# A Factor of Millions
## *Why we made plutonium*

*with André F. Michaudon and Ileana G. Buican*



**The seminal work of (from the top) Irène Curie and Frédéric Joliot, Ernest Rutherford, James Chadwick, and Niels Bohr opened the modern era of nuclear physics.**

Until the middle of the 20th century, humans had managed to extract only chemical energy from atoms. That energy is carried by electrons orbiting around the heavy, tiny nucleus at the atom's center. Released when conventional fuel is burnt or explosives are detonated, that energy is on the order of electron volts per atom. Yet early in the century, scientists—particularly those educated in the great European school of physics—were already engaged in probing the secrets of the nucleus. In 1938, they discovered that the nucleus could be split through a process called fission, unleashing huge amounts of energy. Suddenly, the scale of energy release changed from electron volts to millions of electron volts, which became available for powerful explosives and civilian energy production. And it was the Manhattan Project pioneers who managed to extract the millionfold advantage of nuclear over conventional explosives during a wartime race with Nazi Germany.

### The Discovery of Nuclear Fission

When James Chadwick discovered the neutron in 1932, he paved the way for a greater understanding of nuclear reactions. Indeed, in September 1933, the Hungarian physicist Leo Szilard envisioned that neutrons could be absorbed by a nucleus and induce a repeatable chain reaction that would lead to the controlled release of atomic energy. Szilard then also thought of using neutron-induced chain reactions to create explosions. In 1934, while in London, he filed and was awarded a patent on that concept, thus becoming the legally recognized inventor of the atomic bomb.

That same year, Frédéric Joliot and Irène Curie demonstrated that stable elements could be made radioactive. After irradiating aluminum nuclei with α-particles, they detected the neutrons produced but also a signal of radioactivity. The scientists deduced that the signal was coming from a short-lived phosphorus isotope into which some of the aluminum must have transmuted. Using a chemical precipitation

PATENT SPECIFICATION 630,726

Application Date: June 28, 1934. No. 19157/34.
" " July 4, 1934. No. 19721/34.

One Complete Specification left (under Section 16 of the Patents and Designs Acts, 1907 to 1946): April 9, 1935.

Specification Accepted: March 30, 1936 (but withheld from publication under Section 30 of the Patent and Designs Acts 1907 to 1932)

Date of Publication: Sept. 28, 1949.

Index at acceptance:—Class 39(iv), P(1:2:3x).

PROVISIONAL SPECIFICATION
No. 19157 A.D. 1934.

Improvements in or relating to the Transmutation of Chemical Elements

I, LEO SZILARD, a citizen of Germany and subject of Hungary, c/o Claremont Haynes & Co., of Vernon House, Bloomsbury Square, London, W.C.1, do hereby declare the nature of this invention to be as follows:—

This invention has for its object the production of radio active bodies the storage of energy through the production of such bodies and the liberation of nuclear energy for power production and other purposes through nuclear transmutation.

**In 1934, the irrepressible Hungarian physicist Leo Szilard filed a patent for the atomic bomb and the release of nuclear energy.**

A nucleus, mimicked here by a drop of oil, can deform and break in two.

