Plutonium Up Close . . . Way Close

STOCKPILE Stewardship, the Department of Energy's program for assuring the long-term safety and performance of the nuclear weapons stockpile without underground testing, has created a heightened focus on better understanding plutonium.

At Lawrence Livermore, a number of experiments are under way to measure the structural, electrical, and chemical properties of plutonium and its alloys and to determine how these materials change over time. The measurements will enable scientists to better model and predict plutonium's long-term behavior in the aging stockpile (see "Inside the Superblock" beginning on p. 4 of this issue).

Plutonium's Peculiarities

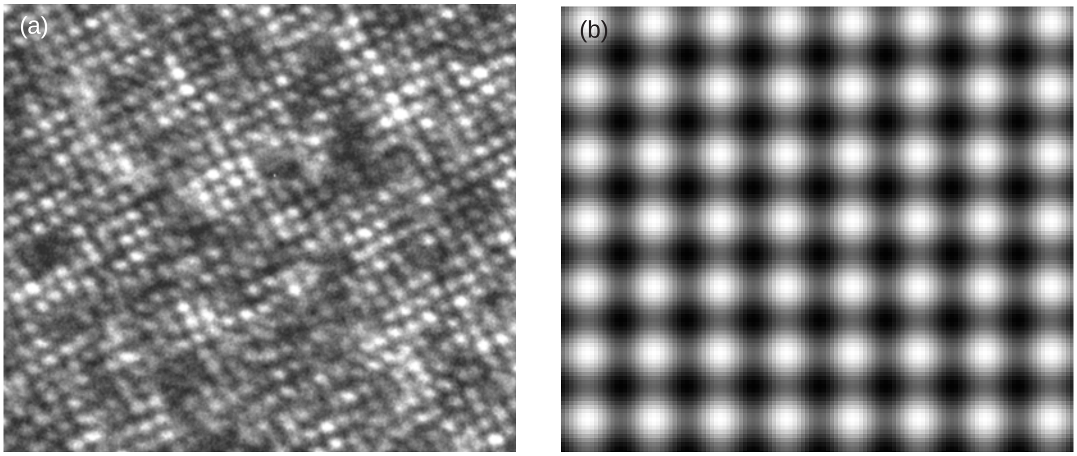
"Plutonium is a complex and perplexing element," notes metallurgist Adam Schwartz. "For instance, plutonium has seven temperature-dependent solid phases—more than any other element in the periodic table. Each phase possesses a different density and volume and has its own characteristics. Alloys are even more complex; you can have multiple phases present in a sample at any given time."

Because plutonium is so complex, surrogate materials cannot give a complete picture of plutonium's characteristics. With the importance of stockpile stewardship, the Laboratory has seen a resurgence of interest and research in plutonium and the other actinide elements (see *S&TR*, June 2000, pp. 15–22). One area that Schwartz, microscopist Mark Wall, and physicist Bill Wolfer are pursuing as part of their stockpile stewardship responsibilities is the evolution of damage to plutonium's structure. As with the atoms of all metals, plutonium atoms form structures on scales as small as a billionth of a meter. These microstructures are constantly changing because of plutonium's radioactive nature. When an atom of plutonium-239 (the isotope of plutonium used in nuclear weapons) decays, it splits into an alpha particle—a helium nucleus with two protons and two neutrons—and an atom of uranium-235. The heavy uranium atom recoils, displacing other plutonium atoms and disrupting the surrounding microstructure. Scientists are concerned that the buildup of gaseous helium atoms combined with other elements in the weapon's environment might gradually change the properties of the plutonium metal.

Seeing Beneath the Surface

To better understand the basic nature of this complex metal and search out the long-term effects of the weapon environment, scientists must know what goes on at the atomic level. To aid this endeavor, the Laboratory acquired a 300-kiloelectronvolt, field-emission transmission electron microscope (TEM) about one year ago. This remarkable instrument uses electrons instead of light waves to "see," so features can be resolved, or viewed at the atomic scale. Where most microscopes can only probe the surface of materials, a TEM looks directly at the internal structure of materials, explains Wall.

Microscopist Mark Wall uses the transmission electron microscope to image the microstructure of plutonium.



(a) An atomic resolution image of plutonium. Such an image was created for the first time ever by the team studying plutonium properties with a transmission electron microscope. (b) A high-resolution computed image of plutonium's atomic structure.

