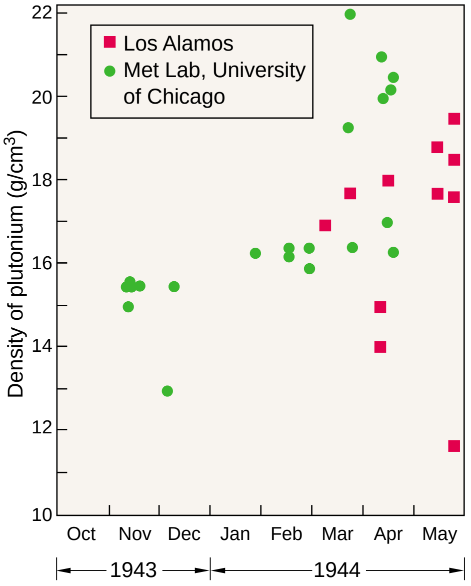
The Plutonium Challenge

Stockpile stewardship



The variations in plutonium density baffled Manhattan Project chemists and metallurgists until about midway through 1944, when they discovered that plutonium had no less than five allotropic phases between room temperature and the melting point.

Challenging as it may be to understand and mitigate the problems of plutonium aging, by far the most difficult problem we face today is the aging of our technical staff. Because plutonium science is enormously complex, we are just now beginning to understand it at a fundamental level. Our approach has therefore been largely empirical. But experience rests with the practitioners, and unfortunately, these practitioners are aging. We are in danger of losing their expertise and advice before we develop a more fundamental understanding of plutonium—one that can more easily be taught and sustained over time.

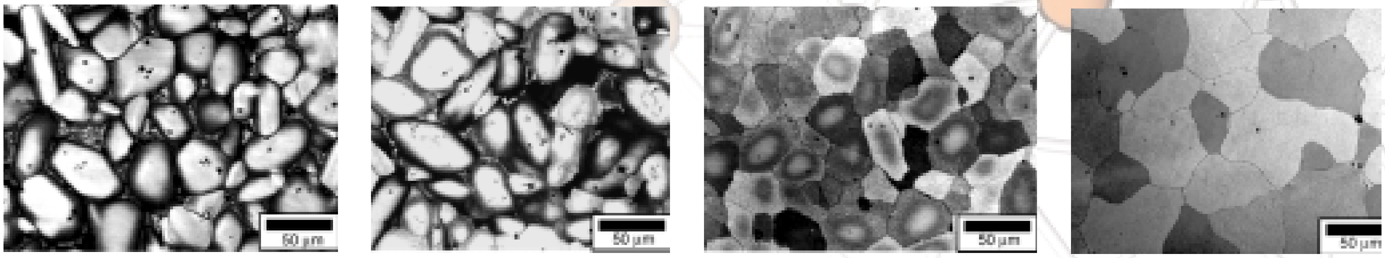
The plutonium pit is at the heart of the bomb. Fabrication of the first pits during the Manhattan Project was a tour de force. In 1944 and 1945, when gram and kilogram quantities of plutonium became available, this metal was found to be at odds not only with itself but also with everyone who touched it. Just enough was learned about this mysterious new element that the chemists and metallurgists were able to reduce the reactor product to metal. They could subsequently purify it, alloy it (so they could stabilize it and press it into shape), coat it (so they could handle it), and keep it together long enough before the plutonium bomb exploded at Trinity and Nagasaki.

Over the following 50 years, Los Alamos scientists and many other scientists around the world tried to decipher the mysteries of plutonium. Fortunately, many of the great academics recruited to Los Alamos during the Manhattan Project kept an affiliation with the Laboratory. Numerous professors spent their summers at Los Alamos and sent their best graduates to work at the Laboratory. Willi Zachariasen, probably the best crystallographer of all times, came from the University of Chicago to continue his wartime quest for understanding plutonium. He eventually determined the incredibly complex monoclinic crystal structure of the α-phase.

It seemed that the more we learned about plutonium, the deeper its mysteries became. The sensitivity of plutonium to thermal changes was matched by similar sensitivities to the application of pressure and to the addition of chemical elements. In fact, plutonium appeared to change phase with very little provocation at all and by almost every transformation mechanism known to scientists.

