmutations. The idea was given its greatest impetus in Rome, under Fermi's leadership. Beginning in early 1934, Fermi and his collaborators carried out a systematic study of nearly every element in the periodic table. Stable elements would be bombarded with neutrons, and Fermi would measure the activity, that is, the intensity of the radiation induced in the irradiated sample.

A major step forward occurred when Fermi and colleagues accidentally discovered that the activity increased dramatically when the incident neutrons were slowed down. Neutrons lose energy when they scatter from hydrogen or other light elements, and the insertion of paraffin (which contains lots of hydrogen atoms) between a neutron source and the irradiated sample was sufficient to slow the neutrons down. Even neutrons that had slowed down to room temperature—so-called thermal neutrons with energies of only a fraction of an electron volt (eV)—would lead to a high activity.

The enhanced activity, an indication that many unstable nuclei were being created, was the result of neutron-radiative capture. A slow neutron can be absorbed by a target nucleus to form a relatively long-lived intermediate state known as the compound nucleus. (The compound nucleus model was proposed by Niels Bohr in 1936.) The binding energy of the absorbed neutron is converted into excitation energy of the compound nucleus, which quickly decays by γ-ray emission to its ground state (or sometimes to an isomeric state). The newly created nucleus—an isotope one mass unit higher than the target nucleus—can be unstable, in which case it decays after a characteristic half-life by emitting either α-particles (α-decay) or β-particles (β-decay).

When neutron absorption is followed by β-decay, a neutron (or a proton) in the unstable nucleus transforms into a proton (neutron), creating a β-particle and a type of neutrino. If a neutron transforms, the β-particle is an electron and the neutrino an electron antineutrino, both of which flee the nucleus. The newly created proton remains in the nucleus, so that a new element—one atomic number higher but with the same mass number—is created. Neutron bombardment could therefore be used to produce transuranic elements. As seen in Figure 3, neutron irradiation of uranium-238 would create uranium-239, which was expected to β-decay to element X-239 with atomic number $Z = 93$.

Confident that he would produce the first transuranic element, Fermi tried his neutron source on uranium. But while he observed β-activity to come from the sample, direct confirmation of a new element escaped him. The chemistry of transuranic elements was not known at that time, and separation of β-emitters from irradiated samples

proved difficult. Using chemical separation techniques, Fermi could only prove that the activity did not come from uranium, lead, or any element between them in the periodic table. Fermi postulated that an unexplained β-activity with a half-life of 13 minutes came from a transuranic element, but in the absence of chemical proof would not go beyond this suggestion.

More troublesome was the fact that the radiations emitted by the neutron-activated samples were remarkably complex. Besides the 13-minute activity were several others that could not be identified nor simply interpreted in terms of transuranic radioactive decays.

In September 1934, Ida Noddack suggested that the complex radiations might be coming from nuclear products lighter than lead. Because lead has 10 protons less than uranium, Noddack was suggesting that absorption of a single low-energy neutron resulted in a substantial breakup of the uranium nucleus. This was unimaginable. At the time, all known nuclear reactions resulted in only minor changes of the nucleus. Noddack's suggestion was largely ignored and Fermi himself, after some calculations, patently dismissed the idea.

Noddack was correct however, and ironically, Fermi could have seen evidence for such a nuclear breakup within his own laboratory in early 1935. At that time, Fermi's group made another attempt to detect transuranic elements from irradiated uranium samples, using a recently built ionization chamber. They postulated that, if neutron absorption followed by β–decay produced transuranic nuclei and if those nuclei had a short half-life for α–decay, they would emit energetic α-particles (according to the Geiger-Nuttal law). To reduce low-energy background from natural α-radioactivity, the Italian scientists placed a thin aluminum foil between the uranium sample and the detector. While stopping low-energy α-particles of natural radioactivity, this foil would be partially transparent to the higher-energy α-particles that might be emitted from short-lived transuranic

