U.S. Weapons Plutonium

Advances in plutonium science enhance understanding of this mysterious element.

Aging Gracefully

ONE of the most important components of a nuclear weapon is the core or pit, a sphere of plutonium-239 that is compressed by conventional explosives to create a nuclear chain reaction. Understanding the performance of plutonium pits is crucial to Lawrence Livermore scientists and engineers who must ensure the safety and reliability of the nation's nuclear stockpile. Planning the future needs of the U.S. nuclear weapons complex also depends on confidence in the long-term stability of the pit and credible estimates for pit lifetimes.

Many physicists, metallurgists, and chemists have worried that the natural radiation produced by plutonium over many years might eventually damage the pit and compromise weapon performance. Although plutonium-239 has a half-life of 24,000 years, its decay rate is high enough to produce a significant amount of damage after only a few decades. In addition, many pits are in deployed weapon systems that are far older than their originally planned lifetimes. To maintain a system past its designed lifetime, weapon scientists need a more thorough understanding of plutonium as it ages.

"People have been studying metals for 3,000 years, but plutonium for less than 70," says Livermore metallurgist Adam Schwartz. Conducting research on plutonium's electrical, chemical, and physical properties is also much more difficult than it is on other metals because plutonium is radioactive, highly toxic, and sensitive to changes in temperature, pressure, and composition. In addition, its properties do not always change in linear fashion.

For years, weapon scientists examined plutonium's bulk behavior by testing nuclear devices at the Department of Energy's Nevada Test Site. However, the nation stopped underground nuclear testing in 1992. In its place, the department launched a vigorous science-based stockpile stewardship program to evaluate and certify the safety, security, and reliability of the nation's weapons.

In 1997, as part of this program, the National Nuclear Security Administration's (NNSA's) Enhanced Surveillance Campaign began funding a $100 million study at Lawrence Livermore and Los Alamos national laboratories to examine how nuclear warhead materials age and determine the likely effects of aging on weapon performance and safety. Livermore's research teams include physicists, chemists, engineers, materials scientists, and computer scientists from the Chemistry, Materials, and Life Sciences; Physics and Advanced Technologies; Defense and Nuclear Technologies; Engineering; Computation; and Energy and Environment directorates.

On November 29, 2006, NNSA announced the results from this effort: "These studies show that the degradation of plutonium in our nuclear weapons will not affect warhead reliability for decades," said Linton Brooks, then administrator of NNSA. "It is now clear that although plutonium aging contributes, other factors control the overall life expectancy of nuclear weapons systems." The laboratories' research teams also determined that the minimum lifetime for most of the plutonium pits in the nation's nuclear weapon stockpile is at least 85 years—25 to 40 years longer than scientists had previously estimated.

JASON, an independent panel of scientists who advise the government on science and technology, reviewed the scientific studies used to assess pit lifetimes. The JASON reviewers concluded that the credible lifetime for most of the pit types is at least 100 years. They also noted that mitigation plans have been proposed or are being implemented for those types with less than 100 years of projected stability.

These findings are the latest chapter in a scientific effort that began in 1941 when plutonium was discovered. Since the Laboratory's founding in 1952, Livermore scientists have made significant contributions to the science of plutonium and its closely related elements, called actinides. (See the box on p. 14.) The

Plutonium Grudgingly Reveals Its Secrets

In 1941, Glenn Seaborg, Edwin McMillan, Joseph Kennedy, and Arthur Wahl synthesized plutonium, element 94, in a cyclotron at the University of California at Berkeley. Since then, scientists have identified 21 plutonium radioisotopes. The most stable isotopes are plutonium-244, with a half-life of 80.8 million years; plutonium-242, with a half-life of 373,300 years; and plutonium-239—the isotope of greatest interest—with a half-life of 24,110 years.

When plutonium-239 undergoes fission, the nucleus releases enormous energy. One kilogram of this metal is equivalent to about 22 million kilowatt-hours of heat energy. When detonated, it produces an explosion equal to about 4 kilotons of chemical explosives.

