Plutonium

A Wartime Nightmare but a Metallurgist's Dream

by Richard D. Baker, Siegfried S. Hecker, and Delbert R. Harbur

In 1942 the theoretical outline for an atomic bomb was clear: compress enough fissionable material long enough to properly ignite a chain reaction. Construction of an actual weapon, however, required translation of "fissionable material" into real pieces of plutonium or uranium metal. These metals had to be free of impurities that would adversely affect the neutron flux during the chain reaction and yet be fabricable enough that precise shapes could be formed. Whether this would even be possible with plutonium was not then known, however, because plutonium was a new, manmade element and the metal had not been produced.

Accounts of the Manhattan Project have neglected (for security reasons, initially) the important metallurgical work that preceded fabrication of these materials into integral parts of real weapons. For example, the Smyth Report* devotes one short paragraph to the wartime work of the entire Chemistry and Metallurgy Division at Los Alamos—a division that in 1945 numbered 400 scientists and technicians. Our article will attempt to fill part of this gap for plutonium by highlighting key developments of the wartime research and will continue with some of the exciting research that has occurred since the war.

Research from 1943 to 1946

The Los Alamos work on plutonium and enriched uranium, the so-called special nuclear materials, was extensive, covering a variety of research problems ranging from purification of material received from reactors to the prevention of oxidation of the final product. Further, because many chemical processes and physics experiments required very pure materials, such as gold, beryllium oxide, graphite, and many plastics, considerable general materials research was also carried out.

Much of the chemistry and metallurgy of uranium was already known from the production of uranium metal for the uranium-graphite reactor pile at Chicago in 1942. The work remaining on enriched uranium included preparation of high-purity metal, fabrication of components, and recycling of residues. However, the most challenging research and development was carried out on the new element plutonium.

Table I gives the important dates in the early history of plutonium and shows the short time—four years—that elapsed between its discovery and its use in the first atomic device at Trinity. The discovery occurred, as predicted by nuclear theory, when uranium was bombarded with 16-million-electron-volt deuterons in the cyclotron at Berkeley. Within about a month it was shown that plutonium-239 fissioned when bombarded with slow neutrons, and a decision was made to build large reactors at Hanford for the production of plutonium—this before the uranium-graphite pile at Chicago had demonstrated that a sustained and controlled chain reaction was even possible! That demonstration soon followed, proving that large quantities of plutonium could be produced, although no plutonium was extracted from the Chicago reactor.

At this point only microgram amounts of plutonium had been separated from the targets used in the cyclotrons. Remarkably, the basic chemistry of plutonium was worked out at Berkeley and Chicago on this microgram scale, and it formed the basis for the scale-up—by a factor of a billion—needed for plants that would eventually separate plutonium from spent reactor fuel. At the same time the first micrograms of the metal were produced at Chicago by the

reduction of fluorides, and preliminary metallurgical properties were determined. However, the influence of impurities on such tiny samples distorted many of the results; for example, the melting point of plutonium was first thought to be about 1800 degrees Celsius, considerably above the true melting point of 641 degrees. Ultimately, the properties of plutonium were found to be incredibly sensitive to impurities.

It had been agreed that Los Alamos would not work on batches of plutonium of less than about 1 gram, and the microgram-scale work continued at Chicago. Finally, in early 1944 Los Alamos received plutonium nitrate samples containing half-gram amounts of the element from the "Clinton" reactor and pilot extraction plant at Oak Ridge. Later, larger amounts were received from the production facility at Hanford.

The plutonium nitrate arrived in relatively impure form, and techniques and equipment had to be developed for a number of processes, including purification, preparation of plutonium tetrafluoride and other compounds, reduction to metal, and metal fabrication. Also, because plutonium was in very short supply, it was imperative to develop processes to recycle all residues.

Initially, the purity requirements for the metal were very stringent because some elements, if present, would emit neutrons upon absorbing alpha particles from the radioactive plutonium, These extra neutrons were undesirable in the gun-type plutonium weapon then envisioned: they would initiate a chain reaction before the material had properly assembled into its supercritical configuration, and this "pre-initiation" would decrease the explosive force of the weapon. The purity requirement for certain elements was a few parts per million and for some, less than one part per million. As a result, all the materials used in the preparation of the plutonium metal, everything from the process chemicals to the containers, had to be of very high purity. This necessitated development work on many materials, including an

TABLE I
EARLY HISTORY OF PLUTONIUM

Plutonium discovered	February 23,1941
Neutron-induced fission of plutonium-239, proved	March 25, 1941
Decision reached for large, full-scale plutonium production	December 6, 1941
First controlled fission chain reaction achieved, proving method for full-scale production of plutonium	December 2, 1942
Preparation of plutonium metal from microgram quantities produced with cyclotron	November, 1943
Gram quantities of plutonium nitrate from experimental reactor received at Los Alamos	March, 1944
Plutonium nitrate from production reactor received at Los Alamos	mid 1944
Plutonium weapon demonstrated with Trinity test	July 15, 1945

extensive effort to obtain pure and nonreactive refractories to contain molten plutonium. The high purity requirements also necessitated the development of new methods for analysis of all materials, including plutonium.
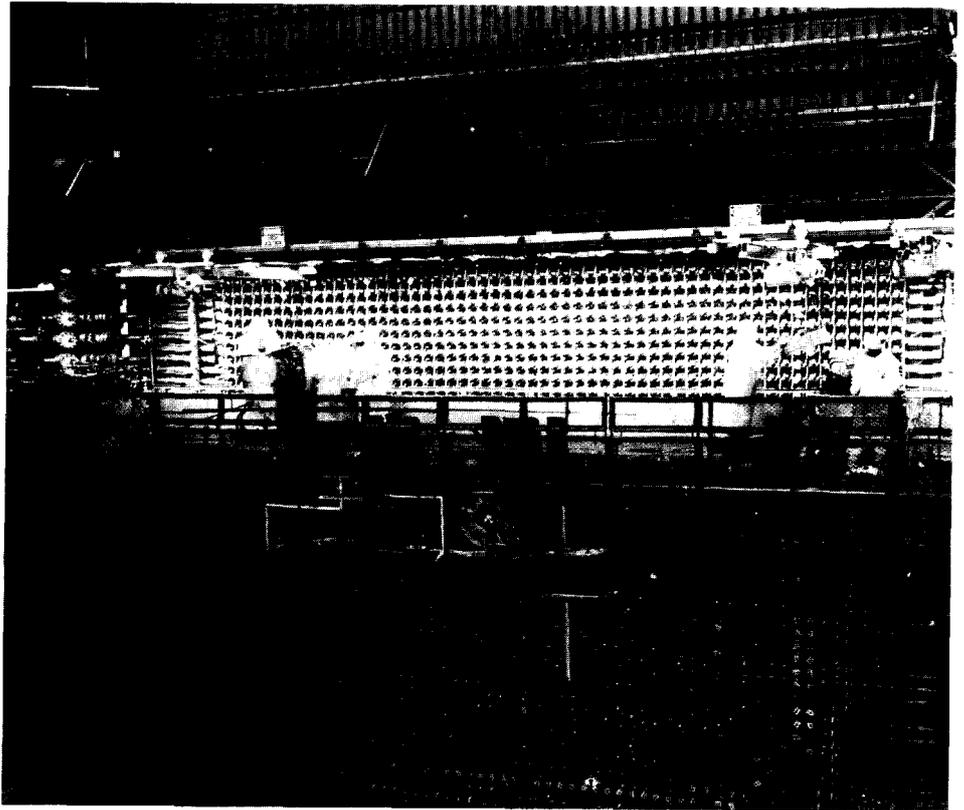
The potential health problem associated with the handling of plutonium had been recognized at Chicago, and work on the subject began with receipt of the first small amounts of plutonium. A Health Group was formed to monitor plutonium work areas, and, within the Chemistry and Metallurgy Division itself, committees were established to design suitable radiation detectors and apparatus for handling plutonium and to formulate safe handling procedures. Because alpha counters then lacked either sensitivity or portability and were in short supply, oiled filter paper was swiped over surfaces to pick up possible stray bits of plutonium and then measured at stationary counters. Similar procedures were used to detect suspected contamination of hands and nostrils. The air-conditioning system in the plutonium laboratory (D Building), which was installed initially to help maintain high purity by filtering out dust, ultimately served the more important function of confining the plutonium. The building was equipped with hoods with minimal ventilation and with the forerunner of the modern glove box—plywood "dry boxes." The successful handling of large quantities of plutonium without serious problems was at that time an outstanding achievement.
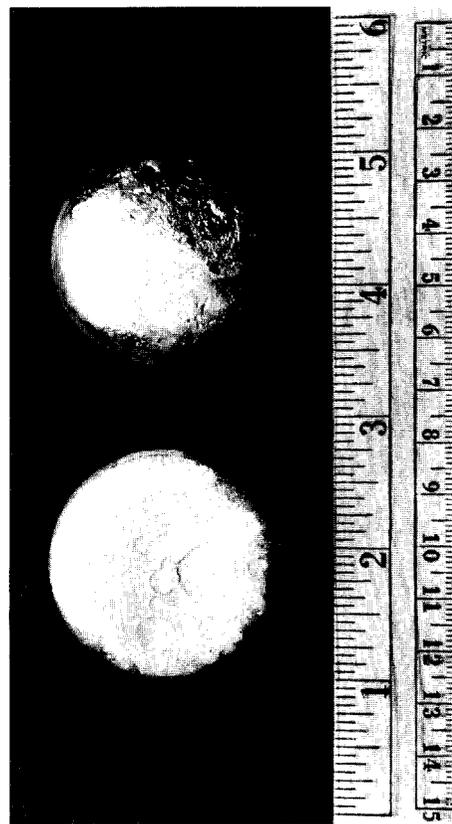
Two early discs of plutonium metal after reduction from the tetrafluoride. Plutonium generally arrived at the Laboratory from the Hanford reactors in the form of a relatively impure nitrate solution. Techniques were developed at Los Alamos for purification, preparation of various compounds, reduction to the metal, and metal fabrication.