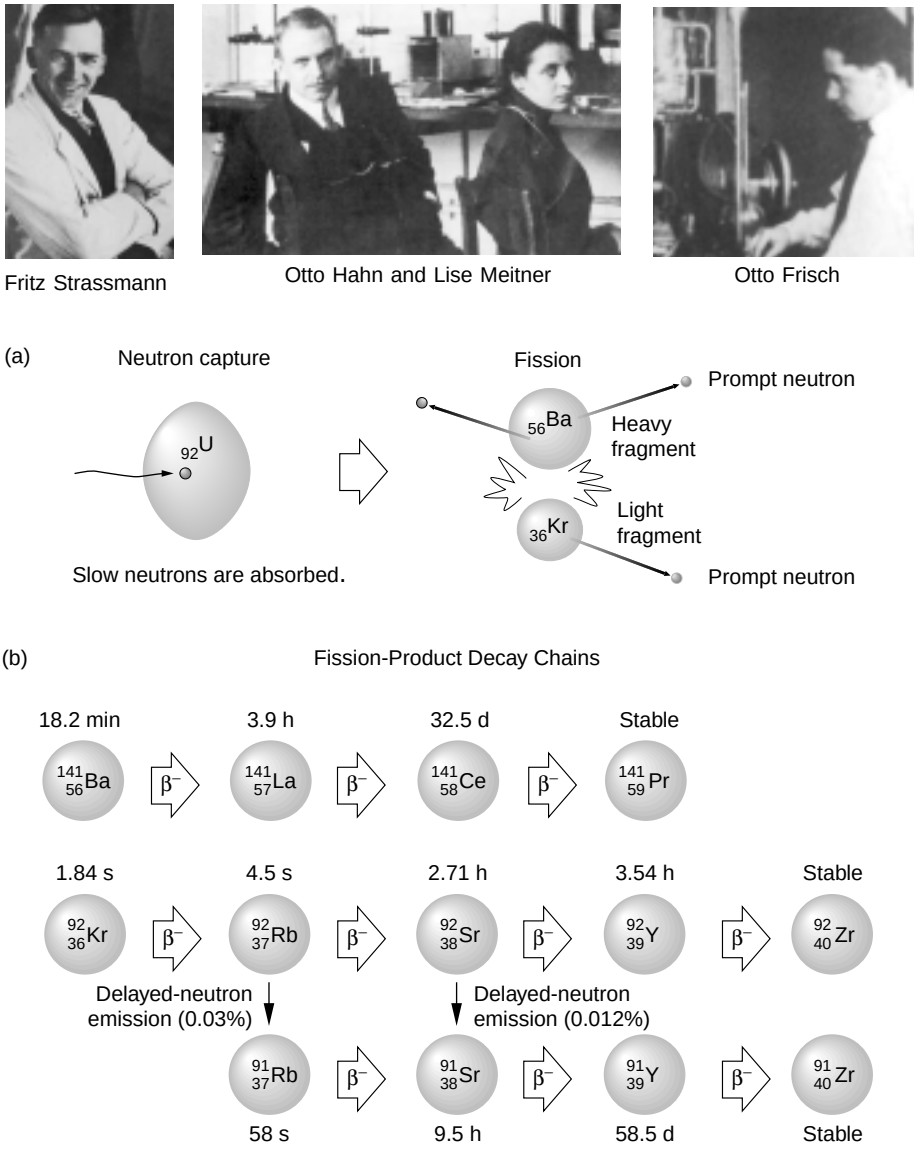
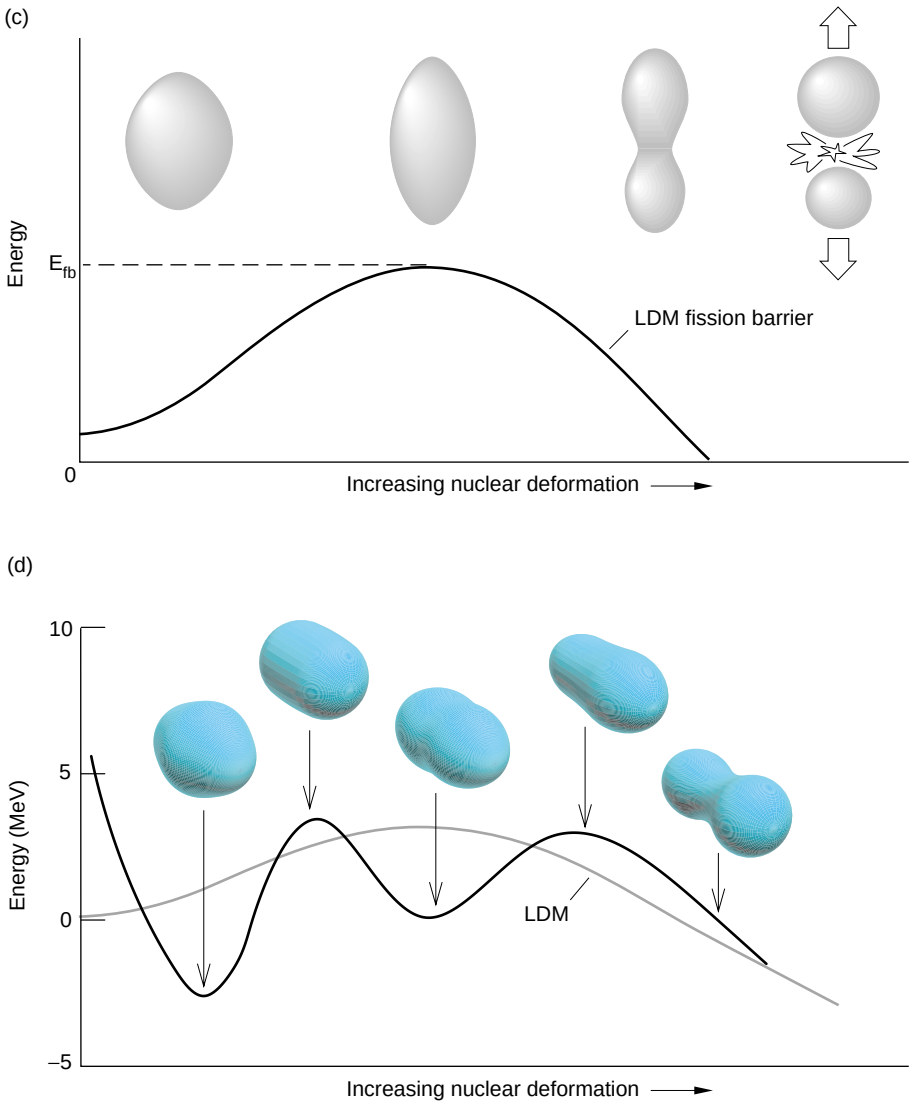