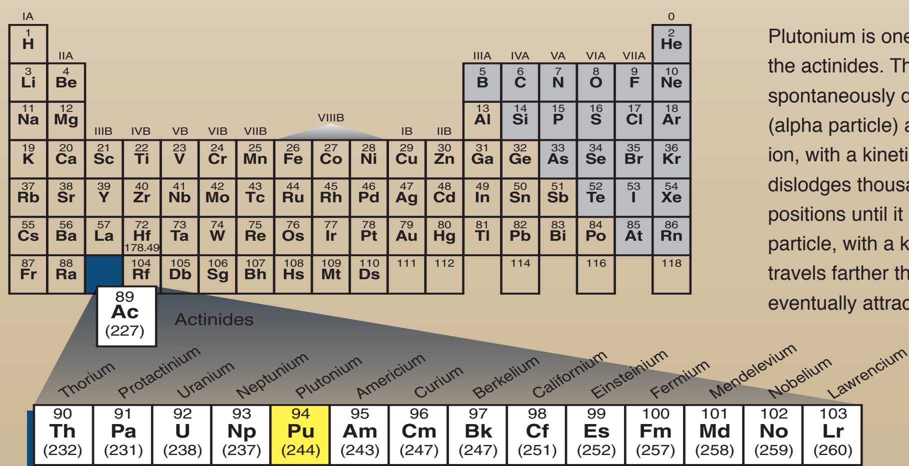
Plutonium is part of a series of 14 radioactive elements called the actinides, which all contain electrons in an outer shell called 5f. Plutonium's complexity derives from its position in the periodic table: the transition point at which 5f electrons change from forming bonds with other elements to being chemically inert.

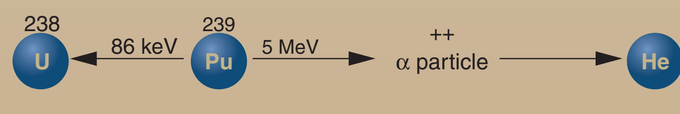
Scientists consider plutonium the most perplexing element in existence. It joins with virtually every other element to make compounds, complexes, or alloys and forms up to 12 chemical bonds to molecules in solution, something no other element can do. Plutonium-239 goes through six solid-state phase transformations, more than any other element. Large volume and density changes occur as it transitions through these six phases to its liquid state at 640°C. Under pressure, it also exhibits a seventh phase.

An atom of plutonium-239 spontaneously decays into a doubly charged helium nucleus (alpha particle) and a uranium-235 ion. The uranium-235 atom has an initial kinetic energy of about 85 kiloelectronvolts. Before this atom comes to rest, it dislodges thousands of other plutonium atoms from their normal positions in the crystal lattice, thereby creating a large collision cascade. The alpha particle, which has an energy of 5 megaelectronvolts, travels farther. However, because it is relatively small, it creates fewer lattice defects.

Within 200 nanoseconds, about 90 percent of these displaced atoms return to a normal lattice position, a process called self-healing. If a defect does not self-heal, the lattice may have with vacancies, where atoms are missing, and interstitial atoms, where atoms are squeezed between other regularly spaced atoms. On average, each atom of plutonium is displaced once every 10 years.

After the alpha particle comes to rest in the lattice, it attracts two electrons from its surroundings to become a helium atom. This process creates about 29 helium atoms per year for every 1 million atoms of plutonium. The aggregation of helium atoms causes helium bubbles to form. Over many decades, this accumulation of helium becomes substantial, although bubbles stop growing once their diameter reaches about 1.4 nanometers. Why bubbles do not continue to expand is one of many questions that remain to be explained about this most mysterious of elements.





Plutonium is one of 14 radioactive elements called the actinides. Through alpha decay, a plutonium atom spontaneously decays into a doubly charged helium nucleus (alpha particle) and a uranium-235 ion. The uranium-235 ion, with a kinetic energy of 85 kiloelectronvolts (keV), dislodges thousands of plutonium atoms from their lattice positions until it comes to rest. The much smaller alpha (α) particle, with a kinetic energy of 5 megaelectronvolts (MeV), travels farther through the lattice but creates fewer defects. It eventually attracts two electrons to become a helium atom.

research has also benefited from studies conducted at the Laboratory's Glenn T. Seaborg Institute. (See *S&TR*, June 2000, pp. 15–22.)

As part of the NNSA study, Livermore scientists used some of the most accurate instruments in the world to measure the microstructural, physical, and chemical properties of plutonium and its alloys. In dynamic experiments such as gas-gun tests and static studies using diamond anvil cells (DACs), they examined how radioactive decay affects plutonium's structure, phase stability, and equation of state (EOS). They also measured the element's density and volume with unprecedented accuracy and reviewed data from past nuclear tests.

Advanced computational models of weapons physics and theoretical studies helped researchers design the experiments and provided data to complement the experimental results. In addition, calculations of design sensitivity determined the extent to which aging pits would affect performance of different weapon systems decades from today. By combining experimental and computational resources, scientists working on the NNSA study derived the most accurate estimates ever obtained for pit lifetimes.