From the 1950s through the 1970s, Los Alamos, Livermore, and Rocky Flats metallurgists, chemists, and engineers extracted as much as possible from the international scientific work on plutonium to help shape the U.S. classified research program. That program provided sufficient knowledge to enable the development of increasingly sophisticated physics designs required by the drive for devices with a constantly higher yield-to-weight ratio. In fact, the drive for improved performance was so relentless that it far surpassed our progress in understanding plutonium at a fundamental level. As a result, much of the engineering performance requirements were met through empirical knowledge and day-to-day experience. Manufacturing plutonium was more of an art than a science. Fortunately, however, some of the engineering requirements experienced during manufacturing, storage, and delivery could be tested in the laboratory.

Because the implosion performance has never been adequately simulated,



As cast

2 h

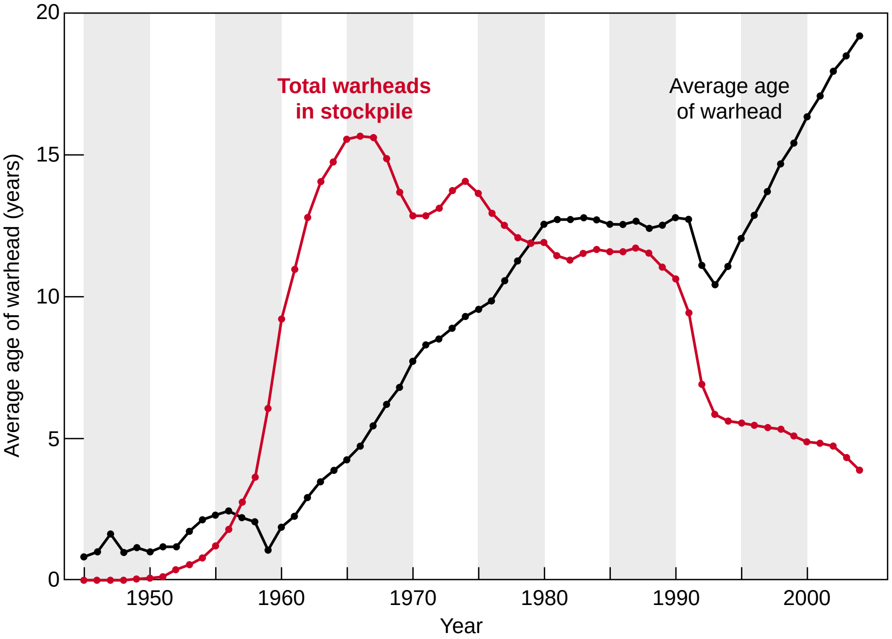
20 h

720 h

A few atomic percent gallium is typically added to plutonium to retain the face-centered-cubic phase, which is easily shaped into components. However, as Pu-Ga alloys cool during casting, gallium segregates and leaves a nonuniform distribution across the metallic grains. This sequence of micrographs demonstrates gradual gallium homogenization during annealing for long times at 460°C. The as-cast sample on the left exhibits regions high in gallium in the grain centers (etched to appear very light). At longer times, the gallium concentration becomes more uniform, as demonstrated by the more uniform coloration within the grains. After 720 h, the sample is completely uniform—the variations from grain to grain result strictly from differences in crystalline orientation.

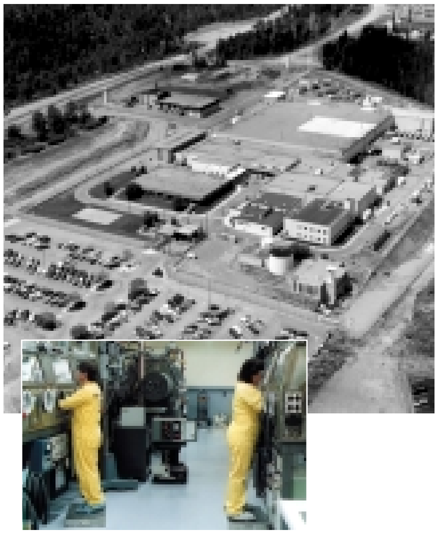
we relied heavily on nuclear testing—first, in the atmosphere and, after 1963, underground. Yet we had to develop great skills in modeling the physics and, with the aid of large-scale computing, we “calibrated” the performance of plutonium during the extraordinarily complex conditions of a nuclear explosion. Problems that were often discovered through calculations or stockpile surveillance were fixed, but they often required nuclear tests to ensure the adequacy of the fix. Some expected concerns about the aging of plutonium were most easily addressed by the replacement of old systems with new, more-capable systems.