Figure 4. Fission

(a) Hahn and Strassmann chemically isolated radioactive barium from a neutron-irradiated sample of pure uranium. They realized that the uranium nucleus had split into two nuclei, one of which was barium. Fission could explain the confusing activity of neutron-irradiated uranium samples that was observed by groups in Rome, Paris, and Berlin. The fission fragments are born with a significant amount of excitation energy that is dissipated through prompt-neutron and γ-ray emission. Because de-excited fission products are far from the β-stability valley, they subsequently β-decay, and then always emit γ-rays and sometimes delayed neutrons. Also, fission does not produce a unique pair of primary fission fragments before prompt-neutron emission or a unique pair of fission products after prompt-neutron emission. (b) The β-decay chains of fission products are illustrated here for barium-141 (*Z* = 56) and krypton-92 (*Z* = 36). They were obtained after prompt emission of 3 neutrons from the primary fission fragments produced in uranium-235 neutron-induced fission. The γ-ray emission that follows β-decay is not shown. The β-decays from krypton-92 and rubidium-92 (*Z* = 37) are also followed by neutron emission (the delayed neutrons) from rubidium-92 and strontium-92, respectively. Later, experiments proved that both prompt



and delayed neutrons were emitted during fission. (c) Lise Meitner and Otto Frisch used the liquid drop model (LDM) to explain fission. The nucleus is considered to be a positively charged drop of fluid acted upon by two opposing forces. The attractive strong force holds the nucleons together and results in a surface tension that shapes the drop into a compact sphere. The electric (Coulomb) force tries to break the drop apart. As the size of the nucleus increases, the surface tension grows less rapidly than the long-range Coulomb force because the strong force has very limited range. The net result is that larger nuclei become increasingly unstable with respect to shape distortions. It requires additional energy to further distort the nucleus, seen by the fission barrier in the potential-energy diagram. When uranium absorbs a neutron, the excitation energy sets the nucleus oscillating, and the extra energy allows it to deform so much that it tops the fission barrier ($E > E_{fb}$). The nucleus breaks in two. The fission fragments carry off about 170 MeV of kinetic energy, more than a factor of 20 greater than the energy released by α- or β-decay, and tens of millions of times greater than that released by breaking chemical bonds. (d) A modern calculation of the fission barrier and nuclear deformation in plutonium-240 is shown here. The double hump (solid line) arises from considering the detailed quantum mechanics of the nucleus. (Calculation courtesy of Peter Möller, Los Alamos National Laboratory.)

elements. The experiment proved unsuccessful—no α-particles were detected—but fission products were undoubtedly produced and would have been detected if the aluminum foil had not been there to stop them!

The multitude of β-activities from neutron-irradiated uranium remained a mystery for quite a while. Indeed, by 1935, two other groups had observed similar startling results: Irene Curie and Pavel Savitch in Paris, and Lise Meitner, Otto Hahn, and Fritz Strassmann in Berlin. A 23-minute activity seemed to originate from uranium-239 and would imply the creation of X-239, but neither the Paris nor the Berlin scientists could explain their results convincingly.

All these studies and speculations took a radically different turn with the discovery of fission by Hahn and Strassmann in December 1938. (Lise Meitner, who was Jewish, had to flee the Nazi regime. She left Berlin in July.) Using radiochemical methods, the team clearly demonstrated that a pure uranium sample contained radioactive barium (Z = 56) after having been irradiated by neutrons. The barium could only come from the splitting, or fission, of the uranium nucleus. Fission could explain the confusing spectrum of radiation that was observed after neutron-induced activation of uranium—the radiation came in part from the great variety of fission fragments and their descendants (see Figure 4). Also, Szilard's vision of nuclear energy released from nuclear chain reactions suddenly switched from dream to reality. The prompt emission of neutrons from the hot fission fragments would provide the neutron multiplication mechanism needed for a chain reaction.

Fission

Although nuclear fission had escaped theoretical prediction, it was immediately explained in terms of existing nuclear models. Like neutron-radiative capture, neutron-induced fission was interpreted as one mode of decay of the compound nucleus. The details could easily