Decay Generates Helium

The isotope plutonium-239 undergoes alpha-particle decay, in which a plutonium atom spontaneously disintegrates, turning into a uranium-235 atom while emitting a high-energy alpha particle (helium nucleus). The alpha particle and the uranium atom fly off in opposite directions, disrupting nearby atoms. As the alpha particle comes to rest, it picks up two electrons and becomes a helium atom. Weapon designers have long sought greater assurance that a pit would retain its size, shape, and strength in the presence of an ever-increasing amount of damage from alpha-particle decay.

One concern is that at some threshold, the accumulating damage might induce a change from plutonium's ductile delta phase used in nuclear warheads to the denser alpha phase, which is more brittle. Such a phase change could cause the crystalline lattice to crack and would likely degrade a weapon's performance. Helium atoms also might collect in large bubbles, weakening the part or otherwise changing its behavior.

Many scientists have also been concerned that a phenomenon called void swelling might occur. Studies of radiation damage in other metals, such as the steels used in nuclear reactors, show that helium buildup, in combination with vacancies in a metal's crystalline lattice, can produce voids in addition to helium bubbles. Voids cause most metals to swell in size, losing their critical shape and strength.

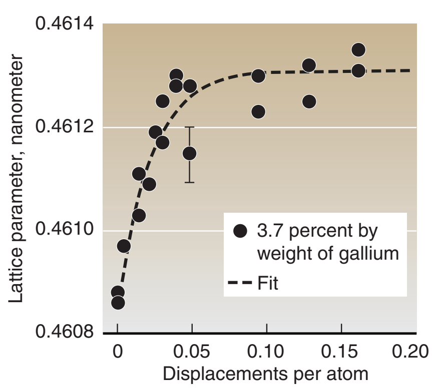
Results from the NNSA study indicate that the accumulation of helium would not significantly change the properties of plutonium in pits up to a century after they were manufactured. The helium bubbles appear to be distributed uniformly throughout the material, with only a small change in properties including a slight increase in volume.

The study also revealed no evidence of void swelling or other catastrophic damage in plutonium over several decades. "We now understand damage mechanisms in plutonium more precisely," says Schwartz. "The radiation damage is slow, and self-healing mechanisms occur in which atoms move back into the lattice from which they were displaced."

Schwartz notes that the behavior of aging plutonium is also important in dismantling nuclear weapons and disposing of pits from retired weapons. In addition, nuclear power plants produce plutonium as a by-product of burning enriched uranium fuel, and this waste must be secured against diversion or theft. The most likely methods for disposing of unwanted plutonium will be to burn it in a future nuclear reactor or sequester it underground in a geologic formation. With either approach, plutonium must be kept for many decades or centuries, and solid scientific understanding is required for its safe handling and storage. (See the box on p. 17.)

Accelerating the Aging Process

Researchers combined experiments and measurements to characterize samples of both old and new plutonium. The oldest weapons-grade plutonium made in the U.S. and available for detailed analysis was about 45 years old, taken from pits retired from the stockpile. The processes used to manufacture this plutonium differ somewhat from those used to make pits in today's stockpile. The oldest samples most directly comparable to current weapons were about 30 years old.



