

Plutonium Metal

In 1943, the Manhattan Project was pursuing two routes to a nuclear bomb, both dominated by the problem of acquiring the necessary nuclear materials. One route involved isolating the rare isotope uranium-235 from the abundant uranium-238 in sufficient quantity to build a weapon. The two isotopes are chemically identical and differ in mass by only about 1 per cent. Somehow the slightly lighter uranium atoms would have to be teased away from the heavier ones. Several separation techniques were under study—gaseous diffusion, electromagnetic separation, thermal diffusion, and the use of a centrifuge—but it was very uncertain whether any of them could produce the required kilogram quantities in a reasonable amount of time.



Ted Magel

The second route to the bomb involved plutonium-239, an isotope that physicists predicted would support a nuclear-fission chain reaction at least as well as uranium-235. But only insignificant traces of plutonium occur naturally on Earth. Large quantities would have to be made in a uranium-fueled nuclear reactor. When the reactor was operating, some of the neutrons from the chain reaction would be absorbed by uranium-238 to produce the unstable isotope, uranium-239. Almost immediately after being formed, uranium-239 would emit a beta particle (electron) to become a new element, neptunium-239, which would emit a second beta particle to become plutonium-239.



Nick Dallas

The total amount of man-made plutonium in existence in 1943 was the approximately 1.5 milligrams that had been made in accelerators. Not until February 1944 could gram quantities become available from the uranium reactor under construction at Clinton, Tennessee, and the needed kilogram quantities could not be expected to become available from the production reactors being built at Hanford, Washington until sometime in 1945.

In the meantime the metallurgists needed information as soon as possible on the bulk properties of the metallic form of plutonium including its melting point, its hardness, and especially its ductility and density. After all, they would be responsible for fabricating the metal into the shapes specified by the bomb designers. Solid pieces of pure plutonium metal large enough for metallurgical experiments—that is, not much less than a gram—were required to make the measurements.*

The need was so urgent that chemists at the University of Chicago's Met Lab and at Los Alamos began research in 1943 on chemical techniques to reduce plutonium compounds to pure metal. Compounds of other metals, particularly uranium, were used as stand-ins in the experiments.

Two young men at the Met Lab, Ted Magel and Nick Dallas, (see the plutonium-worker roundtable, "On the Front Lines") were the first to solve the plutonium metal reduction problem on a scale larger than a few micrograms. Since parallel work at Los Alamos was going poorly and gram quantities were soon expected

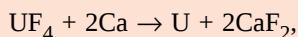
*The first unequivocal production of plutonium metal was carried out on November 6, 1943, at the Met Lab by H. L. Baumbach, S. Fried, P. L. Kirk and, R. S. Rosenfels (Manhattan Project Report CK-1143, December 1943). It was in the form of a few small globules of silvery metal weighing 1-3 micrograms each, scarcely large enough to permit any meaningful measurements of physical properties.

The First Gram

by Ed Hammel

from the Clinton reactor, Oppenheimer wrote a memo on January 18, 1944 requesting that Magel and Dallas come to Los Alamos. About a month after their arrival on February 3, 1944, they produced a shiny 20-milligram button of plutonium easily visible to the naked eye, and three weeks later they prepared a 520-milligram button of pure plutonium metal. These were the first amounts of plutonium metal produced at Los Alamos as well as the largest single buttons of the new element produced anywhere in the world. The technical story of their work is recounted here to illustrate the science and the intense atmosphere of the early plutonium metallurgy work and also to give them long overdue recognition for their contributions.

One basic reaction for reducing a plutonium or uranium salt to a metal is a metallothermic reaction. For uranium, a typical starting compound is uranium tetrafluoride and a typical reduction reaction is:

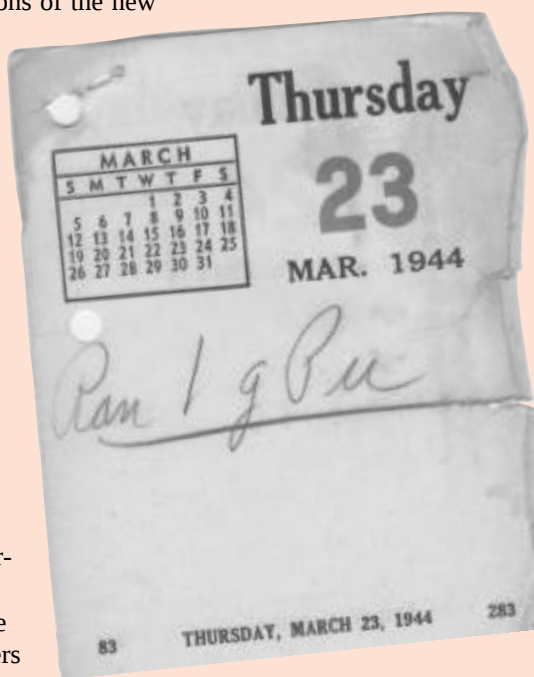


where calcium is the reducing agent. Heating the reagents to temperatures in the vicinity of 400 to 500 degrees centigrade initiates the reaction, which proceeds in the direction shown because fluorine has a much higher affinity for calcium than for uranium. At the same time, and for the same reason, the reaction gives off a great deal of heat—hence the name "metallothermic." Because of the high temperatures and pressures and the high reactivity of the reducing agent, the reaction was run inside a sealed metal container, which the Manhattan Project researchers called a "bomb." The bombs were lined with crucibles made of refractory materials such as metal oxides that would remain intact at the thousand-degree-centigrade temperatures produced in the reaction.

To maximize the yield and purity of the metal product, chemists had to optimize many parameters: the form of the initial uranium or plutonium salt, the reducing agent, the layering of the reagents in the bomb, their mesh sizes (the reagents were powdered), deviations from the stoichiometric proportions, the refractory material for the liner, the rate of heating, the optimum temperature required for initiating the reaction, the time spent at the maximum temperature reached, and finally, whether or not to add other materials that would simultaneously react, thereby producing additional heat (so-called boosters).

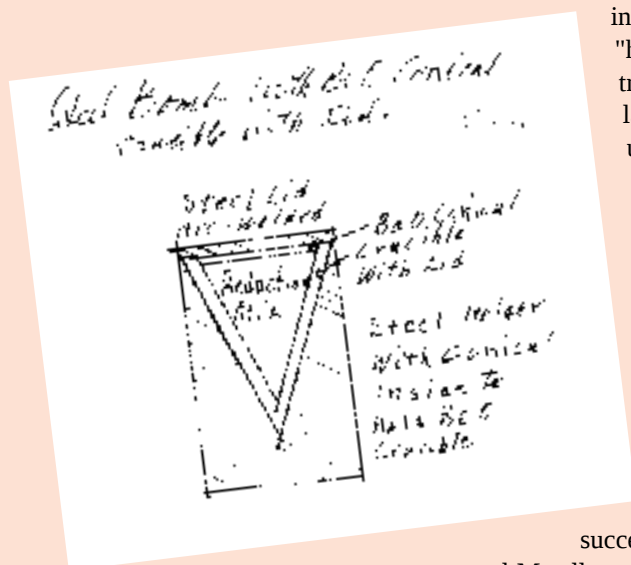
Yet another choice was how to separate the pure molten metal from the slag formed by the reaction products (CaF_2 in the above example). One way was to leave the bomb alone during the heating and let gravity do the work. Uranium and plutonium are far denser than the slag and should therefore naturally coalesce into a single molten globule of metal at the bottom of the crucible. Dick Baker's group at Los Alamos used this "stationary bomb" approach.

But the first batches of plutonium compounds would be very small indeed. The smaller the scale of the reaction, the worse the stationary-bomb approach could be



expected to work. A smaller bomb has more interior surface area in proportion to its volume than a larger bomb and is therefore more likely to lose a larger proportion of the reaction heat through the liner and bomb walls to the external environment. The reaction products might solidify before the new metal could flow through them and coalesce at the bottom of the liner.

Magel and Dallas, while working at the Met Lab in Chicago under Dr. John Chipman, recognized this problem and decided to assist the separation by performing the reduction inside a graphite centrifuge. The bomb was placed on its side in the centrifuge and rotated rapidly as it was being heated. The rotation rate could be adjusted to make the centrifugal force on the molten metal about 50 times larger than the force of gravity, enough to propel the molten metal outward to the tip of the cone-shaped interior of the refractory liner where it would cool into a consolidated mass. The components and operation of their "hot centrifuge" are shown in the box "The Magel-Dallas 'Hot Centrifuge' Technique," page 165. By the end of 1943 Magel and Dallas were using their new technique to make 1-gram buttons of pure uranium metal from uranium fluoride.