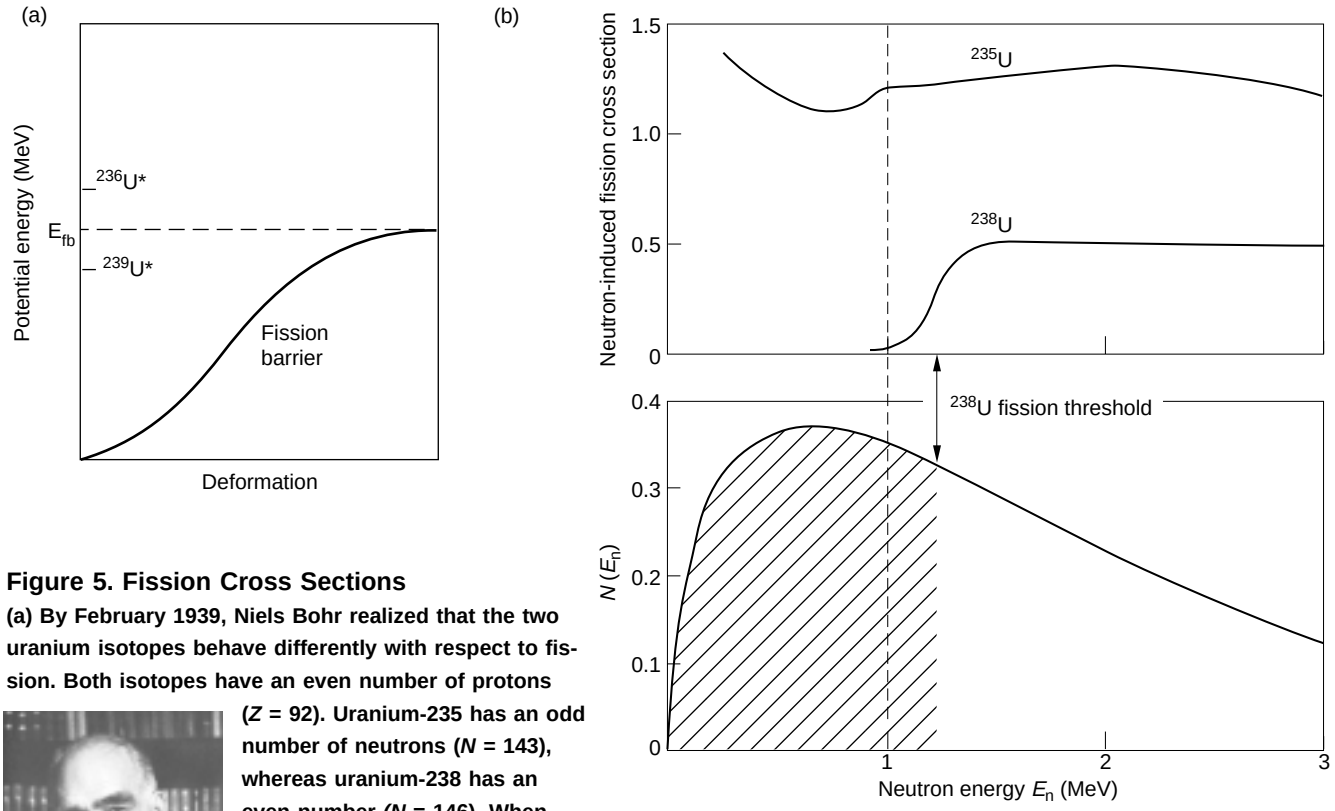


Figure 5. Fission Cross Sections

(a) By February 1939, Niels Bohr realized that the two uranium isotopes behave differently with respect to fission. Both isotopes have an even number of protons (*Z* = 92). Uranium-235 has an odd number of neutrons (*N* = 143), whereas uranium-238 has an even number *(N* = 146). When uranium-238 absorbs a neutron, it forms the even-odd nucleus uranium-239 and gains about 4.8 MeV of binding energy, which goes into nuclear excitation. This amount of energy is not enough to top the fission barrier of about 6 MeV, so uranium-238 needs to absorb a neutron with kinetic energy greater than about 1 MeV in order to fission. But the strong force that binds the nucleus together "likes" to pair up protons and neutrons; hence, when uranium-235 absorbs a neutron to form the even-even nucleus uranium-236, it gains about 6.5 MeV of binding energy. That amount is more than enough to allow the excited nucleus to fission regardless of the incident-neutron energy. Uranium-235 is fissile. (b) The top graph shows the different fission cross-sections for uranium isotopes as a function of neutron energy. At thermal energies of about 0.025 eV, the uranium-235 cross section is about 500 barns. In the bottom graph, the energy spectrum of prompt neutrons emitted in the slow-neutron-induced fission of uranium-235 is quite wide, with an average energy of about 2 MeV. About half of these neutrons cannot induce fission in uranium-238 (hatched area). The fraction that cannot induce fission is even higher in nuclear devices because the neutrons lose energy through nuclear interaction with device components.

Niels Bohr

be described within the context of the liquid-drop model (LDM), originally put forward by George Gamow in 1928 and then given a much more serious development by Bohr in 1937. The LDM ignores the discrete nature of the protons and neutrons. Instead, it models the nucleus as an electrically charged liquid drop that can deform, vibrate, or split. It can also merge with another drop as in fusion reactions. The model provided a straightforward prescription for calculating basic nuclear properties, such as the mass and size of nuclei.

Meitner had remained in close communication with the Berlin group, and within a week of the discovery of fission, she and her nephew Otto Frisch used the LDM to describe the process. In the heuristic picture presented in Figure 4, the drop-like compound nucleus deforms so much upon absorbing a neutron that it elongates into a dumbbell shape that finally scissions into two droplets, also known as fission fragments. (Frisch was the one who coined the term "fission" after a discussion with the American biologist William Arnold about the splitting of bacterial cells.) The electrically charged droplets repel each other, and they instantly fly apart to rapidly reach about 170 million electron volts (MeV) of total kinetic energy after full acceleration. By mid-January of 1939, Frisch had observed these energetic fission fragments with an ionization chamber, and subsequently observed neutron-induced fission in thorium-232.

Experimental studies quickly focused on whether neutron emission accompanied fission. R. B. Roberts, R. C. Meyer, and P. Wang in the United States first showed that "delayed" neutrons were emitted following the β-decay of fission products. But other studies soon revealed

that, as the initial fission fragments form, they immediately release "prompt" neutrons. Fermi, Szilard and collaborators at Columbia University, and Joliot and his collaborators at Paris demonstrated that more than one prompt neutron was emitted during fission. Joliot and collaborators first reported this finding for uranium, with the precise average number of 3.5 ± 0.7 prompt neutrons. The modern figure is now an average of 2.4 prompt neutrons per fission.