The enormous geopolitical changes of the past decade have brought about an entirely different approach to our nuclear weapons responsibilities at Los Alamos. Nuclear weapons remain the cornerstone of U.S. national security strategy, and our job is to keep them safe and reliable into the indefinite future. But we must do so without nuclear testing, according to the provisions of the Comprehensive Test Ban



The number of weapons in the stockpile is decreasing, and in another decade, the ages of most weapons will be well beyond their original design lifetimes.

Treaty. In addition, in 1992, President Bush adopted the policy of not fielding weapons of new design, so we must also forgo the practice of fixing stockpile problems by replacing old designs with new ones. Although the number of weapons in our stockpile is decreasing because of arms reductions agreements with Russia, the remaining weapons are approaching or exceeding their original design lifetimes.



The plutonium facility at Los Alamos is developing the capability to remanufacture small lots of plutonium pits and now carries out all surveillance activities necessary for stockpile stewardship. All experiments and tests are conducted inside a glove-box environment (inset).

Nuclear Weapons Certification

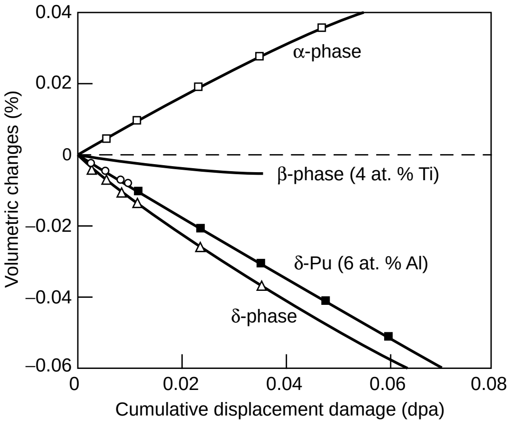
Yearly, the directors of the three nuclear weapons laboratories—Los Alamos National Laboratory, Lawrence Livermore National Laboratory, and Sandia National Laboratories—certify the weapons designed by their labs as safe and reliable without testing. This annual certification drives the stewardship challenge. Plutonium is a particularly demanding part of that challenge because it is the component that we cannot test under conditions that produce a nuclear yield. Many of the other components can be adequately tested under simulated conditions.

Our approach to stewardship is to extend the lifetimes of pits (requalify) or remanufacture the pits in the warheads scheduled to remain in the stockpile. The United States is currently establishing a pit production capability of very limited capacity at Los Alamos. Extending the lifetimes of pits to 50 years and beyond provides a substantial financial incentive because of the high construction costs for new plutonium facilities. Certification of requalified or remanufactured pits is a major challenge for metallurgists, chemists, engineers, and weapon designers.

Because the pits in the stockpile are aging, we must significantly upgrade our surveillance. It is therefore imperative that we develop new, more-sophisticated nondestructive techniques to assess changes caused by aging. New diagnostic capabilities under development may allow detecting changes early and predicting the lifetimes of pits. Several age-related issues about plutonium concern us. Among them are surface changes caused by corrosion and dimensional changes caused by potential phase instabilities. In addition, plutonium undergoes continuous radioactive decay during which it transmutes itself. This radioactive decay leads to long-term chemical changes, as well as short-term self-irradiation damage. To have any hope of assessing the effects of these complex events on the already hypersensitive plutonium lattice, we must develop a better fundamental understanding of plutonium.

Remanufacturing the plutonium pits is another challenging task. The United States has not manufactured a war-reserve plutonium pit in 12 years. Because the Rocky Flats plant is no longer operational, remanufacture will be done at Los Alamos with new people, new equipment, and some new processes. Certifying that such pits are functionally equivalent to those originally manufactured and tested is one of the principal challenges of stockpile stewardship.