Meanwhile, the Los Alamos efforts in metal reduction, using stationary bombs and other methods, were floundering. Baker's group tried to prevent the slag from solidifying too quickly by using an iodine booster which not only adds heat to the reaction but also adds reaction products with low-melting points to the slag. Both effects keep the slag in the liquid state for a longer time. The iodine booster improved the results, but the reductions on the 1-gram scale still produced finely divided metal mixed with slag rather than a coherent metal slug. In January 1944,

Baker also tried the centrifuge approach, but his efforts were not successful. Consequently, J. W. Kennedy, the Leader of the Chemistry and Metallurgy Division, his Associate Director Cyril Smith, and eventually, as noted above, Oppenheimer himself requested Dr. Chipman to transfer Magel and Dallas to Los Alamos as quickly as possible.

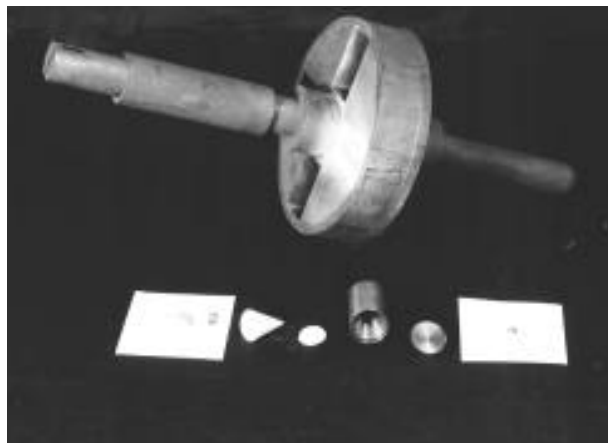
After Magel and Dallas arrived with their equipment, they immediately began performing centrifuge reductions of uranium. Reductions in a centrifuge worked best when the reducing agent was lithium and the liner was made of beryllium oxide. Magel and Dallas also concluded that an iodine booster had essentially no effect on reductions using lithium. Evidently the heat generated by the booster was of little value since the slag in lithium reactions had a sufficiently low melting point to permit plutonium and uranium metal to sink through it easily. Therefore, any further lowering of the melting point by adding iodine was unnecessary.

By March 2, an amount of fluoride (PuF_3) containing 50-milligrams of elemental plutonium was available for reduction. It had been prepared by Laboratory chemists from shipments of plutonium nitrate sent from the Clinton reactor. Magel and Dallas were given the material to reduce to plutonium metal. Probably with some reservations, they first followed the Los Alamos protocol of using calcium as the reducing agent and an iodine booster. The result was a grayish cokey mass containing no agglomerated plutonium. But on March 8, they tried again with another sample, this time using lithium (and iodine again). That experiment produced a shiny 20-milligram button of plutonium. Although the yield of 40 percent was disappointingly low, the result was the first plutonium metal made at Los Alamos and the first made anywhere in sufficient quantity to see without mag-

continued on page 166

The Magel-Dallas "Hot Centrifuge" Technique

The photograph below shows the components of Magel's and Dallas's apparatus for small-scale metal reduction of plutonium and uranium compounds. On the paper in front of the centrifuge rotor is a charge of metal halide (such as PuF_4) and a reducing agent. To the right of the paper is a cone-shaped crucible or liner made by powdering BeO , forming it in a mold, and firing it as clay is fired.



Magel and Dallas put the reducing agent into the crucible first and put the halide on top. They covered the crucible with a double lid (shown to the right of the crucible): the first layer made of either sintered NaCl , BaCl_2 , or LiF , was topped with one made of MgO . They put the crucible inside the cone-shaped interior of the cylindrical steel bomb, displaced the air inside the bomb with argon, covered the bomb with a steel lid, and sealed it shut by welding.

They mounted the bomb into one of the slots of the rotor and packed it tightly in place with more MgO . The rotor was about 15 centimeters in diameter and was made entirely of graphite to give it both strength

and heat resistance. It had four slots so that four reductions could be performed at once. (If the experimenters didn't have four charges, they put dummy bombs into the slots for balance.)

The photograph at right shows the centrifuge. The loaded rotor was placed inside a coil that was attached to a high-frequency electrical generator, and the shaft of the rotor was attached to a drill press through a slot-and-pin connector. When the generator was turned on, the coil would produce a rapidly alternating magnetic field, which would heat the rotor and bombs by induction. During the heating, the rotor would be spun by the drill press at 900 revolutions per minute, which made the force on the bomb's contents about 50 times that of gravity. Magel and Dallas found that the best procedure for plutonium reduction was to heat the spinning rotor and bombs to about 1,100 centigrade, which took somewhat less than five minutes, maintain that temperature for three minutes, and then turn off the generator and let the whole



thing cool but continue the rotation until the temperature reached 400-500 centigrade. When the bomb cooled to room temperature, they sawed it open at the top and removed its contents for examination.