Therefore, the prospect of nuclear energy liberated from fission chain reactions came within reach, but as understood by groups in the United States, Western Europe, the Soviet Union, and later Japan, the prospects of achieving an explosive chain reaction using natural uranium were dim. Shortly after the discovery of fission in thorium, Bohr had analyzed the available data and concluded that only uranium-235, not uranium-238, fissioned when bombarded with slow neutrons. Neutrons of any energy could induce the lighter isotope to fission (it was said to be a fissile material), whereas neutrons had to have energies on the order of 1 MeV or more to induce uranium-238 to fission.

The difference stems from the fact that uranium-235 has an odd number of neutrons and uranium-238 has an even number (both isotopes have an even number of protons). Thus, when uranium-235 absorbs a neutron, it pairs with the odd neutron, creating additional excitation energy (or pairing energy) and pushing the resulting uranium-236 compound nucleus above the fission barrier (see Figure 5). Uranium-238 has no unpaired neutron to match up with the absorbed neutron, and thus no pairing energy and no fission results until the incident neutron brings in the needed energy. Bohr and John Wheeler used the LDM to strengthen the argument in their 1939 landmark paper on fission. In March 1940, by comparing slow -neutron-induced fission in separated uranium isotopes, Alfred O. Nier et al. finally provided experimental proof that uranium-235 was fissile.

In a nuclear device, however, fast neutrons would have to induce enough fission reactions and release enough energy before the material is blown off. In this respect, the role of the two uranium isotopes strongly depends on the energy spectrum of fission neutrons. It was later shown that fission neutrons have a wide energy spectrum with an average energy of about 2 MeV. Given this spectrum, only half of these fission neutrons can induce fission in uranium-238 but this fraction would be even lower in a nuclear device, where the neutron energy is degraded through nuclear interactions with the components of the device (see Figure 5). Even with nuclear data available at that time, uranium-238 appeared unsuitable for nuclear detonations. Uranium-235 is much more suitable, but its isotopic abundance in natural uranium is only 0.7 percent. Thus, a bomb made of natural uranium would be very inefficient. In May 1940, French physicist Francis Perrin estimated the critical mass of natural uranium to be on the order of tens of tons. Shortly thereafter, more-precise calculations by Rudolf Peierls reduced this number to some tons. But similar calculations for pure uranium-235 made by Frisch and Peierls led to a critical mass of a few pounds for this isotope. This was well within the realm of a bomb.

Obtaining pure uranium-235, however, was a severe problem. Because the chemical properties of isotopes are identical, chemical separation of uranium-235 from uranium-238 was not possible. The fissile isotope could be obtained in large amounts only through a separation process that exploited the slight 1.2 percent difference in mass between uranium-235 and uranium-238. The most promising method was a gaseous diffusion method,which had previously been developed by the German physical chemist Klaus Clusius. But the equipment and facilities needed to achieve the separation would have to be constructed on a gigantic scale. Frisch estimated that approximately 100,000 tubes would be necessary. This number was, however, drastically reduced to 5,000 tubes in subsequent developments. Still, with the European continent at war and the possibility that German scientists might be developing a bomb, plans were made to separate uranium isotopes and build a uranium-235 bomb.

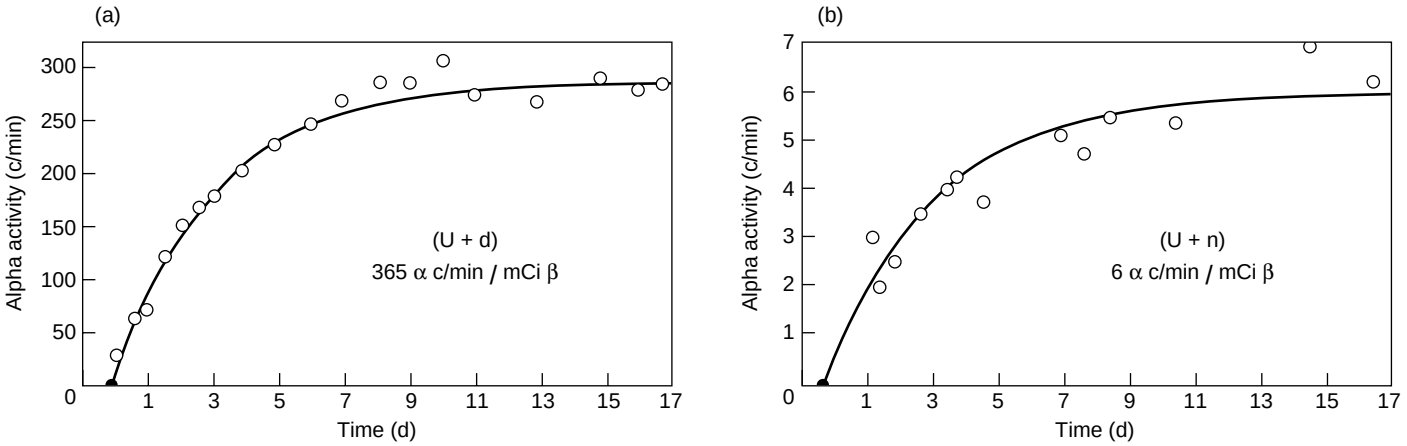
Almost as these plans were being drawn up, Bohr's analysis of fissionable nuclei had another implication. The transuranic element 239 with $Z = 94$ protons and 145 neutrons, could also be fissile. In May 1940, Turner outlined this possibility in a letter to Szilard, who urged secrecy, for he realized that if element-239 with $Z = 94$ were created, subsequent separation of this transuranic element from uranium could be done through chemistry, thus obviating the difficult and expensive need to separate isotopes.