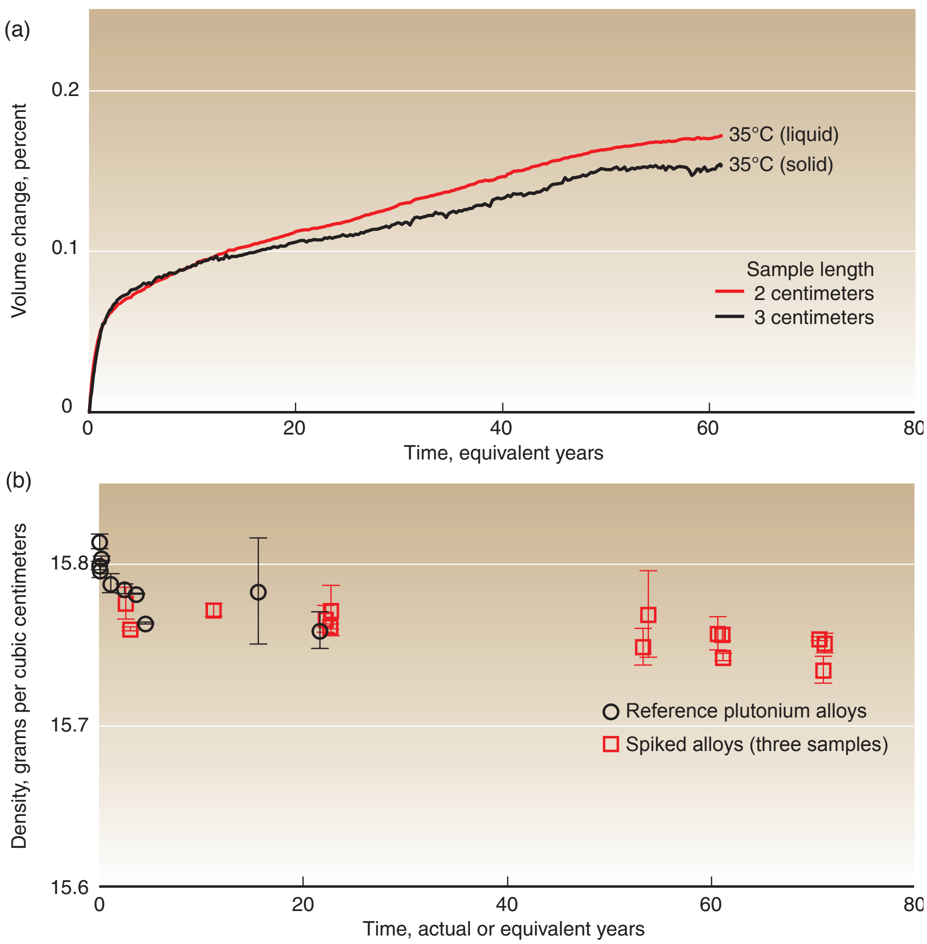
Alpha decay has a measurable effect on the lattice parameter, the distance between two faces of the crystalline lattice in delta-phase plutonium. The rate at which alpha decay knocks an atom from its original lattice position is measured in displacements per atom (dpa). In plutonium, 1 dpa equals 10 years. Experimental data show that early on, alpha-decay-induced radiation damage reaches a steady state where the rate of self-healing almost equals the rate of alpha-decay damage.

Plutonium is the most complex metallic element. Without a thorough scientific understanding of its behavior, some aging effects could appear suddenly. Scientists cannot merely extrapolate the effects found in samples from retired pits to determine the aging mechanisms over time. To simulate the properties of pits many decades into the future, Livermore and Los Alamos researchers accelerated the age of samples by adding isotopes with shorter half-lives. Using a recipe developed by Livermore physicist Bill Wolfer, the researchers “spiked” an alloy of weapons-grade plutonium-239 with 7.5 percent by weight of plutonium-238, which has a half-life of 87 years. Plutonium-238 is used to provide electrical power for deep-space probes such as the National Aeronautics and Space Administration’s Galileo mission to Jupiter.

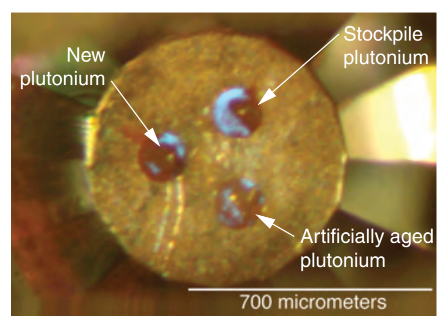
This spiked alloy accumulates radiation damage at a rate 16 times faster than weapons-grade plutonium alone. “Spiking gives us 16 years additional aging for every year of natural aging,” says chemical engineer Karen Dodson, who has supervised the production of artificially aged plutonium. The oldest spiked samples, manufactured about 5 years ago, are now equivalent to 80-year-old plutonium. Because the damage rate is much higher in the spiked plutonium, those samples are maintained at a slightly elevated temperature to ensure that the self-healing rate is appropriately accelerated as well.

Livermore metallurgists made several batches of artificially aged plutonium alloys in a glove box under inert atmosphere to prevent oxidation. For each production batch, dozens of samples were machined at various sizes for the dynamic and static experiments. For example, samples measuring 2 and 3 centimeters long were manufactured for dilatometry experiments, which measure dimensions to extreme accuracy. Metallurgists also made reference samples that did not contain plutonium-238.

Chemist Brandon Chung is measuring property changes as a function of age for the various samples, including those that are artificially aged. He records changes in density, dimension, tensile and compressive strength, and hardness,



(a) Dilatometry measurements show a sharp rise and then an extremely slow increase in volume as plutonium ages over decades. (b) Immersion density experiments show a marked density decrease in both spiked and reference plutonium alloys, followed by a very slow, linear decrease.



Three plutonium samples, each 100 micrometers in diameter, are squeezed in a diamond anvil cell to determine how radioactive decay affects the metal’s structure. The samples shown here include recently produced plutonium, artificially aged plutonium, and plutonium retrieved from a retired pit several decades old.

all properties related to the element's composition and crystalline microstructure.