The photograph at left show a longitudinal cross section of a bomb that was fired in the graphite centrifuge. In this particular specimen, the layer of slag is clearly seen on top of a button of uranium metal. The button is located in the tip of the crucible. The black spongy deposit clinging to the upper part of the cone is metal mixed with slag, which meant that the yield of pure metal was low in this particular reduction.

continued from page 164

nification. Many other 50-milligram runs were made with PuF_4 , PuF_3 , and PuCl_3 , as well as with other reducing agents. At this scale the results varied (about one third of them were successful).

During the three weeks following the initial success, Laboratory chemists prepared in succession two samples of PuF_4 , each containing a gram of plutonium. Much to the dismay of Magel and Dallas, Eric Jette, the leader of the Plutonium Metallurgy Group, and Cyril Smith decided to give the first 1-gram sample to Dick Baker for an attempt at reduction in the stationary bomb. The attempt produced only questionable microscopic droplets of plutonium dispersed in slag.



Ted Magel

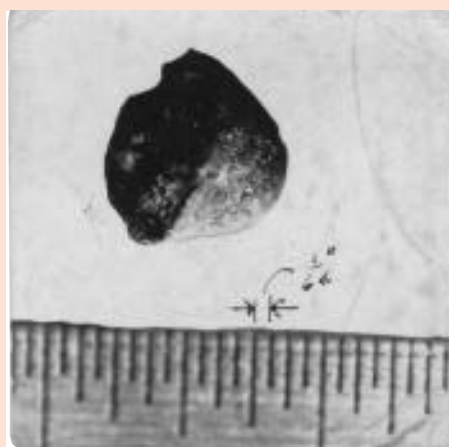


Figure 1. The first gram-scale piece of plutonium metal in history. It was made by Ted Magel and Nick Dallas at Los Alamos on the night of March 23, 1944 and weighed 520 milligrams.

When the second sample became available, Jette and Smith requested Magel and Dallas to attempt a centrifuge reduction on March 24th in the presence of a number of dignitaries. Magel decided on the 23rd to do the experiment without a crowd present. That night he and Dallas performed the reaction with lithium and no booster. When they cut open the bomb, they found a 520-milligram button of plutonium, shown in Figure 1. Again the yield was inexplicably low, but the metal was shiny and soft enough to cut with pliers; both qualities indicate purity. The button was immediately used for crucial metallurgical and chemical studies. From April to early June, Magel and Dallas made eight more buttons on the one-gram scale, all of which were successful, and four of which are shown in Figure 2. In total, they performed about 300 centrifuge reductions between February and June; twenty-five of them were plutonium reductions.

During the course of their work, both Magel and Dallas experienced various accidental exposures to plutonium, which later qualified them for membership in the so-called UPPU club, Wright Langham's follow-up study of wartime plutonium workers who received intakes of plutonium (see "On the Front Lines").

In the summer of 1944, Magel and Dallas started small-scale work on purifying plutonium, especially from light-element contaminants. They set up high-vacuum, high-temperature remelting systems to evaporate residual light element impurities from the reduced buttons of plutonium. Light-element impurities are a problem because they absorb alpha particles from the decay of plutonium and emit neutrons. The neutrons can then initiate a chain reaction in the plutonium before two subcritical assemblies have been able to come together to form the planned supercritical mass. The removal of light-element impurities was therefore considered crucial for minimizing the neutron background and preventing a preinitiation of the gun-type plutonium weapon.

During that summer, Baker made a systematic study of small-scale, stationary-bomb reactions. He found that PuCl_3 was a better starting material than PuF_4 and then went on to develop reliable techniques using this halide for producing gram-scale buttons of plutonium. Because stationary bombs were much more convenient than centrifuges and did not require lithium as a reductant nor the use of

beryllium oxide crucibles (both of which contributed high levels of light-element impurities to the resulting plutonium), Baker's method turned out to be preferable for production of plutonium in quantities greater than one gram.