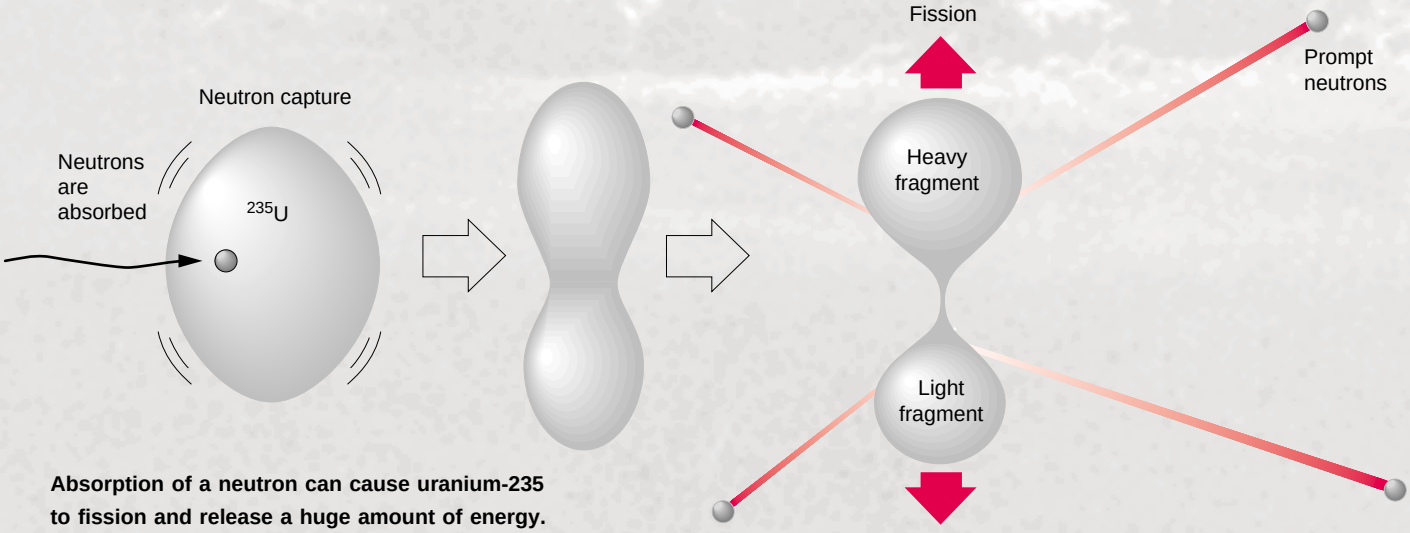
method, Joliot and Curie separated the source of the radioactive signal from the aluminum target and proved that the signal was indeed coming from phosphorus. Their technique for studying nuclear transmutations was adopted universally, and the neutron, which has no electric charge, was immediately seen as an ideal "projectile" for inducing nuclear transformations.

In Rome, Enrico Fermi, Edoardo Amaldi, Emilio Segrè, and Franco Rasetti systematically bombarded all the elements in the periodic table with neutrons. Finally, they bombarded uranium, hoping to create elements beyond uranium (or transuranic elements). Although Fermi had shown that slow neutrons (those whose energies are on the order of electron volts or less) created more radioactivity than fast ones, the Italian scientists did not identify any clear signal of transuranic elements from the array of induced radioactivities. Nor did they know that, in the process of bombarding uranium with neutrons, they must have produced nuclear fission. Later, Emilio Segrè reminisced about those events and admitted that, "We did not seriously entertain the possibility of nuclear fission although it had been mentioned by Ida Noddack, who sent us a reprint of her work. The reason for our blindness, shared by Hahn, Meitner, the Joliot-Curies, and everybody else working on the subject, is not clear to me even today." (*A Mind Always in Motion: the Autobiography of Emilio Segrè*, 1993, Berkeley, CA: University of California Press.)

In December 1938, the German chemists Otto Hahn and Fritz Strassmann discovered that one of the induced activities in neutron-irradiated uranium came from an isotope of barium, a nucleus with roughly half the mass and charge of the uranium nucleus. The scientists were astonished at their findings.

Their collaborator Lise Meitner, then in exile in Sweden, and her nephew Otto Frisch quickly explained the new phenomenon in terms of Niels Bohr's liquid-drop model. When it absorbs a neutron, the positively charged nucleus begins to vibrate, behaving like a drop of liquid. As shown in the illustration below, the vibration deforms the large uranium nucleus so much that the nucleus splits into two smaller nuclei that fly apart, a process accompanied by a tremendous release of

(From top left): Enrico Fermi, Fritz Strassmann, Otto Hahn and Lise Meitner, and Otto Frisch.




**Absorption of a neutron can cause uranium-235 to fission and release a huge amount of energy.**

energy. After talking with biologist William Arnold about fissioning bacteria, Frisch borrowed the terminology for splitting nuclei.

On January 26, 1939, Niels Bohr publicly announced the discovery of fission at an annual theoretical-physics conference at George Washington University. Physicists from the United States, the Soviet Union, and Western Europe immediately understood the implications—building an atomic bomb from uranium became a possibility. The fission products are created in an excited state and would likely lose energy by "boiling off" neutrons (referred to as prompt neutrons). These neutrons could be captured by other nuclei and induce more fission events, as Szilard had envisioned in 1933. Depending primarily on the arrangement and density of the fissile material, fission could proceed in either a controlled, self-sustaining manner and produce nuclear power or in a "runaway" fashion and result in a nuclear explosion.