When nuclear tests were allowed, we could work around what we did not understand about plutonium by testing its performance. Now, we must understand plutonium better, then test it in every conceivable way permitted, and finally have the designers test their confidence by comparing the new computational results with those stored in the archives. Better understanding necessarily means incorporating the influence of microstructure on performance. Consequently, computational requirements will increase by several orders of magnitude if microstructure-based materials models are to be incorporated into the physics design codes. Such increased sophistication in materials behavior drives much of the need for the Accelerated Strategic Computing Initiative (ASCI) of the Department of Energy.



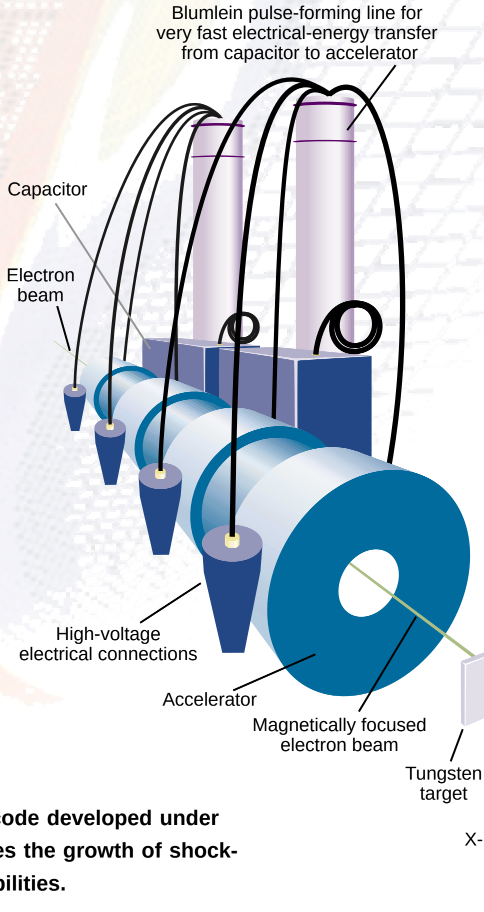
Self-irradiation produced these volume changes in plutonium at cryogenic temperatures. The volume changes eventually saturated at approximately 10% for the α-phase and 15% for the δ-phase. Fortunately, much of the lattice damage from self-irradiation anneals out at ambient temperature. However, we are still studying the effects of helium and transmutation products such as americium, uranium, and neptunium.

The U1A tunnel complex at the Nevada Test Site (above) is being used to study the response of plutonium alloys to shock loading. The results are incorporated into computer simulations of the nuclear-weapon implosion process. Experimental alcoves for these experiments and the diagnostics alcove (top right) are approximately 1000 feet underground.

The Challenge

The technical challenge is to keep the stockpile weapons safe and reliable without nuclear testing. To predict the lifetimes of existing pits or to remanufacture pits so that they can be certified will require a better understanding of how plutonium ages and how microstructure affects performance. To succeed in our stockpile stewardship mission, we will have to combine such understanding with significantly increased computing power and permissible experiments. Improving our fundamental understanding of plutonium requires that we continue to work closely at the frontiers of actinide science with the academic community and the international research community. Indeed, we must continue to attract and retain the best and the brightest of the next generation of scientists and engineers.

From a policy and societal point of view, the U.S. government must deter all our country's potential adversaries with a smaller number of nuclear weapons. We, who work at the nuclear weapons laboratories, must be able to assure our leaders that the weapons we designed and retained in the stockpile will work reliably if they ever have to be used.■



DARHT (photo below), a dual-axis x-ray facility now under construction at Los Alamos, will provide 3-D digital x-ray images of nonnuclear implosion tests. The 4000-A, 20-MeV pulsed-electron beam from an advanced accelerator (diagram below, left) produces intense nanosecond x-ray pulses that can capture very high resolution images of the detonation and implosion. The National Ignition Facility (NIF) at Lawrence Livermore National Laboratory will produce high-energy densities that overlap those produced in nuclear weapons and will eventually be used to implode tiny fusion capsules. The Z pulsed-power accelerator at Sandia National Laboratories, the world's most powerful laboratory x-ray source, is being used to study material properties and radiation transport at very high densities and temperatures.

RAGE, a 3-D code developed under ASCI, simulates the growth of shock-induced instabilities.