His dilatometry and immersion density measurements on newly made spiked and unspiked alloys showed a limited period of significant volume expansion (and its corollary, density reduction), followed by extremely slow changes in volume and density. He found no indication of void swelling. "Plutonium ages at a much slower rate than we originally thought," says Chung, who will continue to measure the samples as they age.

DAC Applies the Pressure

Physicists Choong-Shik Yoo and Hyunchae Cynn conducted experiments on plutonium using DACs. In these experiments, a small mechanical press slowly squeezes a microgram or so of material between two small, flat-tipped diamonds, achieving pressures as high as 100 gigapascals. To better examine actinides under extremely high pressures, Laboratory researchers developed "designer" DACs, which have advanced sensors integrated into the cell. (See *S&TR*, December 2004, pp. 4–11.)

The two scientists conducted the DAC experiments at the Advanced Photon Source (APS) at Argonne National Laboratory. APS is the brightest x-ray source in the world and can reveal minute changes in crystalline materials. In this series of experiments, the Livermore team examined plutonium samples at different stages of the aging process. Samples included material recently produced, artificially aged, and taken from retired pits up to 45 years old.

The experiments used three samples of different ages in the same DAC. Each sample measured 0.1 millimeter in diameter and was secured within a metal gasket. The samples were heated either electrically or by a laser to several thousand kelvins. As pressure was slowly increased, the researchers tracked volume changes that occurred when samples transitioned from one phase to another. The APS experiments

Plutonium Futures Conference Growing in Popularity

One of the most important international forums for discussing the science of plutonium and other actinides is the Plutonium Futures Conference, held every three years since 1997. The conference highlights the latest research on the physical and chemical properties and environmental interactions of plutonium and other actinide elements.

Lawrence Livermore hosted the most recent conference in July 2006 at Pacific Grove, California. The program consisted of lectures, invited papers, and plenary sessions, which included policy makers and scientific leaders. U.S. and international scientists, engineers, faculty, and students from universities, research institutes, and nuclear complexes attended the conference, which attracted nearly 400 participants—many more than organizers had predicted.

Topics included the safe storage and long-term management of surplus weapons materials and large inventories of actinides generated by civilian nuclear power plants. Participants also discussed actinides in the environment; their properties, chemistry, quantum mechanics, and electron structure; and methods to detect them. "The technical basis for addressing these issues requires intensive and increasing understanding of the underlying plutonium and other actinide science and technology," says Livermore physicist and conference organizer Mike Fluss.

"Plutonium is the linchpin of any nuclear energy strategy," says Fluss. "It is a by-product from burning uranium. In future fuel cycles, engineers must find methods to dispose of it." The Department of Energy's recent nuclear energy initiative, called Global Nuclear Energy Partnership, proposes to reduce nuclear waste by using new proliferation-resistant technologies to recycle these fuels. According to Fluss, such an effort requires solid scientific underpinnings.

"The study of plutonium is a 21st century grand challenge for chemists, materials scientists, and solid-state physicists," says Fluss. "Each Plutonium Futures Conference presents an opportunity to bring these communities together." In particular, scientists believe that if they could fully understand the complex interactions of plutonium's 5f electrons, they could understand the electron behavior of any other element.

Lawrence Livermore hosted the 2006 Plutonium Futures Conference, which highlighted current scientific research on plutonium and other actinides. Nearly 400 participants attended from throughout the world.