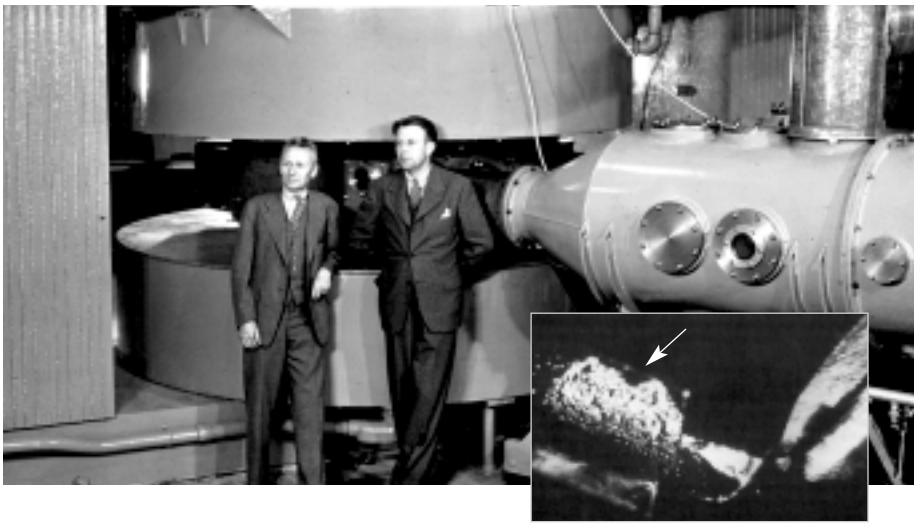
By February 1939, Niels Bohr had deduced that uranium-235 is the only naturally occurring fissile uranium isotope (a fissile isotope is one in which fission can be induced by neutrons of any energy, no matter how small). In September that same year, Bohr and Wheeler published a theoretical analysis of fission. The nuclear physics is such that the odd number of neutrons (143) in uranium-235—as opposed to the even number of neutrons (146) in uranium-238 (both isotopes have 92 protons, an even number)—makes all the difference. But natural uranium contains only 0.7 percent uranium-235. The rest is uranium-238. Using natural uranium for a bomb was clearly inefficient because too few fission events could occur in 99.3 percent of the material.

A successful fission bomb would require a few kilograms of uranium-235, but obtaining those quantities was difficult because the fissile isotope could not be chemically separated from uranium-238. In the meantime, however, the 1939 paper by Bohr and Wheeler had inspired several scientists to postulate that a transuranic element with 94 protons and 145 neutrons—plutonium-239—should also be fissile. Plutonium offered the attractive alternative of a fissile element that could be chemically separated from uranium. But every atom of plutonium would have to be made.

**After Ed McMillan discovered neptunium, he started the process of making plutonium. But it was wartime, and McMillan was summoned to work on radar at the Massachusetts Institute of Technology. As Joseph Kennedy (below, right photo) later put it, McMillan got on the train, and the others found plutonium.**

**Art Wahl (left) vividly remembers his excitement the night he and Kennedy first isolated plutonium. "By the time I finished the second part of the experiment—reducing the oxide and recovering the plutonium—Kennedy had gone home. So, I went to bed with the secret of the discovery of plutonium all to myself."**

**(From center to right): Glenn Seaborg, Art Wahl, and Ed McMillan accept a plaque from Undersecretary of the Interior John Carver, Jr., on the occasion of the 25th anniversary of plutonium's discovery. The ceremony took place in Gilman Hall, in front of Room 307, where plutonium had been isolated.**