At first plutonium metal was prepared at Los Alamos either by lithium reduction of plutonium tetrafluoride in a centrifuge, the metal settling out as the closed reaction vessel rotated, or by the electrolysis of fused salts containing plutonium. Soon, however, calcium reduction of the tetrafluoride was perfected. The vessel used to contain this reaction was called a stationary "bomb" because the reaction was highly exothermic and the metal product settled out in the closed, nonrotating vessel simply by gravity. This technique became the preferred method and was used to prepare plutonium for almost all metallurgical studies and for the nuclear devices.
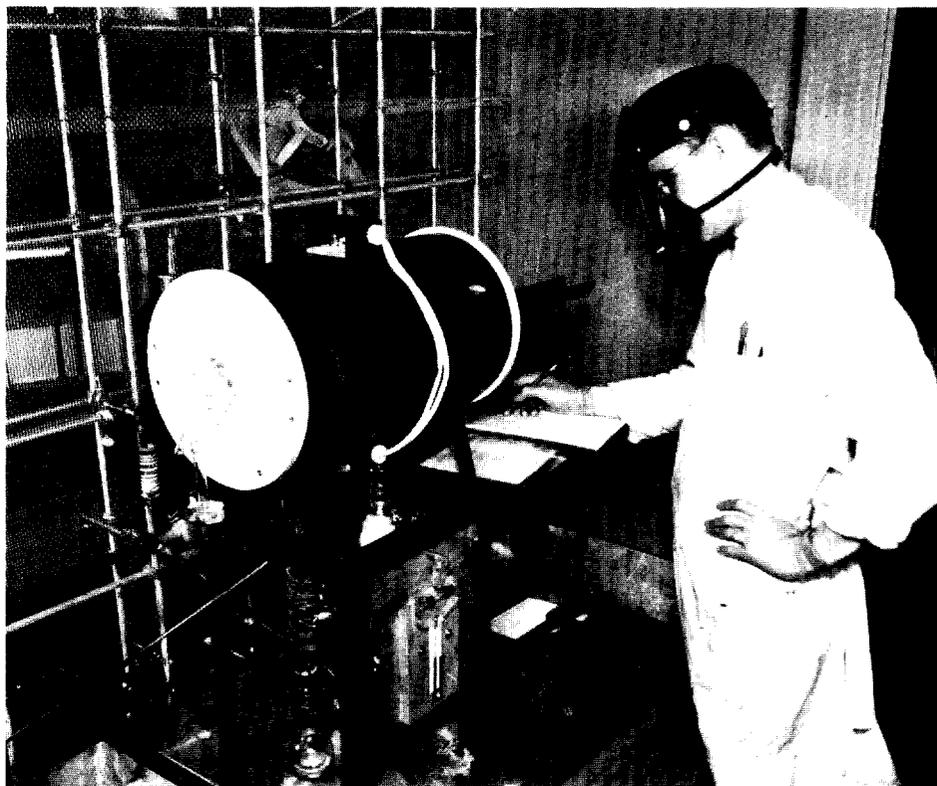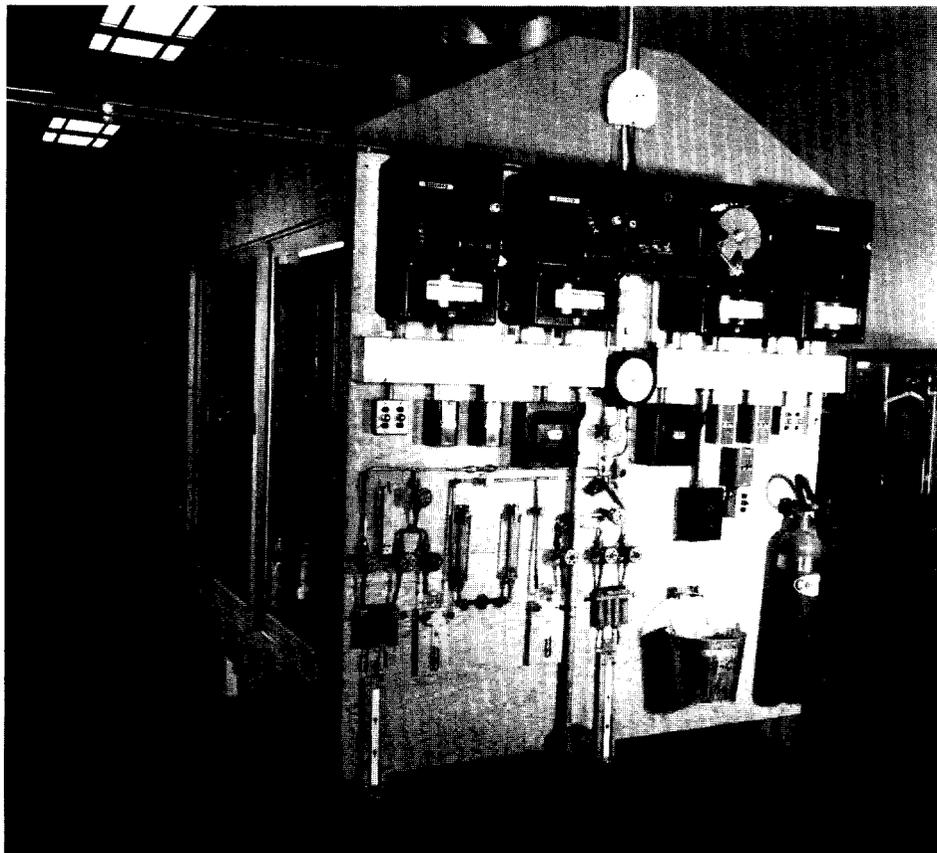
The microgram metallurgy at Chicago had provided values for the density of the metal that clustered about either 16 or 20 grams per cubic centimeter. This bimodal spread, due surely in part to impurities, nevertheless pointed toward interesting metallurgy by hinting that the element had more than one phase. Working with larger amounts, Los Alamos refined these measurements and by the middle of 1944 had discovered that plutonium was a nightmare: no less than five allotropic phases existed between room temperature and the melting point. Unfortunately, the room-temperature alpha (a) phase was brittle, and the metal experienced a large volume change when heated and then cracked upon cooling. These properties made fabrication very difficult, and there was not enough time for detailed fabrication development on the a-phase material. It was thought likely that another phase would be malleable and easily shaped; the problem was how to stabilize such a phase at room temperature. It was then discovered that alloying plutonium with small amounts of aluminum stabilized the delta (8) phase, which was, in fact, malleable. However, aluminum was one of those elements that emitted neutrons upon absorbing alpha particles and so would exacerbate the pre-initiation problem. Beneath aluminum in the periodic table was gallium, which did not

The first production reactor at Hanford with workmen loading uranium into its honeycomb face. (Photo courtesy of the Department of Energy.)

D Building, the air-conditioned wartime plutonium laboratory, bristling with separate external vents.

undergo this type of nuclear reaction. Plutonium-gallium alloys were found to be stable in the δ phase and could be hot-pressed into the required hemispheres. Thus the problem of fabrication was solved. To avoid oxidation of the metal and to contain the radioactivity, the pieces were ultimately coated with nickel.

In July 1944 it was discovered that the plutonium-239 generated in the high-neutron-flux production reactors at Hanford contained too much plutonium-240. Plutonium-240 was undesirable because it had a much higher spontaneous fission rate than plutonium-239 and emitted far too many unwanted neutrons. As a result, pre-initiation in the gun weapon could not be avoided without the difficult task of separating these isotopes. Instead an intense effort was mounted to develop an implosion weapon in which pre-initiation could be avoided because of its higher assembly velocities. This turn of events allowed the purity requirements for the metal to be somewhat relaxed, simplifying many of the process operations. The necessary pieces of plutonium were then fabricated in time to construct the Trinity and Nagasaki devices.

The extreme press of time during the war allowed for the immediate problems of fabrication, stability, and oxidation protection to be solved only empirically. A comprehensive program of basic research on this most fascinating element had to wait until after the war. In Table II we summarize the properties of plutonium metal known in 1945.

Postwar Research and Development

As the war ended, construction began at DP West site on a new, more permanent facility for the plutonium effort. This activity reflected the government's decision to increase production of nuclear warheads and, thus. to scale up all processes associated with the fabrication of plutonium metal parts.