Seven months later, Fermi and Segrè came to the same conclusion. They also understood that the neutron flux within a nuclear reactor—a device that allows for a self-sustaining, controlled chain reaction—would be extremely high. Fermi had believed that a controlled chain reaction could be achieved in natural uranium if slow neutrons were used. He realized that neutron-radiative capture by the uranium-238 reactor fuel would then produce large quantities of element-239.

Studies on chain reactions were soon concentrated at Columbia. The use of graphite as a neutron moderator proved feasible when measurements of neutron attenuation in a large graphite assembly proved that the absorption of neutrons by carbon was lower than anticipated. Also, measurements of the multiplication factor in a lattice structure of graphite and uranium gave hope that a multiplication factor greater than 1 could be achieved. Therefore, a nuclear reactor could possibly be made with such a lattice.

The Discovery of Neptunium

The discovery of fission in the waning days of 1938 had cleared the way for more-precise studies on transuranic



Glenn Seaborg

Art Wahl

Figure 6. Discovery of Plutonium at Berkeley
After failing to detect plutonium-239 from the neptunium-239 sample produced in neutron-irradiated uranium, the Berkeley team irradiated uranium with deuterons. This deuteron-induced reaction produced two neptunium isotopes—neptunium-239 with a β-decay half-life of 2.3 days and neptunium-238 with a β-decay half-life of 2.1 days. The data in (a) show the α-activity in the neptunium sample from deuteron-irradiated uranium. It was observed with a new high-sensitivity ionization chamber. Although neptunium-238 contributed only 5% of the total β-activity, the product of that decay (plutonium-238) produced more than 98% of the observed α-activity. The α-activity in (b) originated from plutonium-239 that had grown in a neptunium sample from neutron-irradiated uranium. (Early experiments by McMillan and Abelson were not sensitive enough to detect this small plutonium-239 α-activity.)

elements. Taking into account the radioactivity of fission products, the German team confirmed that a previously observed β-activity with a 23-minute half-life actually came from uranium-239. While they failed to detect the first transuranic, their work helped an American research effort to succeed.

At Berkeley, Edwin McMillan began using cyclotron-produced neutrons to irradiate a thin foil of uranium. The energetic fission fragments would fly out of the foil. McMillan discovered that the foil retained some activity. In addition to the known 23-minute activity, indicative of uranium-239, another 2.3-day activity remained. Although this last activity could not have originated from fission fragments, the source behaved chemically like the rare earths, which are abundant in most fission fragments. This puzzling inconsistency led McMillan and Philip Abelson to repeat the experiment. The second time, they found that the 2.3-day activity came from an element with chemical properties different from those of any other element but close to those of uranium.

Moreover, McMillan and Abelson demonstrated in May 1940 that the 2.3-day activity grew from uranium-239. They carefully purified the activated uranium sample and then obtained precipitates at 23-minute intervals, using equal amounts of cerium as a carrier. The activity of the precipitate was measured a day later to allow any uranium-239 to totally decay away while preserving most of the 2.3-day activity. The intensity of the latter activity showed a 23-minute exponential decay as a function of the time the precipitate was taken from the activated uranium sample. This demonstrated that uranium-239 decays with a 23-minute half-life to a new radioactive transuranic element that has a 2.3-day half-life.

McMillan and Abelson had thus discovered the first transuranic element with mass number 239 and atomic number $Z = 93$. They called it neptunium—after the planet Neptune, which was next in line beyond Uranus in the solar system. Because the chemical properties of neptunium-239 were different from those of rare earths but close to those of uranium, McMillan and Abelson suggested for the first time that neptunium might belong to a second "rare earth" group of similar elements starting with uranium.

Their suggestion was prophetic. Glenn Seaborg later proved that this second "rare earth" group actually starts after actinium ($Z = 89$). Today, the group is referred to as the actinides. (See the box "The Actinide Concept" on page 368.)

The Discovery of Plutonium

The next step was the formation and identification of the transuranic element with $Z = 94$, soon to be dubbed "plutonium" even before its discovery. (The new element was named after Pluto, after the next planet beyond Neptune in the solar system.) As a follow-up of their studies on neptunium-239, McMillan and Abelson had already attempted to identify plutonium-239 as

the product of the 2.3-day decay of neptunium-239. Plutonium was expected to decay by emission of α- rather than β-particles, and so McMillan and Abelson looked for radiation from α-decay (or spontaneous fission[2]) from a purified sample of neptunium-239 obtained from neutron-irradiated uranium. They failed to detect any, and the negative result led them to postulate that the half-life for these two processes in plutonium-239 was greater than a million years.