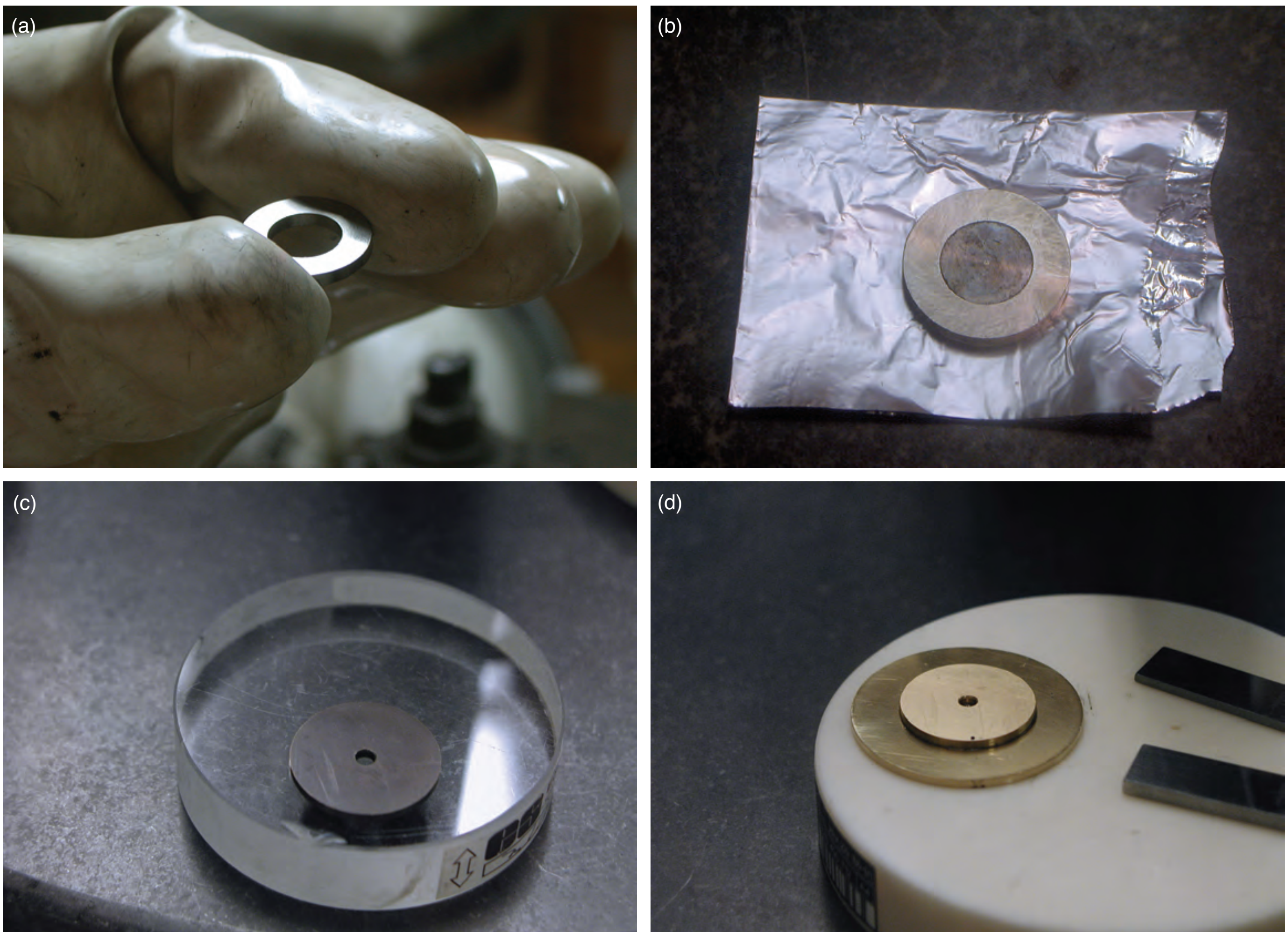
revealed no significant differences among the plutonium samples and no sudden or unexpected changes in properties. (See the bottom right figure on p. 16.)

Consistent Data from JASPER

Researchers at the Joint Actinide Shock Physics Experimental Research (JASPER) Facility acquired the first simultaneous dynamic comparison data for naturally aged plutonium obtained from retired pits versus newly produced plutonium. JASPER is a 30-meter-long, two-stage light-gas gun located at the Nevada Test Site.

Designed to gather EOS data on plutonium, the gas gun hurls projectiles at speeds up to 8 kilometers per second at plutonium targets. The impact produces an extremely high-pressure shock wave (about 600 gigapascals) in the target, raising its temperature to as high as 7,000 kelvins. (See *S&TR*, June 2004, pp. 4–11.)

For these experiments, targets were made of old plutonium pressed into a disk of new plutonium and machined extremely flat (to 1.5-micrometer variation). The disk had an outer diameter of 32 millimeters, and the old plutonium had an inner diameter of 19 millimeters. Nineteen pins



Researchers used the gas gun at the Joint Actinide Shock Physics Experimental Research Facility to simultaneously shock samples of aged and new plutonium. The sample assembly sequence shows (a) a ring of new, machined plutonium; (b) old plutonium pushed into the ring; (c) the concentric rings after being machined; and (d) the concentric rings after they are polished, coated with gold, and bonded to a larger ring of new plutonium.

placed in the two samples gathered shock velocity data.

JASPER engineer Matt Cowan explains that with this target design, the two concentric samples experienced identical shock waves at precisely the same time. "Testing the two samples simultaneously is better than conducting separate experiments because gun velocities are always slightly different," says Cowan.

The results unequivocally showed no statistically significant difference in the EOS of the new and old plutonium. "The experiments achieved EOS data with an accuracy never achieved before," says physicist Neil Holmes, chief JASPER scientist. "The DAC and gas-gun experiments produced entirely consistent data. One technique is dynamic, the other static, but the answers were the same."