Because the plywood dry boxes of old D Building posed a tire hazard, they were

The hydrofluorination of plutonium. The upper photograph shows the chemical hoods in D Building used for this process, which converted the oxide to the tetrafluoride. Four furnace controllers are at the top of the panel with one controller open showing the temperature program cut into its rotating disc. Note the bucket of calcium oxide to be used for treatment of hydrogen fluoride burns. The lower photograph shows one of the hydrofluorination furnaces inside the hood.

TABLE II

PROPERTIES OF PLUTONIUM METAL KNOWN IN 1945'

| Phase	Temperature Range of Stability ('W)	Crystal Structure	Density (g/cm')	Average Linear Expansion Coefficient (per 'C)	Electrical Resistivity (@cm)	Temperature Coefficient of Resistivity (@cm per "C)
Alpha	Below 117	Orthorhombic (?)	19.8	55×10^{-6}	150 at 25°C	-29.7×10^{-4}
Beta	117 to 200	Unknown (complex)	17.8	35×10^{-6}	110 at 200°C	- o
Gamma	200 to 300	Unknown (complex)	Unknown	36×10^{-6}	110 at 300°C	˜ 0
Delta	300 to 475	Face-centered cubic	16.0	-21×10^{-6}	102 at 400°C	$+1.5 \times 10^{-4}$
Epsilon	415 to 637	Body-centered cubic	16.4	4×10^{-6}	120 at 500°C	- 0
Liquid	Above 637±5					

ᵃFrom Cyril Stanley Smith, Journal of Nuclear Materials 100,3-10 (1981).

replaced with stainless-steel glove boxes. To better contain the plutonium, the glove boxes were equipped with elaborate ventilation-filtration systems devised to keep the atmosphere within each glove box at a lower pressure than the surrounding air so that any leak in the system would not release plutonium to the room. In addition, the breathing air in the laboratories was filtered and changed several times each hour.

Since all of the processes for purification, preparation, and fabrication of the metal and for recycling of the residues of plutonium and enriched uranium were developed at Los Alamos during the war, there was no other place for the production of nuclear warheads. It was decided that Los Alamos should not continue in production but should concentrate on research and development. The transfer of all the special processes to be used in the new production plants was a major postwar undertaking. Plutonium processes were transferred to Hanford, Savannah River, and Rocky Flats. The enriched uranium processes were transferred to Oak Ridge.

The work at DP West thus settled into a program of basic research and development, and major advances were made in the fledgling plutonium technologies of vacuum casting, metal working, machining, electrorefining, and aqueous processing of scrap. Several plutonium reactor fuels, both metallic and ceramic, and the plutonium-238 heat sources for thermoelectric generators for space and other missions had their beginnings at DP West.

In 1978 the plutonium activities at DP West were moved to the newly completed Los Alamos Plutonium Facility, the most modern and complete plutonium research
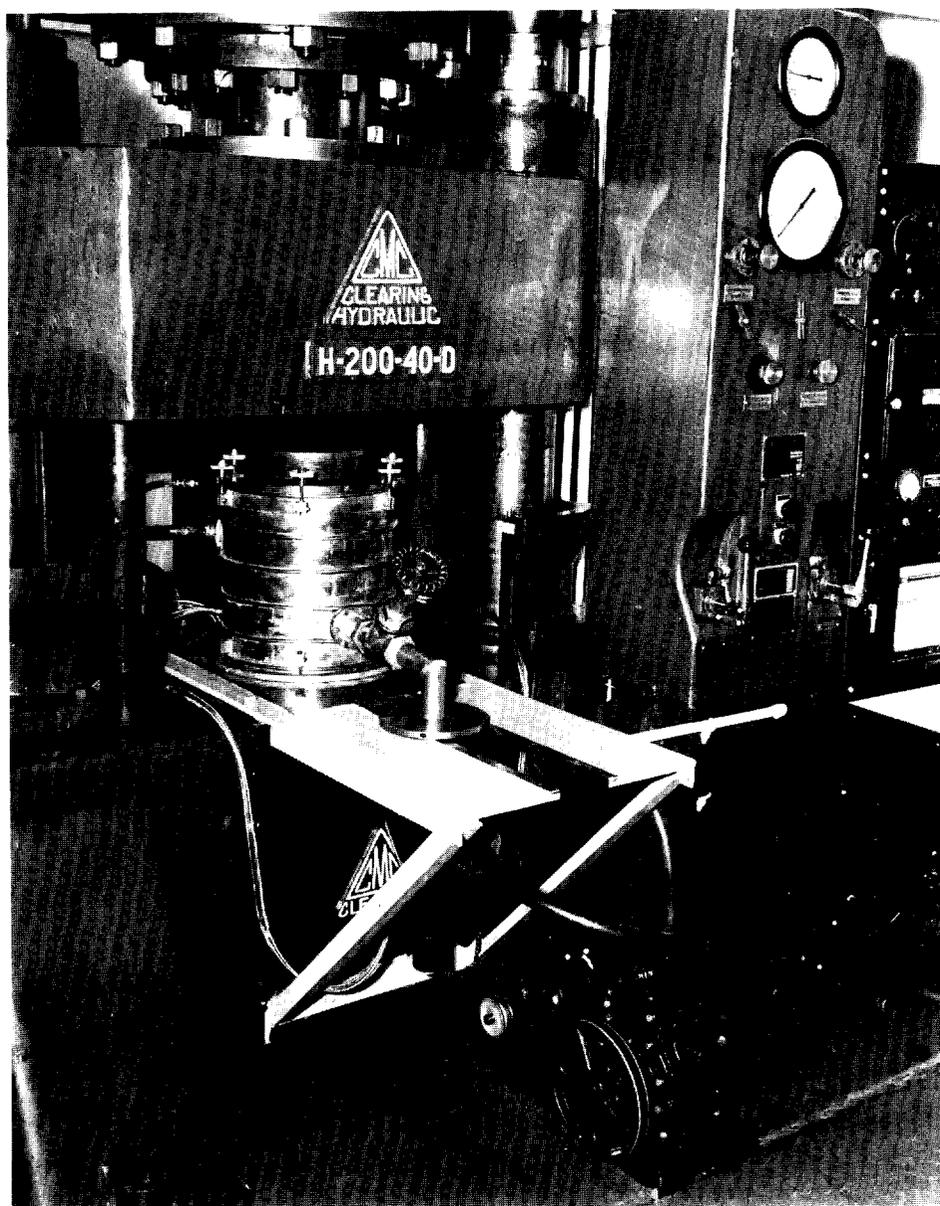
Presses used during the war to hot press plutonium metal into the shapes required for the Trinity and Nagasaki devices.
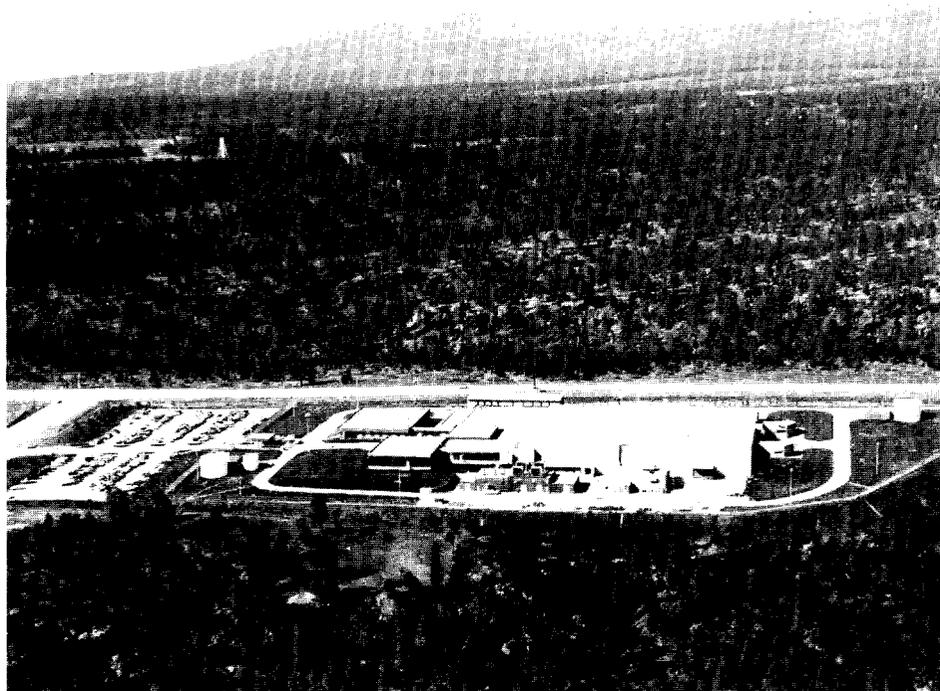
The current Los Alamos plutonium research and development facility. Note, in contrast to the wartime laboratory, the absence of external ventilation on the large research building to the right (upper photograph). The facility uses the most modern equipment, including a computerized plutonium accountability system (lower photograph).

and development center in the country. It incorporates state-of-the-art designs and equipment for the safe containment of plutonium and the protection of workers during all credible accidents or natural disasters, including earthquakes and tornado-force winds.