McMillan left Berkeley shortly thereafter to help develop radar for the war effort while Kennedy, Seaborg, Segrè, and graduate student Art Wahl continued the pursuit of plutonium. A major step forward occurred when the group used the 60-inch cyclotron at Berkeley to irradiate uranium with deuterons with the hope that another plutonium isotope, shorter-lived than plutonium-239, could be formed, whose α-decay could be more easily detected. Studies of γ-rays and β-rays emitted from these deuteron-irradiated uranium samples revealed the presence of two neptunium isotopes—neptunium-239 and another that accounted for about 5 percent of the β-ray activity. This other isotope (later identified as neptunium-238) had a half-life of 2.1 days, close to that of neptunium-239. These neptunium samples also showed the existence of an α-emitter whose chemical properties were found to be different from those of neptunium, uranium, and all other known elements. The only possibility was that this α-emitter was the second transuranic element—plutonium—which was created by the β-decay of neptunium.

The growth of plutonium from the neptunium samples was later studied in the spring of 1941 from the radiation emitted by these samples with the help of a newly developed ionization chamber that could be placed behind the poles of the magnet. With this arrangement, β–rays were deflected away from the ionization chamber but not the α–rays, which could then be more easily detected. The growth studies showed that the α-decay detected from neptunium chemically separated from deuteron-irradiated uranium samples came mostly from plutonium-238, which was produced by the β-decay of the neptunium-238. (see Figure 6). Although this decay accounted only for 5 percent of the total β-decay in the sample, the grown-in plutonium-238 has a relatively short half-life (88 years) and therefore a relatively high α-activity.

The same sensitive technique was used with neptunium samples chemically separated from neutron-irradiated uranium, and the α-ray activity from plutonium-239 was finally detected. Previous experiments with neutron-irradiated uranium were not sensitive enough to detect the relatively small α-activity of plutonium-239, which is due to its relatively long half-life (24,000 years).

Making Bulk Plutonium

Small but ponderable quantities of plutonium-239 (0.5 microgram), produced using cyclotrons, soon became available and were used to demonstrate that fission induced by thermal neutrons was larger by a factor of 1.28 than for uranium-235. A larger plutonium-239 sample (3.5 micrograms) was used in July 1941 to demonstrate that its fission cross section with fast neutrons was also larger than that for uranium-235. Although uncertainties still existed about the number of prompt fission neutrons emitted, it already appeared that plutonium-239 was a strong competitor to uranium-235 for nuclear explosives.

Then, a remarkable effort followed for making enough plutonium to build a bomb. At the beginning of 1942, all work on plutonium was supervised by the Metallurgical Laboratory at the University of Chicago and was placed under the leadership of Arthur Compton. Three missions were given to this laboratory: (1) to find a system using natural uranium in which a controlled chain reaction could take place, (2) to find a chemical separation technique for plutonium formed in the irradiated uranium fuel, and (3) to obtain theoretical and experimental nuclear data relevant to an explosive chain reaction with uranium or plutonium.

The first objective, which had already been started by Fermi and Szilard at Columbia, gained momentum when the work shifted to the University of Chicago. In an abandoned squash court, Fermi assembled what he knew would be a self-sustaining nuclear reactor. Twenty-five feet high and 20 feet wide, the reactor was built from 349,263 kilograms of pure carbon cut as large graphite bricks. Blind holes were drilled into about a quarter of the bricks, and about 36,500 kilograms of uranium oxide and 5600 kilograms of pure uranium metal were pressed into thousands of fuel elements and dropped into the holes. An active brick layer contained an array of evenly spaced uranium plugs. These were sandwiched between two dead layers of solid graphite. As the two types of layers were alternated and stacked, a full 3-dimensional lattice of uranium plugs was formed (see photo on article's opening page).

Collisions with the carbon atoms in the graphite would slow the neutrons to thermal energies, whereupon they would diffuse until they encountered a uranium-235 atom. The chain reaction was held in check by cadmium control rods that would absorb neutrons without fissioning. As the number of layers increased, the reactor came increasingly close to critical mass. On December 2, 1942, with the final layers in place and the control rods removed, the reactor went critical and sustained the world's first manmade nuclear chain reaction.

Several steps were taken to meet the second objective—the bulk-scale chemical separation of plutonium from reactor fuel. By the end of 1942, about 500 micrograms of mostly plutonium-239 had been obtained from large quantities of neutron-irradiated uranium. The irradiation had been carried out at the cyclotrons at Berkeley and Wash-

[2]Spontaneous fission, wherein the nucleus undergoes fission without an initiating neutron, was discovered in uranium by George Flerov and K. A. Petrjak of the Soviet Union in June, 1940.

ington University. Chemical studies with these first milligram samples showed that it would be possible to separate plutonium from other elements in the reactor fuel.

Subsequent developments on a larger scale included the construction of a pilot nuclear plant at Clinton, Tennessee. This plant was to produce plutonium in quantities larger than the fraction of a milligram obtained from cyclotrons in order to demonstrate the possibility of plutonium separation on an industrial scale and study the biological effects of radiation. The Clinton reactor started operation on November 4, 1943, by producing 500 kilowatts, and it reached 1800 kilowatts in May 1944. Plutonium was separated from slugs of irradiated uranium fuel by remote control and behind thick shields. The first slug was treated for plutonium separation on December 20, 1943, and by March 1944, several grams of plutonium were available. These larger quantities of plutonium were also essential in obtaining nuclear data on plutonium.

The last step toward the production and separation of plutonium-239 was the construction of three nuclear piles on the Hanford site, whose purchase was approved by General Groves almost immediately after the creation of Met Lab. These reactors were designed to supply enough plutonium-239 for nuclear devices. The first pile started operation in September 1944, and the three piles were in operation by the summer of 1945.