Images and Modeling Aid Studies

To supplement the data from these experimental activities, Schwartz worked with staff associate Mark Wall to directly observe plutonium samples with an electron microscope. The ability to directly image the accumulation of self-irradiation damage is critical to understanding the element's aging process, says Schwartz. Using Livermore's 300-kiloelectronvolt, field-emission transmission electron microscope (TEM), he and Wall observed spherically shaped helium bubbles, each about 1 nanometer in diameter—too tiny to be seen with conventional TEM instruments. The bubbles form as helium-filled vacancies migrate and coalesce. (See *S&TR*, March 2001, pp. 23–25.)

Schwartz has not observed voids in aged specimens with the TEM, although he regularly finds high densities of nanometer-size helium bubbles. "Although the number of helium bubbles grows over time, the bubble size is limited," says Schwartz.

Modeling and simulations done by Wolfer and others has aided the plutonium imaging and experimental effort. In one project, Wolfer studied whether the delta-phase plutonium–gallium alloy could eventually convert to a more stable phase, such as alpha, which is 25 percent more dense. His calculations show that self-radiation damage is, surprisingly, a key factor in stabilizing delta-phase plutonium.

According to Wolfer, gallium atoms in the delta-phase alloy tend to aggregate, which over time could contribute to a transition to another phase. However, plutonium decay disrupts any nearby gallium aggregation. "It's a dynamic but stable situation that contributes to plutonium's graceful aging," says Wolfer.

He and chemical engineer Alison Kubota are using Livermore's Blue Gene/L supercomputer to simulate collision cascades of uranium-235 atoms created from the alpha decay of plutonium atoms. The simulated reactions have a volume of 30 cubic nanometers and occur over a span of 10 picoseconds. The simulations, which use 32,768 processors and require 30 hours of computational time, depict an entire cascade of atomic collisions from one alpha-decay reaction.

Design Sensitivity

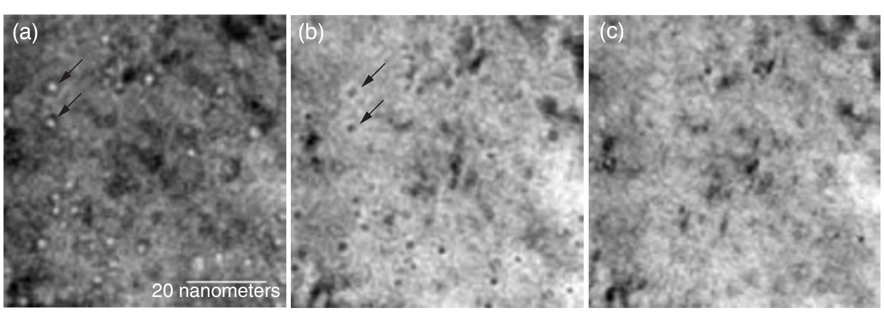
Data and models from this research improve the fidelity of design codes used to calculate the likely change in pit performance over the next few decades. Design sensitivity was then calculated for every weapon design in the stockpile.

"We take models that have been validated by nuclear testing, apply the documented effects of plutonium aging, and determine whether differences in the amount of aging affect the yield," says physicist Kris Winer. "Different devices have different performance margins, but we found that the effects of plutonium aging are very small."