After the war the continued improvement in process chemistry and applied metallurgy of plutonium came about through a better understanding of its basic properties. Aqueous processes were developed for separating plutonium from virtually every element in the periodic table. The wartime "bomb" reduction process was augmented with other pyrochemical processes such as direct reduction of the oxide and electrorefining. These processes not only yielded a purer product but also minimized the amount of plutonium-bearing residues and the associated radiation exposure of personnel.

Plutonium casting was first carried out with ceramic crucibles and molds because they were known to be compatible with molten plutonium. The discovery that slightly oxidized tantalum was quite unreactive with molten plutonium led to the development of reusable foundry hardware. Also, the development of several ceramic coating processes, based on either calcium fluoride or the stable oxides of zirconium or yttrium, permitted the use of easily machined graphite molds. It was discovered that microcracks resulting from the multiple phase changes that occur as the metal cools and freezes could almost be eliminated by casting the pure metal into chilled aluminum molds, a process that virtually by-passes most of the intermediate phase transformations.

The development of new plutonium alloys for both reactor and weapons use proceeded hand in hand with the determination of the equilibrium phase diagrams of plutonium with most other elements, and the associated complex crystal structures of phases and compounds. Early on we realized that we were dealing with alloys that were metastable in many environments. Thus, it became
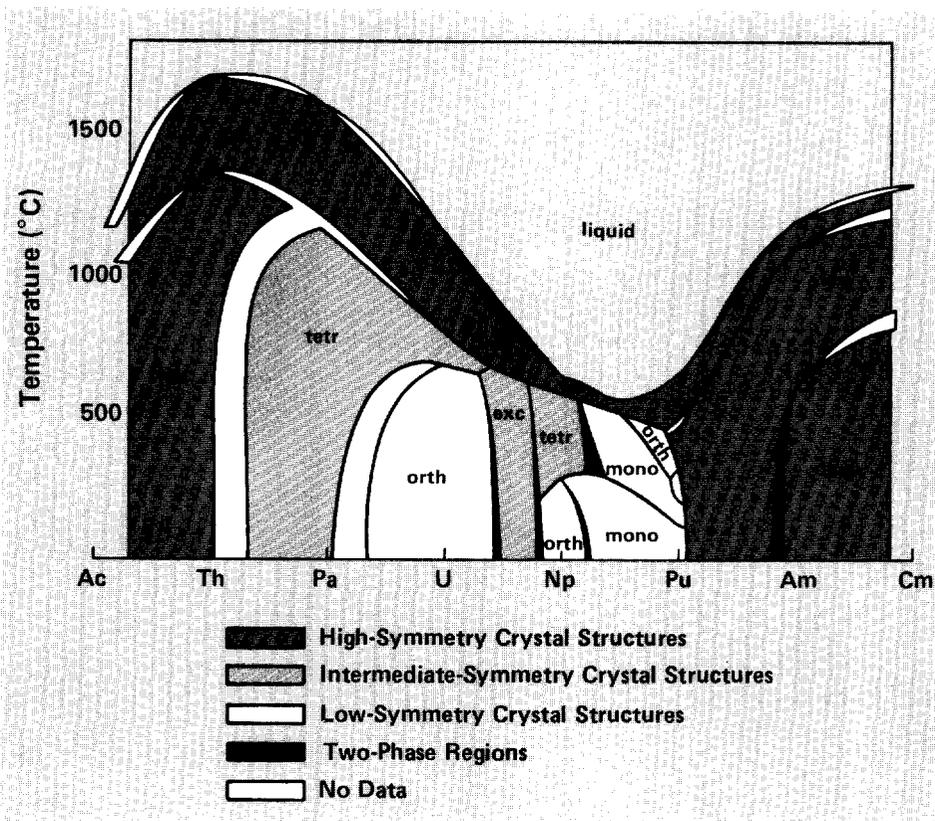
Fig. 1. The actinides and the configuration of their outermost electrons. This series of elements is characterized by the filling of 5 f electron orbitals. (The 5 f orbitals of thorium lie above the Fermi level but are sufficiently close to induce some f character in the electrons near the Fermi level. Thorium is therefore regarded as the first actinide.) The properties of the 5 f electrons, particularly their participation in atomic bonding, are the key to the unusual properties of these elements.

imperative to understand microsegregation of alloying elements and phase stability during all processing steps. Complex heat treatments were developed to homogenize the alloys or to further stabilize the proper phases.

In 1954 the purity of plutonium was increased sufficiently that a new phase, delta prime (δ'), was discovered between the already known δ and epsilon (ε) phases. While this new phase proved to be inconsequential to applied plutonium technology, its discovery certainly showed the necessity of using high-purity, well-characterized plutonium in basic research. A seventh allotrope of plutonium, the zeta (ζ) phase, was discovered many years later in 1970 during careful studies of the equilibrium pressure-temperature phase diagram of plutonium. This phase exists only at high temperature over a limited pressure range and has such a complex crystal structure that it still today

has not been positively identified.

Current Understanding of Plutonium

During the postwar period major activities in plutonium metallurgy in the United States were centered at Argonne National Laboratory, Hanford, Lawrence Livermore Laboratory, Rocky Flats, and of course, Los Alamos. Important contributions were also made at the Atomic Weapons Research Establishment's facility at Aldermaston in the United Kingdom and at Centre d'Etudes Nucleaires de Fontenay-aux-Roses in France. Except at Los Alamos much of the research activity was terminated or severely curtailed in the 1970s. However, we are currently seeing a revival of plutonium research at several locations.

In spite of many years of concentrated research and great strides in the practical aspects of plutonium metallurgy, this field is

still in its infancy. A comparison with steel supports this perspective. The metallurgy of steel has been studied intensely in many countries for more than 100 years, yet important discoveries are still commonplace. Metallurgists have learned to manipulate the three allotropic phases of iron to tailor the properties of steel to specific applications. The six allotropic phases of plutonium and its much wider range of crystal structures and atomic volumes provide many more possibilities—and pitfalls.

The focus of the postwar research was to study all aspects of the behavior of this new element and, thus, be prepared for all of its peculiarities. In contrast, the focus of the past decade has been to exploit the complexities of plutonium. Much of the effort has been devoted to alloy development and the determination of structural properties and has resulted in several new alloys with interesting properties. Many of these results re-

Fig. 2. *A connected phase diagram of binary alloys of the actinides (prepared by E. A. Kmetko and J.L. Smith) shows the preponderance of low-symmetry crystal structures, the large number of phase changes, and the depression @melting points in the vicinity of plutonium. The crystal structures are body-centered cubic (bcc),face-centered cubic (fcc), tetragonal (tetr), orthorhombic (orth), exotic cubic (exe), monoclinic (mono), and double hexagonal close-packed (dhcp).*

main classified.

In the past decade we have also turned our attention toward a more fundamental understanding of plutonium on the atomic level. This effort has opened a most fascinating chapter in solid-state physics—the electronic properties of the actinides, the seventh period in the periodic table. Interest in the actinides had stemmed primarily from their special nuclear properties. Yet, it is the properties of the electrons (not the nuclei) that govern all chemical and structural behavior. The actinides are characterized, as shown in Fig. 1, by the progressive filling of *5f* electron orbitals. It is the participation of these *5f* electrons in atomic bonding that leads to the peculiar and complex behavior of actinide metals and alloys.

Although details of the *5f* bonding in the actinides are still being contested, it is gener-

ally agreed that the *5f* electrons in the early actinides (through plutonium) are not fully localized and thus participate in bonding. The *5f* bonding increases to a maximum at plutonium and vanishes as the electrons become localized near americium. The effects of *5f* bonding on the behavior of the lighter actinides are dramatic, and most dramatic for plutonium. We will highlight here only three of the most important effects. These are demonstrated in Fig. 2, a connected binary alloy phase diagram of the elements in the actinide series. First, as one moves from actinium to plutonium, a change from highly symmetric cubic to low-symmetry crystal structures occurs. Second, the number of allotropic phases increases. Finally, the melting points decrease dramatically.