Met Lab's third objective—obtaining precise nuclear data on plutonium-239—was met by experiments carried out at Berkeley and at the new, secret Los Alamos site, where J. Robert Oppenheimer had been appointed director. Although early data already indicated that plutonium would be fissile, more precise and sometimes new data were needed to design a weapon. Most of these data were obtained by measurements of fission cross sections and fission neutrons. The number of prompt neutrons emitted per fission of plutonium-239 proved to be higher than for uranium-235, which enhanced the potential for using plutonium-239 as a weapons material.

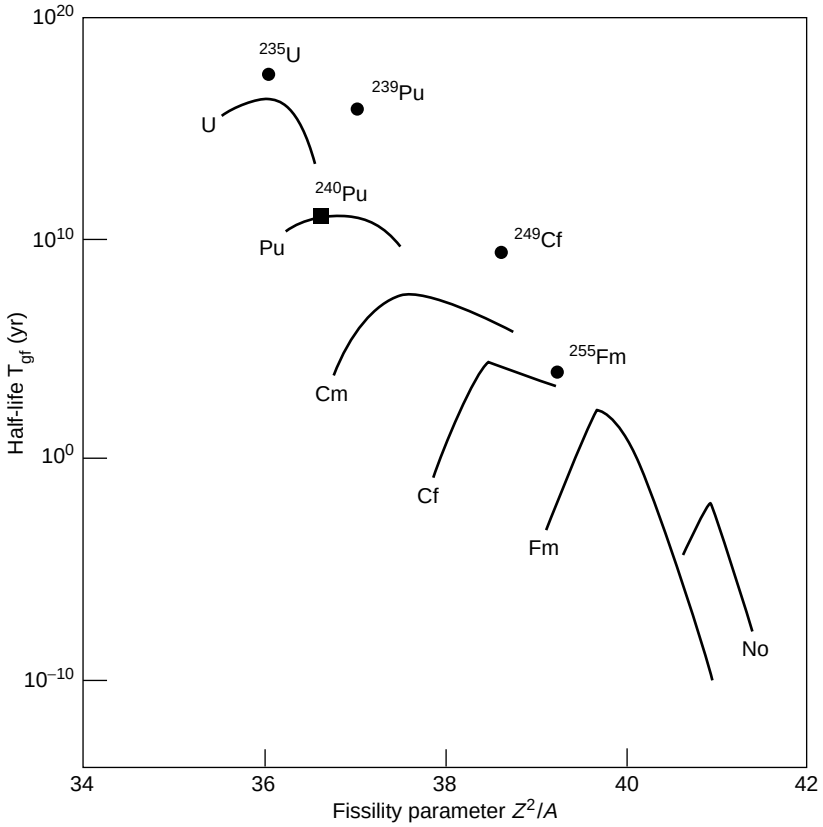


Figure 7. Spontaneous Fission Half-Lives

Experimental spontaneous-fission half-lives T_{gf} for isotopes of transuranic elements are plotted as a function of the fissility parameter Z^2/A (Z and A are the atomic and mass number, respectively, for each isotope). The solid lines are drawn through the half-lives of isotopes having even numbers of protons (Z) and neutrons (N). These even-even isotopes have spin 0 and positive parity. The solid square is for plutonium-240. The spontaneous fission half-lives of isotopes having even Z but odd N (such as uranium-235, plutonium-239, californium-249, and fermium-255), indicated by filled circles, are systematically higher than those for the even-even isotopes of the same element. These even-odd isotopes have a spin different from zero and a parity that can be either positive or negative. Their spontaneous-fission half-lives are longer because the fission barrier is higher to accommodate their spin and parity. Similarly, the α-decay half-life of even-odd isotopes is higher than for neighboring even-even isotopes (see the discussion of plutonium-238 and plutonium-239 in the text) because the α-particles have to penetrate a higher potential barrier—another manifestation of a spin effect—in these even-odd nuclei.

Although these plutonium-239 data were encouraging, other data seemed to indicate that plutonium-239 might not be used in the weapon that was being designed at Los Alamos. The plutonium samples obtained from the Clinton pilot plant were emitting an unexpectedly high neutron intensity from spontaneous fission, which had not been seen in the samples from the Berkeley cyclotron. The high neutron flux and long irradiation times in the Clinton reactor allowed plutonium-239 itself to capture neutrons and become plutonium-240, which has a high spontaneous fission rate. The spontaneous fission rate in plutonium-239 is hindered by a spin effect that makes the fission barrier higher than for plutonium-240 (see Figure 7). The plutonium from the Hanford piles would have an isotopic composition similar to that of the

plutonium produced in the Clinton pile.

At the time of this discovery, the favored weapon design was the "gun-type" design, wherein a subcritical amount of fissile material would be literally fired—like a bullet from a gun—into another subcritical mass. The rapid assembly of a critical mass from these two fissile pieces would result in a runaway chain reaction and a nuclear explosion. But the ever-present emission of neutrons from spontaneous fission would cause premature fissioning of the plutonium before a critical mass was achieved. A plutonium gun-type weapon would thus fizzle.

The use of plutonium in a nuclear detonation, therefore, required the formation of the critical mass with a faster method. This new method was found with the implosion of a plutonium shell, whose efficiency was later demonstrated at the Alamogordo test. ■

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