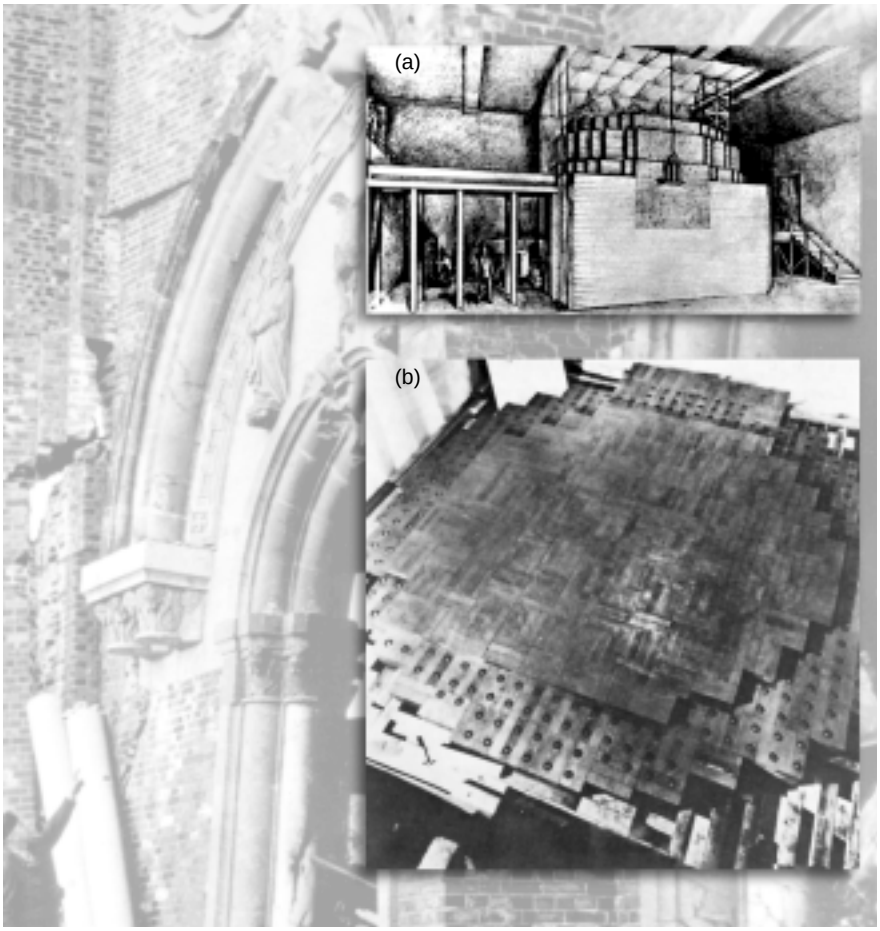


**(From the left): Donald Cooksey and Ernest O. Lawrence stand in front of the historic 60-in. cyclotron designed by Lawrence. The primary deuterons and secondary neutrons from the cyclotron were used to irradiate uranium samples. The uranium decayed to neptunium, which decayed to plutonium. Plutonium amounts made here in 1941 were too small to be seen but sufficient for initial analysis of chemical properties. The inset shows a 2.7-μg sample of plutonium oxide produced by Seaborg at the Washington University cyclotron. It is placed on a platinum weighing boat and has been magnified 40-fold.**

## Creating Plutonium

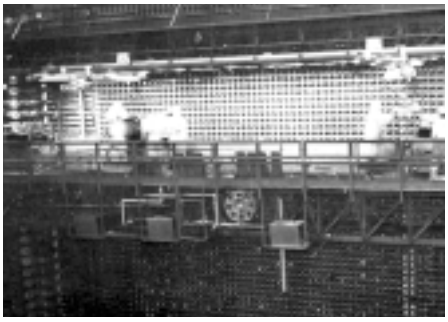
Until 1939, the worldwide attempt to identify transuranic elements had not been successful. That year, however, the American scientists Ed McMillan and Philip Abelson were the first to prove the existence of element 93. After bombarding uranium-238 with neutrons in the 60-inch cyclotron at the University of California at Berkeley, they used a clever sequence of measurements to prove that element 93 was growing in from the decay of uranium-239. They called it neptunium, after the planet Neptune. Neptunium was expected to decay to element 94 (plutonium), which would signal its presence through α-decay. Although minute quantities of the long-lived plutonium-239 must have been made, its radioactivity could not be detected. Shortly thereafter, McMillan's colleagues Seaborg, Kennedy, Wahl, and Segrè bombarded uranium-238 with deuterons and detected α-decay from the shorter-lived plutonium-238. Finally, on the night of 23 February 1941, Seaborg, Kennedy, and Wahl isolated a minute quantity of element 94, which a year later they officially named plutonium for the planet Pluto.