How can these effects be explained? To

begin, the wave functions off electrons (like those of *p* electrons and unlike those ofs and *d* electrons) have odd symmetry. This property is not compatible with symmetric cubic crystal structures but rather favors the low-symmetry crystal structures and, thus, the stability of monoclinic and orthorhombic phases. Only with increased temperature and lattice vibrations is the *f* character sufficiently overcome in plutonium to permit cubic crystal structures. Beyond plutonium the localization (nonbonding) of the *f elec-*trons leads to a return of more typical metallic behavior. Also, because the *f elec-*trons are just on the verge of becoming localized and magnetic, small changes in temperature, pressure, or alloying have dramatic effects on phase stability and properties. Hence, allotropy is promoted. Finally, the *f* electrons bond quite easily in the liquid phase because its less rigid structure increases rotational freedom. This ease of bonding promotes the stability of the liquid (or, equivalently, limits the stability range of the solid) and lowers the melting points.
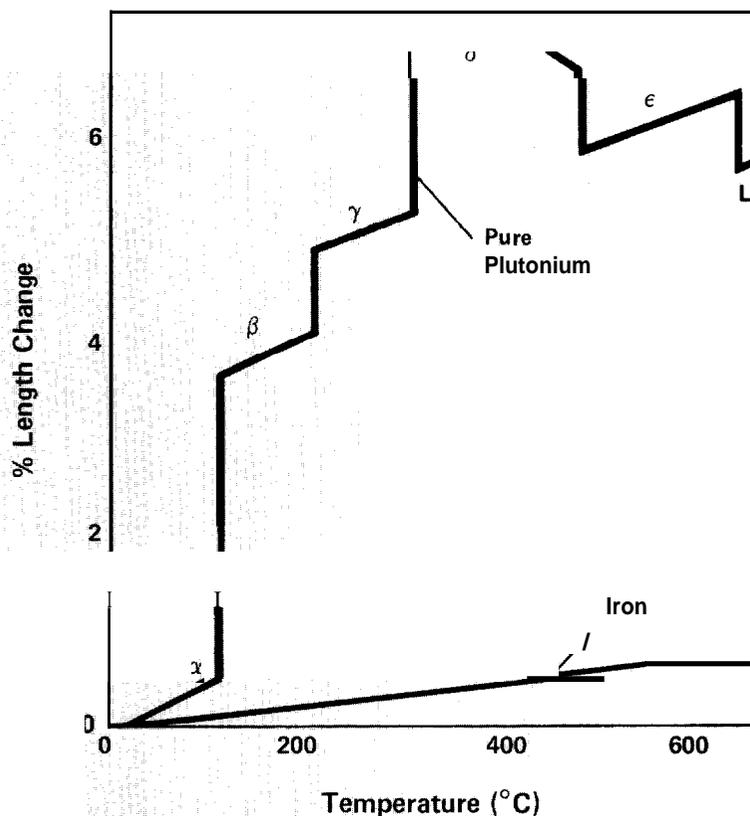
We see that the *f* electrons are the cause of many of plutonium's peculiarities and complexities, which have important practical consequences. Its low melting point and limited solid stability are particularly important because, as a liquid, plutonium is extremely reactive and corrosive and hence difficult to contain. Liquid plutonium also has the greatest known surface tension and viscosity among metals because off bonding. A less obvious consequence arises from the fact that most rate processes in solids depend upon homologous temperature, that is, temperature relative to the absolute melting point. Hence, diffusion and other thermally activated processes are quite rapid at room and slightly elevated temperatures.

The most significant consequence of plutonium's large number of phases is thermal instability of the solid. This property is best illustrated by a plot of length change during heating. Figure 3 compares the behavior of

plutonium with that of iron. Most phase transitions in plutonium are accompanied by large length and thus volume changes. Such volume changes are difficult to accommodate in solids at relatively low temperatures without loss of physical integrity. In addition, plutonium's α, β, γ, and ε phases have very large thermal expansion coefficients. For example, the thermal expansion coefficient of the a phase is about five times greater than that of iron. Therefore special compatibility problems arise wherever plutonium is in contact with other metals. Figure 3 illustrates two exceptionally peculiar properties of plutonium: the negative thermal expansion coefficients of the δ and δ' phases and the contraction upon melting, which results from increased *f*- electron bonding in the liquid phase.

The crystal structures and the corresponding densities are also listed in Fig. 3. Note that the three structures that are stable at temperatures closest to room temperature are of low symmetry. The cubic structures that are typical of most metals appear only at high temperatures where the 5 *f*-electron bonding is overwhelmed. The low-symmetry structures (especially the a phase) exhibit very directional bonding. The a-phase monoclinic structure is essentially covalently bonded. Its unit cell contains 16 atoms with 8 different bond lengths ranging from 2.57 to 3.71 angstroms. Consequently, most of its physical properties are also very directional. In addition, the *a* phase is a poor conductor and is highly compressible.

The low symmetry and nearly covalent nature of bonding in the a phase greatly affect its mechanical properties, which more nearly resemble those of covalently bonded minerals than those of metals. The a phase is strong and brittle because the low symmetry controls the nature and motion of defects. The face-centered cubic δ phase, on the other hand. behaves much like a normal metal. In fact, the δ phase possesses the strength and malleability of aluminum. One must remember, however, that at δ-phase temperatures

Fig. 3, *A plot of percentage length change as a function of temperature illustrates the dramatic changes that occur with each of plutonium's phase changes. The more sedate behavior of iron is shown for comparison.*

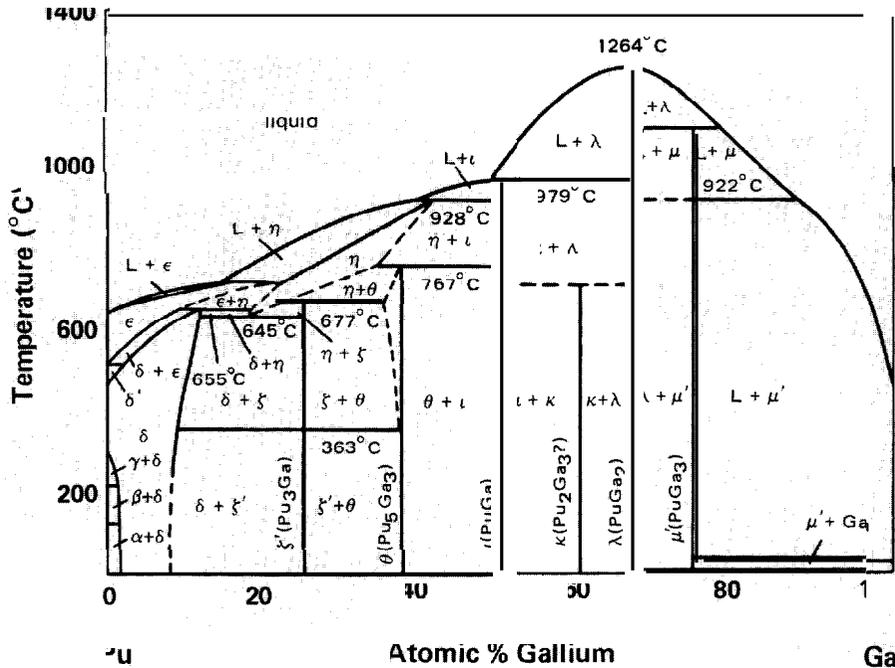|	Crystal Structure	Density (g/cm^3)
α	Simple Monoclinic	19.86
β	Body-Centered Monoclinic	17.70
γ	Face-Centered Orthorhombic	17.14
δ	Face-Centered Cubic	15.92
δ'	Body-Centered Tetragonal	16.00
ε	Body-Centered Cubic	16.51
L	Liquid	16.65

Fig. 4. The plutonium-gallium phase diagram serves as an example of the complexities that occur when plutonium is alloyed. The region of concern during the war was the lower left corner where the malleable δ phase extends down to room temperature at gallium concentrations below 10 per cent.

the *5f* bonding is essentially gone.

The δ phase can be stabilized to room temperature by alloying. As we pointed out earlier, this fact was already recognized during the war and led to the use of gallium to stabilize this phase. It is now recognized that most trivalent solutes, such as gallium, aluminum, cerium, americium, iridium, and scandium, stabilize the δ phase. Figure 4 shows the plutonium-gallium equilibrium phase diagram as determined at Los Alamos in the postwar era. Note the expanded field of the δ phase on the left and the enormous complexities that result from alloying plutonium. The δ phase in plutonium alloys behaves much like a normal metal and has several advantages over the a phase, including excellent ductility (fabricability), a much larger range of thermal stability, and a lower thermal expansion coefficient (nearly zero for most alloys).

So far we have not mentioned the effects of pressure. As one might expect, hydrostatic pressure tends to collapse the low-density crystal phases. Hence, in pure plutonium the δ phase disappears at pressures of less than 1 kilobar. Here is where the seventh allotrope of plutonium, the ζ phase, appears before giving way to α or β phases at high pressures. Only moderate pressures are required to collapse the alloyed δ phase to higher density phases. When dealing with alloys at high pressures, we are faced with the problem of what happens to the solute atoms, since they are generally insoluble in the α and β phases. This topic and the question of the response of alloys under nonequilibrium cooling conditions typify the fascinating world of nonequilibrium phase transformations in plutonium, which is beyond the scope of this article.