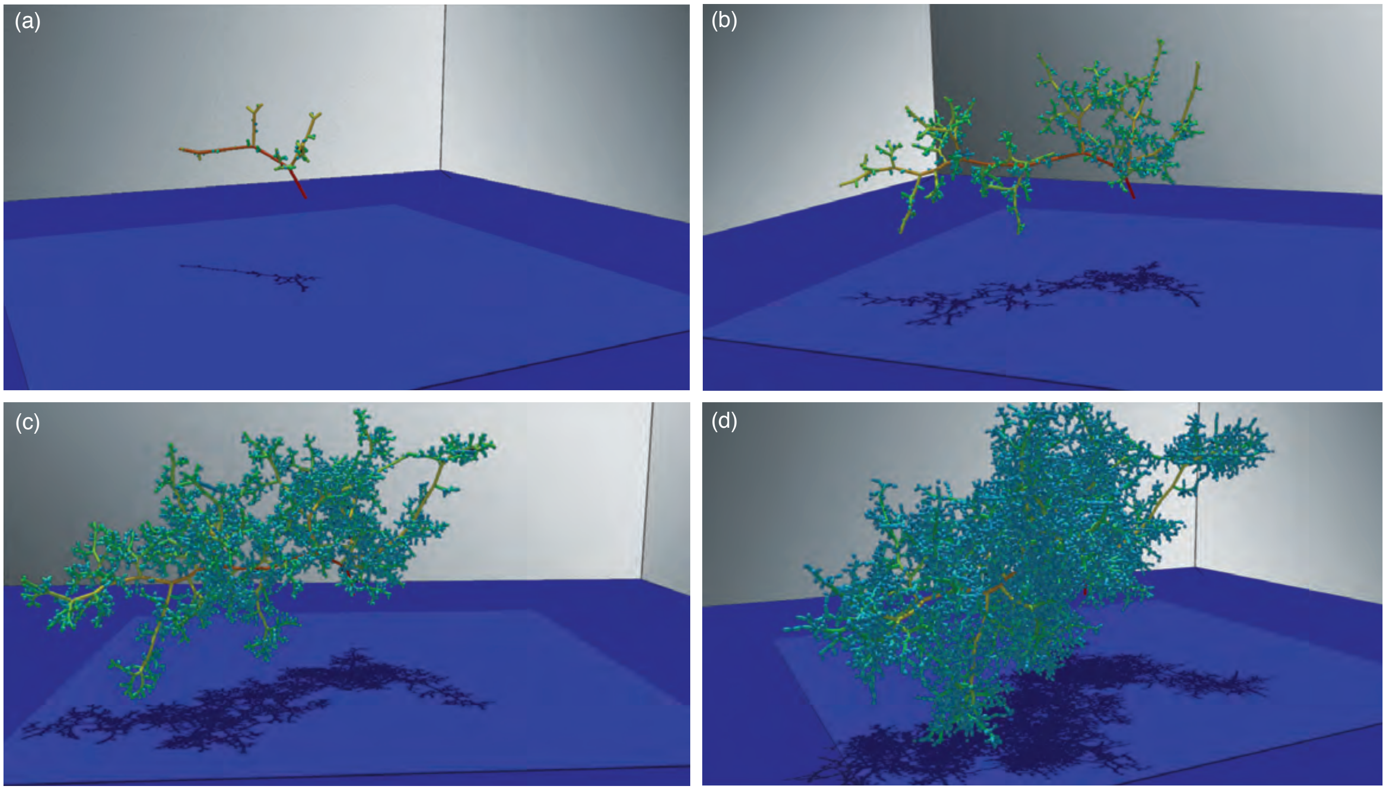
A small number of past underground experiments tested the same weapon design using plutonium samples at different stages of the aging process. "We compared the results of these detonations and determined what portion of the differences are caused by plutonium aging," says Winer. Again, these differences were small with large uncertainties.

An Informed Future

"Until NNSA commissioned this study, we didn't know with precision many of the details involved in plutonium aging," says Schwartz. "Now, we're finding out



Images of aged plutonium taken with a transmission electron microscope reveal helium bubbles measuring about 1 nanometer in diameter. (a) An underfocused image reveals tiny bubbles as a dark fringe surrounding a light dot. (b) In an overfocused image, bubbles appear as light fringes surrounding a dark dot. (c) A focused image shows no contrast from the bubbles.



(a–d) Frames extracted from a simulation run on Livermore's Blue Gene/L supercomputer show four steps in a collision cascade that occurs when a uranium-235 atom forms from the spontaneous alpha decay of a plutonium-239 atom. The newly formed uranium atom (in the red-colored region) begins to collide with plutonium atoms, forming a treelike structure. The entire simulation, which involved thousands of atoms, modeled a 10-picosecond process over an area of about 30 nanometers. Colors indicate energy levels, where red is high (85 kiloelectronvolts) and blue is low (4 electronvolts).

not only how plutonium ages but why. We still don't have all the details, but we have quantitatively improved our knowledge."

According to chemist Patrick Allen, leader of Livermore's plutonium aging study, "We now have a much better scientific understanding of several aspects of plutonium aging and so have greater confidence in the reliability of our pits and the stockpile." He points out, however, that nuclear weapon systems are extremely complicated, composed of thousands of different parts belonging to dozens of integrated systems.

"The pit is just one of a warhead's many components," he says. "Components such as high explosives and organic materials also require investigation so we can understand how aging affects their performance and stability as well."

The aging assessments of plutonium continue, as scientists track the properties of naturally and artificially aged samples. Additional data and analysis will allow them to refine minimum lifetime estimates for each stockpile system. In the meantime, experts can make more-informed decisions about America's nuclear forces and NNSA's future complex.

—Arnie Heller

Key Words: actinides, Advanced Photon Source (APS), alpha decay, diamond anvil cell (DAC), Global Nuclear Energy Partnership, helium, Joint Actinide Shock Physics Experimental Research (JASPER) Facility, nuclear power, plutonium, Plutonium Futures Conference, stockpile stewardship, uranium.

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