As early as 1939, American scientists, many of whom were refugees from Fascist regimes in Europe, had already started organizing a secret project that would take advantage of the newly discovered fission process for military purposes. Under the code name the Manhattan Project, it was officially established in August 1942. The Metallurgical Laboratory (Met Lab) at the University of Chicago became one of four Manhattan Project secret sites. It was tasked with producing plutonium-239 from uranium-238 in a reactor pile. The low neutron flux available at cyclotrons can produce only minute amounts of plutonium. A self-sustaining chain reaction in a reactor pile, on the other hand, would produce an extremely high neutron flux that could transform large amounts of uranium-238 into plutonium-239. But first, as told in the caption to the right, the fission chain reaction would have to be demonstrated.

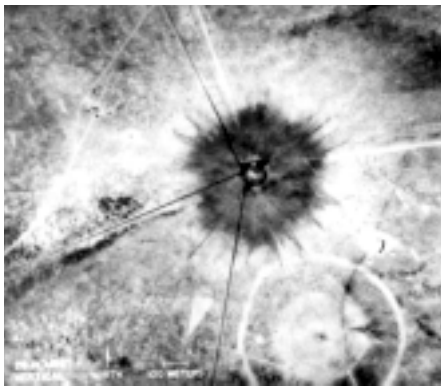



**Relying on their experience with building a uranium-graphite lattice at Columbia University, Fermi (above) and Szilard codesigned the Chicago pile—an artist's rendition is shown in (a). Fermi and coworkers built it. Stagg Field (background picture to the left), a Gothic-style monument to the university's football pride was chosen as the site for the chain-reacting pile. The work for building the pile was slow, laborious, and dirty. Made of uranium oxide and uranium metal components held by graphite bars in a lattice arrangement—see (b)—the pile contained 349,263 kg of graphite, 36,507 kg of uranium oxide, and 5,617 kg of uranium metal. On December 2, 1942, after the last cadmium control rod had been pulled out to the next required position to allow the neutron intensity to increase, the pile went critical. And so, what had begun as Szilard's scientific speculation turned into a wartime reality. Inevitably, plutonium must have been created in the pile, but the goal of this experiment was to prove the fission chain reaction.**

## Making the Atomic Bomb



**This is one of the first three plutonium production reactors at Hanford, Washington.**



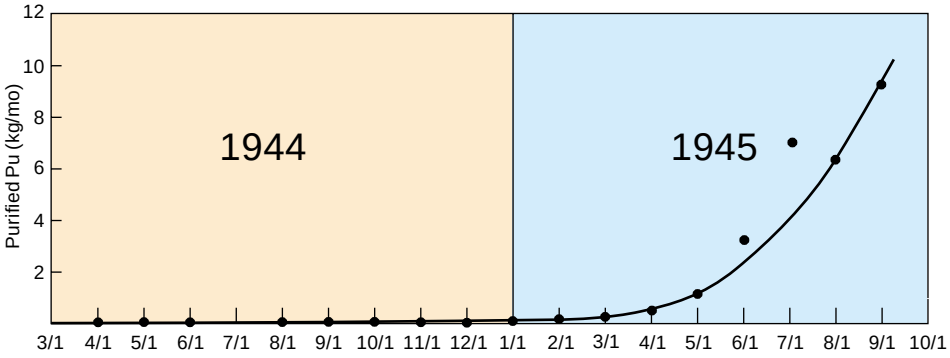
**During the Trinity Test, crater and heat effects scarred the desert at Ground Zero.**


**At Los Alamos, the world's first atomic bombs were designed and built.**

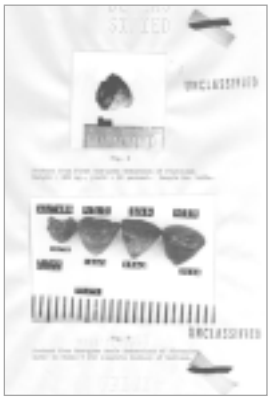
After Fermi and coworkers had demonstrated that natural uranium could sustain a fission chain reaction, the scaled-up uranium-graphite piles at Hanford, Washington, started producing plutonium-239. Hanford was the second site of the Manhattan Project. Eventually, there would be enough plutonium for three bombs—one to be tested at Trinity Site, another that would destroy Nagasaki, and a third that would end up not being used. Simultaneously, at Clinton (later to be called Oak Ridge), Tennessee, uranium-235 was being separated from uranium-238 for the bomb that would destroy Hiroshima. At this third secret site, there was also a plutonium-producing reactor. Los Alamos became the Manhattan Project's fourth site. In October 1942, following Gen. Groves's proposal, J. Robert Oppenheimer was named director of Project Y at Los Alamos, where the world's first atomic bombs were to be designed and built with material shipped from Clinton and Hanford.