Plutonium is without question the most complex and interesting of all metals. More so than in any other metal, a fundamental understanding of its metallurgical behavior must be rooted in an understanding of electronic structure. We have highlighted the peculiarity and complexity of plutonium resulting from the *5f* electrons. The complexity, hidden until after the war, makes the accomplishments of the metallurgists and chemists during the Manhattan Project even more remarkable. ∎

Further Reading

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Criticality
The Fine Line of Control

by Hugh C. Paxton

I n the early days of the Manhattan Project, no one had experience in handling the large quantity of fissionable material needed to build a weapon because, quite simply, it hadn't been made yet. That was soon to change as Oak Ridge began to separate small amounts of uranium-235 and to prepare for processing kilogram amounts. This large a quantity posed the danger of accidental criticality—setting off a fission chain reaction—as scientists on Project Y well knew. But, as Feynman relates,* the demands for secrecy meant that this information was not widespread:

. . . The higher people [at Oak Ridge] knew they were separating uranium, but they didn't know how powerful the bomb was, or exactly how it worked or anything. The people underneath didn't know at all what they were doing. . . . Segre insisted they'd never get the assays right, and the whole thing would go up in smoke. So he finally went down [from Los Alamos] to see what they were doing, and as he was walking through he saw them wheeling a tank carboy of water, green water—which is uranium nitrate solution.
He says, "Uh, you're going to handle it like that when it's purified too? Is that what you're going to do?"
They said, "Sure-why not?"
"Won't it explode?" he says.
. . . The Army had realized how much stuff we needed to make a bomb—20 kilograms or whatever it was—and they realized that this much

material, purified, would never be in the plant, so there was no danger. But they did not know that the neutrons were enormously more effective when they are slowed down in water. And so in water it takes less than a tenth—no, a hundredth—as much material to make a reaction that makes radioactivity. It kills people around and so on. So, it was *very* dangerous, and they had not paid any attention to the safety at all.

Thereafter, criticality safety became an important focus at Oak Ridge and Los Alamos, but when I arrived in Los Alamos, late in 1948, the state of the art was still fairly primitive. I was asked to head the critical assemblies group in Pajarito Canyon. With this assignment I became the Laboratory's immediate expert on nuclear criticality safety, although I had no pertinent background. Now, from the vantage point of today's abundant criticality information, I realize I should have been dismayed. But then there existed only a few-page summary of experimental data from Los Alamos, a couple of reports giving Oak Ridge measurements, and no reliable calculations (excellent methods were being developed but remained unconfirmed). This amount of information was certainly not overwhelming.

I had to learn rapidly the techniques for avoiding accidental criticality in processing, fabricating, storing, and transporting fissile materials. (At that time we had plutonium

and uranium enriched in uranium-235; uranium-233 was added later.) These techniques were meant to control any variable that affects criticality, such as mass, dimensions, density, and concentration in solution. Criticality also is influenced by nearby objects that act as neutron reflectors, returning neutrons that otherwise would be lost to the fissile material. As mentioned in Feynman's tale, neutron moderation, especially by intermixing the fissile material with hydrogenous material, such as water, is particularly important to criticality. Hydrogen is very effective at moderating (decreasing the energy of) fission neutrons by scattering, and these less energetic neutrons are much more effective at initiating further fissions.

In the late 1940s it usually was necessary to compensate for insufficient data by introducing large factors of safety. This situation was acceptable for operations in processing plants because production rates of fissile material were still low. Weapons, however, were another matter. Design subtlety had not yet reduced their content of fissile
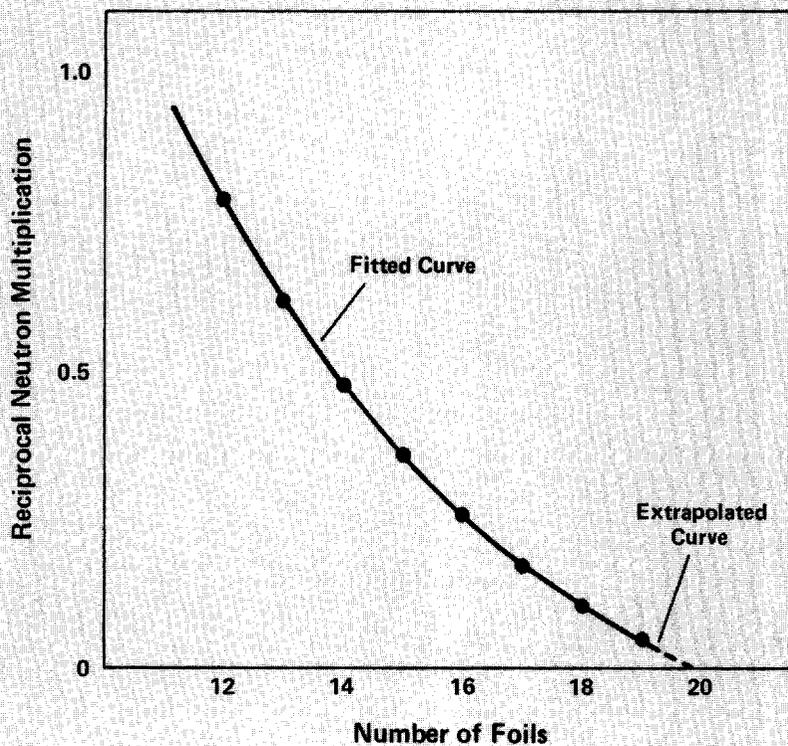
Fig. 1. The data points above were obtained from neutron count-rate measurements on a "sandwich" containing, alternately, slabs of Lucite (a neutron moderator) and foils of enriched uranium. As the sandwich is allowed to approach the critical state by adding uranium-Lucite layers one by one, the neutron count rate rises rapidly. Plotted above are reciprocal neutron multiplication values (ratios of count rate for the original sandwich to count rates as each layer is added) versus number of foils. Extrapolation of the fitted curve to zero establishes the critical number offoils.

material. and many weapons contained as much fissile material as could be introduced safely. Excessive safety factors could not be tolerated, and special measurements by the critical assemblies group were required for reasonably, but not excessively. safe designs.

Because the Pajarito group was capable and smoothly functioning when I arrived, it performed well while I learned from it about the conduct of critical experiments and their relation to weapon design. I learned about neutron-multiplication measurements with so-called long counters that responded uniformly to neutrons with a wide range of energy. I learned how multiplication, represented by neutron count rate, increases as the mass of plutonium or enriched uranium is increased and tends toward infinity as criticality is approached. The critical mass could be established. however, without actually reaching it. A plot of reciprocal neutron multiplication versus fissile mass (or other variable used to approach criticality) extrapolates to zero at criticality (Fig. 1) and

thus establishes the critical mass by means of subcritical measurements.

To appreciate the significance of criticality, let us first note that a nuclear explosion is the result of a runaway fission chain reaction in which neutrons from fission produce an increased number of fissions, which in turn produce an increased number of neutrons, and so on. The term supercritical describes this state. In the critical state the fission rate and the number of neutrons remain steady. A sphere of the most dense phase of plutonium is just critical at a mass of 10.5 kilograms if bare, but the critical mass drops to about 6 kilograms if the plutonium is surrounded by a natural uranium reflector that returns neutrons to the plutonium. A more spectacular decrease, to a critical mass less than 0.6 kilogram, may occur in a uniform mixture of plutonium and water surrounded by a water reflector. This decrease is a result of neutron moderation by hydrogen.

Strictly, the steady-state fission chain re-

action occurs at *delayed* criticality. That is, it depends upon the delayed neutrons emitted during decay of the fission products as well as the prompt neutrons emitted during fission. At steady state the delayed neutrons constitute less than 1 per cent of the total neutron population. The addition of a small amount of fissile material (1 per cent for plutonium and 2 per cent for uranium) to a critical mass produces *prompt* criticality. That is, delayed neutrons no longer influence the chain reaction, and fission power increases so rapidly that it is uncontrollable. If the increment between delayed and prompt criticality is termed 100 cents, prompt criticality may be exceeded a few cents without damaging a uranium metal system, but the intense radiation pulse would endanger a person nearby. At an excess of 10 cents, damage to the system would begin. The damage would become severe at a 15-cent excess, and the runaway chain reaction would lead to an explosion at an excess of 50 cents or less.

In weapon design it is important to know the delayed critical state because it must be exceeded during detonation but must not be attained during assembly, storage, and transportation, As plutonium and enriched uranium began to accumulate at Los Alamos. priority was attached to experiments that determined critical conditions by extrapolation from subcritical measurements. Before 1946 these urgent experiments had been conducted manually by persons who remained beside the experiment. Typically, the experiments involved the stepwise addition of reflector material to a fissile core with a multiplication measurement at each step.

Twice, criticality was attained accidentally during these experiments. The first incident, in 1945, resulted in fatal radiation injury to Harry Daghlian. It occurred when a heavy uranium block slipped from Daghlian's hand onto a near-critical assembly consisting of a plutonium ball and a natural uranium reflector. The damaging radiation consisted of neutrons and gamma rays from the intense

fission chain reaction. Manipulation by hand continued until Louis Slotin suffered a similar fate about a year later. Again something slipped—in this case a screwdriver being used to lower a beryllium reflector shell toward the same plutonium ball involved in the earlier accident. The shell dropped instead of being held short of criticality. In neither accident was equipment damaged. Manual control was outlawed after the second accident, and the facility in Pajarito Canyon was rushed to completion.

At the Pajarito facility experiments are carried out by remote control from a control room one-quarter mile away. (Other critical assembly facilities of the time used massive shielding, rather than distance. for personnel protection.) The building in which the experiments were carried out (Fig. 2) was called the kiva, a term borrowed from the Pueblo Indians and referring to their ceremonial chambers. The facility became available for subcritical measurements in 1947 and for critical operation a year later. In subsequent years two other kivas were added. Separate control rooms for the three kivas are located in a central building.