The availability of gram-scale quantities of plutonium permitted the Los Alamos metallurgists to attack in a multi-faceted and coherent way the so-called variable density and crystal-structure problems. Puzzling variations in density and crystal structure had been seen in different metal specimens since the time of plutonium's first production on the microgram scale at the Met Lab, and the possibility of allotropism had been raised as early as February 1944 by R. Mooney and W. H. Zachariasen at the Met Lab. Nevertheless, at Los Alamos, the results of specific attempts to settle this issue were ambiguous until June 1944. Research did finally show that plutonium has more complex allotropic behavior than any other known metal, and this property made the task of producing the necessary shapes for weapons even more difficult.

Toward the end of the summer of 1944, the light-element impurity problem suddenly became irrelevant: It was discovered that reactor-produced plutonium from Hanford would contain significant amounts of plutonium-240. That isotope undergoes spontaneous fission and therefore would add much more to the neutron background than the light elements ever could. Since there was no practical way to remove it, the project had to abandon the gun-type weapon and replace it with an implosion device in which the speed of the assembly would eliminate the possibility of neutron-induced preinitiation. It also meant that Magel and Dallas were no longer needed to solve light-element purification problems, and they decided to leave Los Alamos and join Dr. Chipman, who had moved to MIT. There they helped make large crucibles of various refractory materials for use by Baker's reduction section and Ed Hammel's remelting, alloying, and casting section. Thus their work for the Manhattan Project continued even after they left Los Alamos. ■

Further Reading

Richard D. Baker, Siegfried S. Hecker, and Delbert R. Harbour. 1983. Plutonium: a wartime nightmare but a metallurgist's dream. *Los Alamos Science* 7: 142-151.

E. F. Hammel. *Recollections of Plutonium Metallurgy Work in D-Building, 1943-1945*. To be published.

Lillian Hoddeson, Paul W. Henriksen, Roger A. Meade, and Catherine Westfall. 1993. *Critical Assembly A Technical History of Los Alamos during the Oppenheimer Years, 1943-1945*. Cambridge University Press.

C. A. Levine and G. T. Seaborg. 1951. The occurrence of plutonium in nature. *Physics Review* 73: 3278.

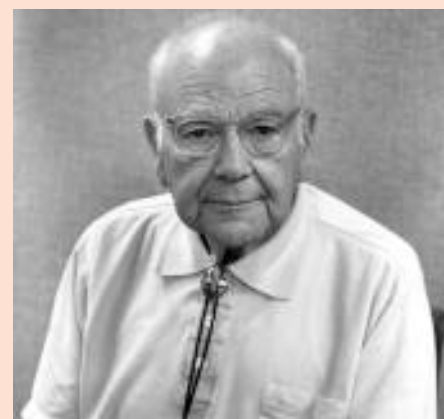
R. L. Kathren, J. B. Gough, and G. T. Benefiel. 1994. *The Plutonium Story: The Journals of Professor Glenn T. Seaborg, 1939 - 1946*. Columbus: Battelle Press.

J. W. Kennedy and the C.M. Smith & CM Division Staff. 1944-1945. Monthly progress report for the chemistry and metallurgy division. Los Alamos Scientific Laboratory reports, 1944-1945.

T. T. Magel, D. B. Loeb, and N. Dallas. 1944. The preparation of plutonium metal on the one-gram scale by means of the graphite centrifuge. Los Alamos Scientific Laboratory report, LA-124 (August 1944).



Figure 2. Four more plutonium metal buttons made by Magel and Dallas during the spring of 1944.



Edward F. Hammel joined the Laboratory in 1944 as a section leader in the Chemistry and Metallurgy Research Division, where his principal responsibility was remelting, alloying, and casting plutonium metal. In 1945 he was appointed group leader of the Metal Physics Group, which was responsible for determining the physical properties of plutonium. In 1948 Ed became group leader of the Low Temperature Physics and Cryoengineering Group and was responsible for organizing a program to study helium-3. During that year Ed and his collaborators were the first to liquefy helium-3 and to test its properties at low temperatures. They searched for superfluid behavior down to 0.7 kelvin, a remarkable feat for the times. Their search was unsuccessful because helium-3 becomes a superfluid at an unexpectedly low temperature of less than 3 millikelvins. From 1970 to his retirement in 1979, Ed held management positions in various energy related projects including the study of superconducting transmission lines and energy storage. In 1955 Ed was awarded the American Chemical Society gold medal for his work on helium-3. He received his A.B. in chemistry from Dartmouth College and his Ph.D. in physical chemistry from Princeton University.