Once the neutron-irradiated fuel elements in the reactor had been dissolved in an aqueous solution of mostly nitric acid, the plutonium nitrate slurry extracted would be shipped to Los Alamos and converted to plutonium chloride or fluoride. That compound would then be reduced to plutonium metal, whose material properties would have to be studied and understood. Ted Magel and Nick Dallas were the first to solve the plutonium reduction problem on a scale larger than a few micrograms. They obtained plutonium metal in a graphite centrifuge.

However, to scale up the reduction process, Dick Baker adapted the "bomb" reduction technique, which had been developed at Iowa State University for uranium, to the reduction of plutonium. He reduced plutonium halide to metal in a metallothermic reaction of the type $PuF_4 + 2Ca \rightarrow Pu + 2CaF_2$. The reaction took place inside a sealed steel container with a refractory liner in an inert-gas atmosphere at high temperatures. That is why it was nicknamed the bomb reduction technique. Plutonium




**During 1944, monthly deliveries of plutonium metal were minuscule (see graph above). From these quantities, Magel and Dallas produced a 50-mg button on March 9, 1944. By March 23, they had produced the first 1-g button (to the right, upper half of photo). Four more plutonium metal buttons made by Magel and Dallas during the spring of 1944 are pictured to the right (lower half of photo). Kilogram quantities did not arrive at Los Alamos until May 1945, when the Hanford reactor started making plutonium in large quantities. Ed Hammel eloquently tells the remarkable story (see page 48) of how the Manhattan Project pioneers learned just enough about this enormously complex metal to be able to craft it into a nuclear device in a very short time.**



metal was a precious commodity. In a letter to Gen. Groves, Oppenheimer pointed out that, by the end of August 1944, "we have received 51 grams of this material. The material has been used for approximately 2500 separate experiments. The overall loss per experiment has been about 1 percent." After each experiment, plutonium had to be recycled. In Oppenheimer's words, the "material we have dissipated" (that is, the 1 percent per experiment) "will be paid for many times over by the effectiveness with which we can deal with production lots when they become available."

At Los Alamos, work had initially centered on developing a gun-type bomb, which required high purification limits for plutonium. But in July 1944, plutonium produced in the Hanford reactor was found to contain not only the isotope 239 but also enough of the isotope 240 to create a problem. The heavier isotope would spontaneously fission and produce neutrons, which would trigger the bomb prematurely. The gun design was therefore promptly abandoned for the plutonium bomb, and the effort shifted toward an implosion design. Ironically, not only did the new work ease some impurity constraints, but it also required that metallurgists add an "impurity"—a few atomic percent gallium—to stabilize the cubic crystal form of plutonium that could be easily pressed into the final shape for the Trinity and Nagasaki bombs.

By July 1945, Los Alamos chemists and metallurgists had learned enough about the mysteries of plutonium to shape it into hemispheres and trust that a plutonium pit would hold together for the required number of weeks before the atomic bomb was to be used. Yet, the fact that the scientists decided to test the plutonium bomb and thus use almost half the quantity of material available at that time is a measure of how uncertain they were that the bomb would work as expected. The test at Trinity Site on July 16, 1945, was completely successful. On August 9, 1945, the plutonium bomb destroyed Nagasaki. ■

**Some of the leaders of the Los Alamos Project are pictured here. (Top row from the left): J. Robert Oppenheimer was the first director of Los Alamos (1942–1945). Physicist Seth Neddermeyer laid the foundations of the implosion program at Los Alamos. John von Neumann, a consultant to the Manhattan Project, was a great mathematician and physicist who invented the concept for a rapid implosion device. Cyril Smith was in charge of metallurgy. He suggested adding a second element to stabilize the δ-phase of plutonium. Hans Bethe led the Theoretical Division in predicting critical masses and explosive yields. (Bottom row from the left): Eric Jette, a physical chemist, headed the section in which plutonium reductions, remelting, alloying, and casting were conducted. George Kistiakowsky developed the explosives lenses that made the implosion concept work.**