Fig. 2. (a) The original kiva, photographed from an Indian cave in the nearby wall of Pajarito Canyon, and (b) its control room, which was first housed in an existing shack. The racks contain controls for gradually separating and bringing together the parts of a critical assembly, displays of the long-counter responses that indicate neutron multiplication, radiation monitors that trigger a scram (automatic disassembly) if the level should become higher than intended, and a television screen for viewing the assembly. From left to right, Vernal Josephson, Roger Paine, Lester Woodward, and Hugh Karr. Paine and Woodward were military personnel who contributed invaluably to our critical experiments.
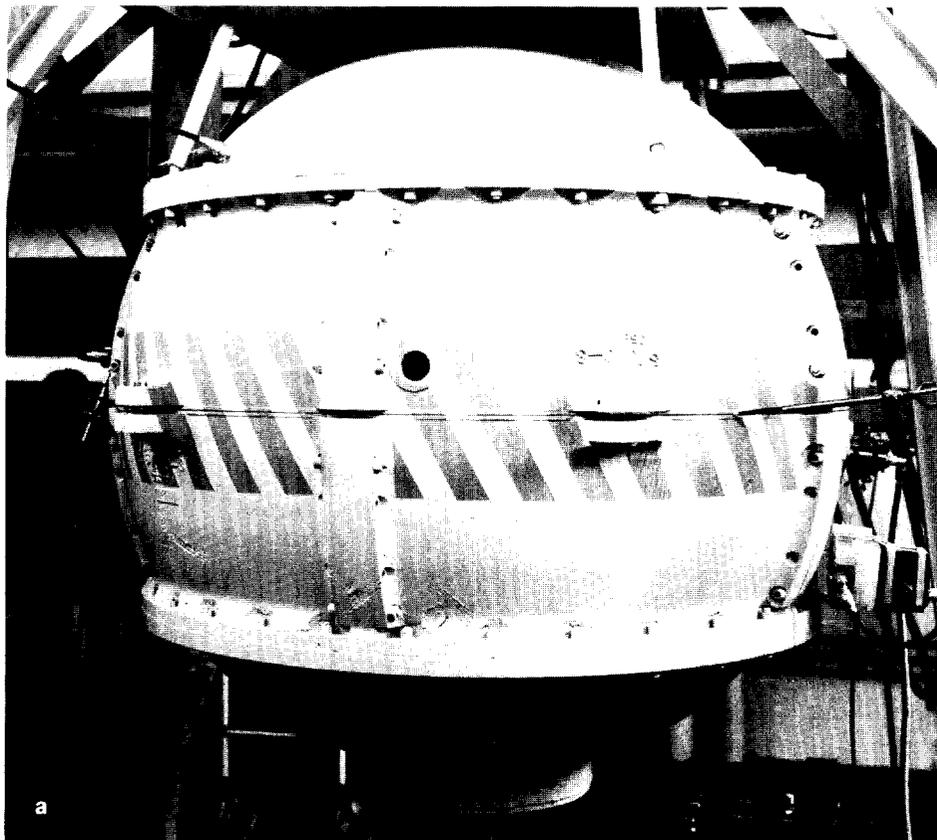
The Bomb Mockup (Fig. 3), the first remotely controlled machine for bringing together two parts of a near-critical assembly, was similar in size to Fat Man, the Nagasaki weapon. The two hemispheres of the Bomb Mockup were separated, and a core of fissile material was placed in a recess in the lower hemisphere. After personnel retreated to the control room, remotely actuated controls brought the two hemispheres together and instruments recorded the neutron count rate. The process was repeated with increasing masses of fissile material until extrapolation to criticality was acceptable.

These subcritical neutron-multiplication measurements with the Bomb Mockup demonstrated safe loading of implosion-weapon components, confirmed the intended reactivity (deviation from the critical state) of production cores, and provided safety guidance for new implosion-weapon designs. To

Fig. 3. (a) The Bomb Mockup, a simulation of an implosion weapon in Kiva I. After a fissile core was placed in a cavity in the lower hemisphere, neutron count rates were measured as the two hemispheres were gradually brought together by remote control. Before personnel could re-enter the kiva, the two halves of the mockup had to be separated. Neutron-multiplication measurements in this mockup established subcritical limits for weapons of more advanced design than the Nagasaki weapon. (b) An adult version of mud pies was an essential preliminary to experiments with the Bomb Mockup. Surrounding the fissile core in the mockup was a material that simulated the neutron reflection and moderation properties of high explosives. The photograph shows the material being mixed and tamped into parts of the mockup. Identifiable are William Wenner holding the bucket, Gustave Linenberger in the center foreground, and James Roberts standing above.
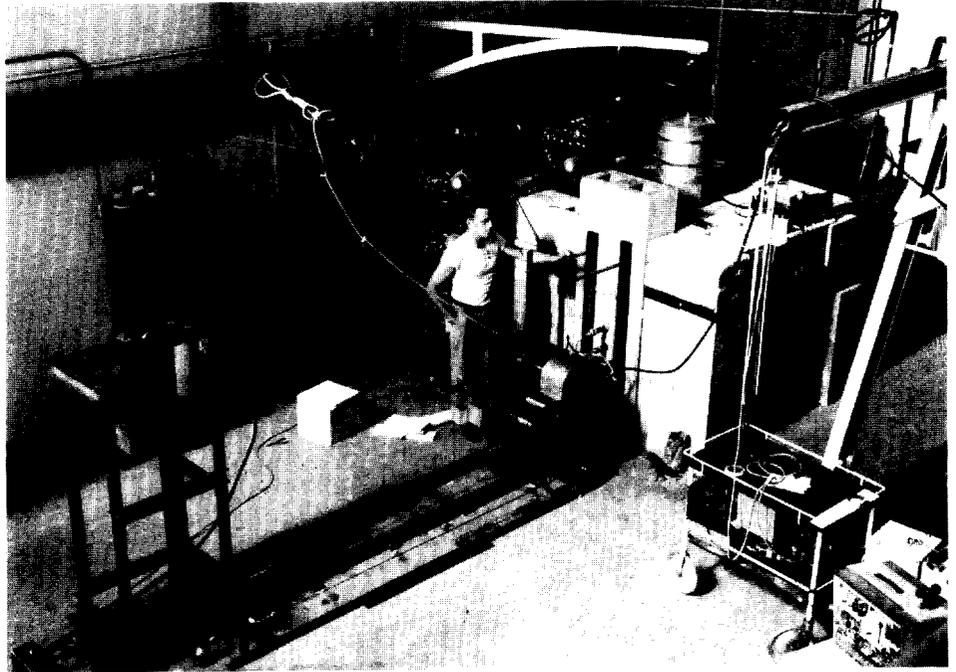
Fig. 4. A concrete vault in Kiva I for criticality tests on weapon cores arranged as they might be during storage. As many as 27 cores (the country's entire stockpile) were supported on two lightweight frames similar to jungle gyms (within the vault in this photograph and shown schematically in Fig. 5). Each frame was mounted on a track and could be moved in and out of the vault by remote control. A portion of the vault wall-a "door"—moved with each frame. Raemer Schreiber is shown beside the one visible drive mechanism and track (the other drive mechanism and track are hidden behind the vault). The number of cores on the frames was increased a few at a time, and neutron multiplication was measured as the frames were moved into the vault and the doors closed. Stringent security measures were maintained during these experiments, including a special contingent of military guards, machine gun emplacements on the walls of Pajarito Canyon, and a requirement that all personnel wear distinctive jackets while moving between buildings. Operations were conducted around the clock to minimize the time the stockpile was removed from its usual location.

supplement experiments with the Bomb Mockup, flooding tests confirmed subcriticality should a core fall accidentally into a body of water. The flooding tests were carried out in a temporary setup consisting of a tank that was filled by remote control and had a large dump valve as a safety device. Other safety tests involved cores surrounded by paraffin, concrete, and natural uranium.

Information to guide the safe storage of weapon components was obtained in 1947 with another temporary setup (Fig. 4). It consisted of a concrete vault of adjustable size that was closed by remote control and opened automatically when the radiation near the vault exceeded a safe level. Multiplication measurements on arrays of implosion-weapon cores or capsules as they were built up stepwise within the vault (Fig. 5) provided the required guidance. Some years later these measurements were supplemented by neutron-multiplication tests on arrays of cores in storage arrangements simulated at Rocky Flats and, finally, by other measurements at an actual storage site.

Only once did we use a live weapon for measurements at Pajarito Site. The purpose was to determine how well our high-explosive mockup material simulated the neutron reflection and moderation properties of real high explosive. The tests were performed on Sunday so that few people would be at risk if something should go wrong. There was one scary moment when the capsule assembly stuck as it was being inserted by remote control into the high explosive. (Neutron multiplication was so low that this difficulty was corrected easily by hand.) On comparing notes with those who brought the high-explosive assembly, we learned that they breathed a sigh of relief when they left our dangerous fissile material behind, just as we did when they departed with their dangerous high explosive.

At no other time was explosive permitted at our facility. Over the years mockup material was improved to simulate precisely the elemental composition of high explosive. Thus it became prudent to test the material to be sure that the simulation was not so good that it, too, might be explosive.

Criticality considerations for gun-type weapons differed from those for implosion weapons because of the requirement that the total mass of fissile material become supercritical as soon as its subcritical components were engaged. Experiments on a new design first established the total fissile mass needed for the weapon. Then, the measured separation of components at criticality provided a basis for choosing a safe initial separation. Other tests demonstrated safety of assembly operations, including reaching down into the cavity to perform manual adjustment with components in place. As gun devices became smaller than the Hiroshima weapon, experimental safety guidance had to include the effects of surrounding materials in, for example, the breech of a naval gun.
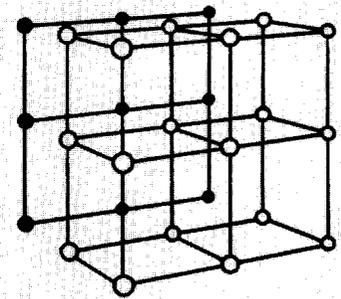

Fig. 5. Schematic arrangement of weapon cores during the criticality tests with the vault shown in Fig. 4. Two separate frames supported the cores at the positions represented by the solid and open circles.
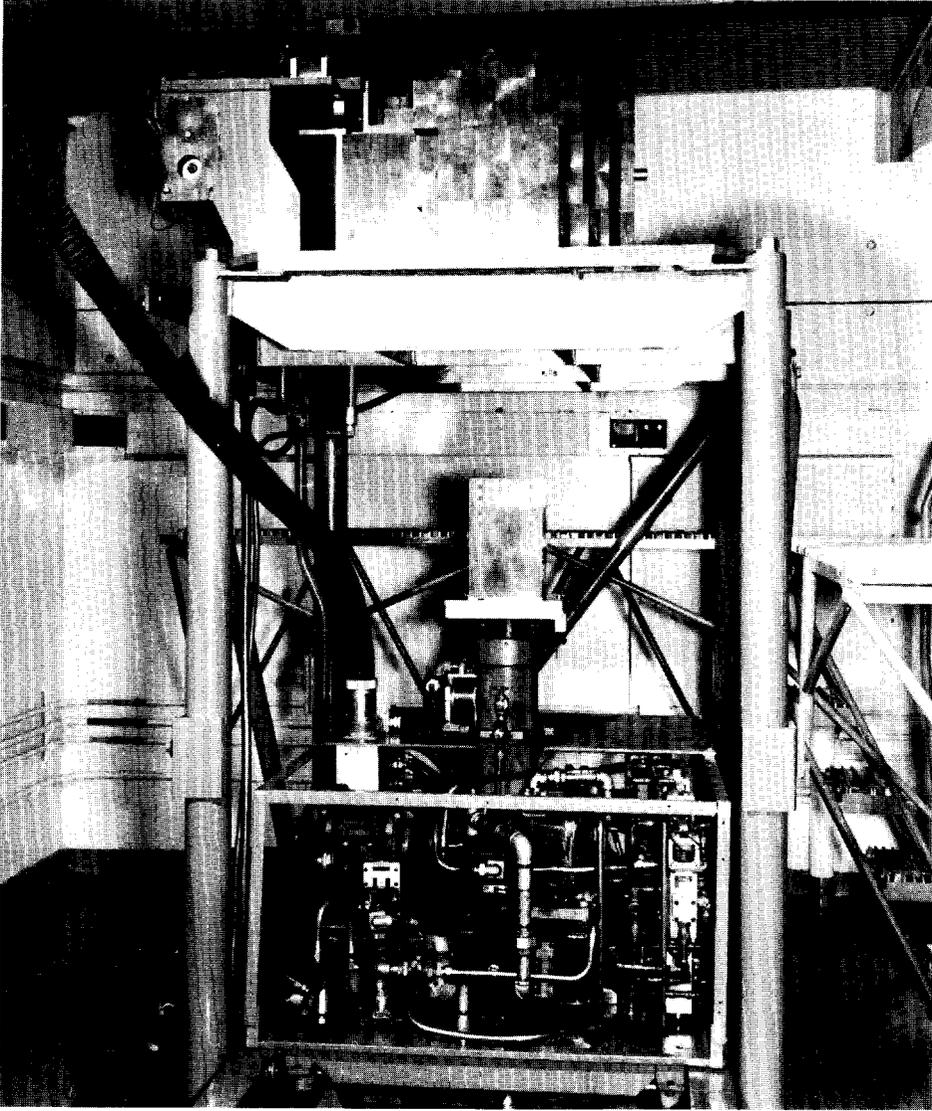
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Fig. 6. The Topsy critical assembly. The central box-like structure contains an enriched-uranium core embedded in some natural uranium reflector. This structure is raised by remote control into a cavity in the main reflector body, the pile of large uranium blocks above. Spherical or cylindrical cores were approximated by arrays of half-inch cubes of enriched uranium.

The first critical assembly for this purpose (Fig. 6) began operating in late 1948. Named Topsy—she just grew—the assembly consisted of a nearly spherical core of highly enriched uranium embedded in thick natural uranium. Topsy was followed in 1951 by a bare sphere of highly enriched uranium, named Lady Godiva by Raemer Schreiber because, like the lady of Coventry, she was unclad. Ultimately we also obtained data on plutonium and uranium-233 assemblies as bare spheres and spheres reflected by thick natural uranium. Other simple assemblies consisted of combinations of fissile materials of interest to weapon designers, some in thin reflectors of various materials. Over the years hundreds of critical specifications have accumulated, which, when used for validation, have greatly expanded the range and reliability of detailed neutronic calculations.

Criticality control is necessary in aspects of the weapons program other than weapon safety. Accidental criticality must be avoided in the purification of fissile material, the production of metal, the fabrication of components, and the recovery of scrap. Other nuclear programs. such as the production of reactor fuel, involve similar operations and therefore require similar criticality information for safety measures. Criticality data from Los Alamos have been incorporated in compilations and safety guides and standards. Thus the scope of Los Alamos criticality safety activities has been national and even international. For example, Los Alamos has hosted two international meetings on criticality, and our short courses on criticality safety, conducted in cooperation with the University of New Mexico, have been attended by interested persons from other countries. ■

Interaction among most simple implosion weapons of modern design is not a consideration except, perhaps, for clustered configurations. For some fission-fusion devices, however, interaction of weapons may be sufficiently important to require measurement. In one instance we tested an array of fission-fusion weapons that simulated a shipboard storage proposal. The tests were carried out at an assembly site because transportation of the weapons to a critical assembly facility was undesirable.

In the 1950s the critical assemblies group became involved in reactor-related activities culminating in the Rover rocket-propulsion reactor program. Although these activities eventually occupied most of our effort, weapon tests retained the highest priority.

We had to be prepared for short-notice safety checks on each device destined for testing in the Pacific or Nevada. Typically, about one day was available for the safety check between completion of the device and shipment to the test site. Obtaining meaningful data on short notice was challenging but exhilarating.

Measured criticality data for easily calculated systems have also been of value for improving or confirming the detailed neutronic calculations that enter weapon design.

Further Reading

Hugh C. Paxton, "Thirty-Five Years at Pajarito Canyon Site," Los Alamos Scientific Laboratory report LA-7121-H, Rev. (1981).

Hugh C. Paxton, "A History of Critical Experiments at Pajarito Site," Los Alamos National Laboratory report LA-9685-H (to be published).

Prompt Criticality Under Control

L ady Godiva became the forerunner of the family of fast-pulse reactors at Los Alamos, Sandia National Laboratories, White Sands Missile Range, Aberdeen Proving Ground, and Oak Ridge National Laboratory. These reactors simulate the radiation from a weapon that occurs beyond the weapon's blast-damage range and therefore are used to test instruments, rocket guidance systems, and electronic equipment for proper functioning in the presence of a weapon burst.

In mid 1953 Lady Godiva, essentially an unreflected sphere of highly enriched uranium, was coaxed gingerly to prompt criticality (the usually forbidden region) and slightly beyond. The typical result was radiation from a sharp, intense fission puke terminated by expansion of the uranium. Although the intent was simply to confirm predictions about the assembly's behavior at superprompt criticality, these pulses were immediately in demand as nearly instantaneous sources of radiation for experiments in areas ranging from biology to solid-state physics, and soon they were used to proof-test instrumentation and controls that were supposed to withstand the radiation from a nuclear explosion.

The total of about 1000 prompt pulses from Lady Godiva was not without incident, for twice the safe limit beyond prompt criticality was overstepped. The first incident did not cause irreparable damage, but in the second uranium parts became too badly warped and corroded for further use. The assembly was then replaced by Godiva 11, designed specifically for burst production. This first of the fast-pulse reactors has been succeeded at Los Alamos by Godiva IV. ●

Top. The Lady Godiva critical assembly of highly enriched uranium. A nearly spherical, unreflected critical assembly was formed as the upper cap was dropped and the lower cap was slowly raised. Lady Godiva was portable and was even operated outdoors to eliminate the effects of neutron reflection from the kiva walls.

Bottom. Lady Godiva after the accident that led to her retirement. The enriched-uranium parts were severely warped and corroded, having approached the melting point at the center of the assembly. The support was damaged as a result of mechanical shock.