The Inside Scoop with the Transmission Electron Microscope

According to Mark Wall, the new 300-kiloelectronvolt transmission electron microscope (TEM) leased by the Laboratory is the best of its kind in DOE's weapon complex. "Having a high accelerating voltage allows us to see through thicker specimens, facilitating more microstructural observations and better image resolution," says Wall.

The TEM is used to characterize the internal structure of a wide variety of materials, not just plutonium. It not only can image the microstructure directly, but can also identify the phases present in a specimen. The TEM characterization techniques are cataloged here under headings that describe what they do (although there is some overlap among the techniques):

Characterization of Atomic Structure

High-Resolution Atomic Structure Imaging: Directly resolves the atomic structure of crystalline materials down to individual columns of atoms.

Characterization of Microstructure, Defects, and Phases

Bright Field: Images the internal microstructure of materials, including grain and defect structures such as dislocations and voids. Can also be used to observe precipitates or inclusions.

Dark Field and Weak Beam: Allows researchers to link diffraction information with specific phase regions in the sample. Weak-beam imaging is dark-field imaging at higher resolution and is primarily used for imaging closely spaced defect structures on the nanometer scale.

Electron Diffraction (Selected Area Diffraction) and Microdiffraction: Both techniques help researchers identify internal crystal structures. Selected area diffraction allows researchers to view and record the electron diffraction pattern from selected areas as small as 0.5 micrometer. Microdiffraction allows analysis of regions as small as 1 nanometer.

Convergent Beam Electron Diffraction: Reveals diffraction details that provide additional three-dimensional crystallographic and symmetry information.

Lorentz Microscopy: Images directional variations in the magnetic field within thin samples.

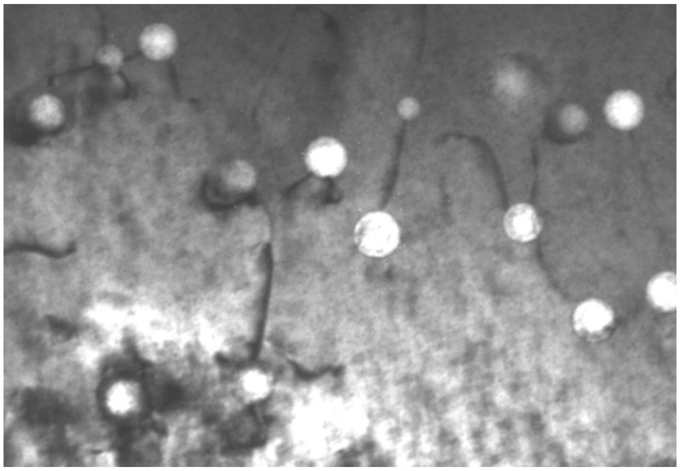
In Situ Microscopy: Allows researchers to record the evolution of a material's microstructure during heating, cooling, and mechanical deformation.

Characterization of Chemical Composition and Impurities

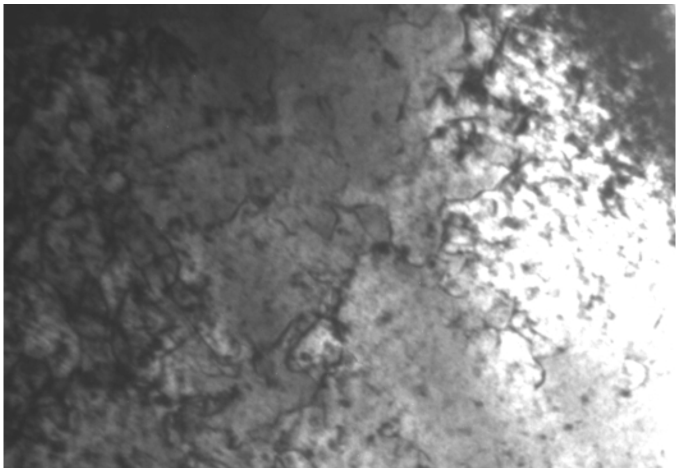
Energy-Dispersive Spectroscopy: Produces x-ray spectra that reveal the presence and amount of elements (for carbon and heavier elements).

Parallel Electron Energy Loss Spectroscopy: Complements energy dispersive spectroscopy, in that it is more sensitive to light elements, including lithium and heavier elements.

*Energy-Filtered Transmission Electron Microscop*e: Acquires real-time, quantitative chemical "maps" of a specific region with a resolution as small as 1 nanometer.



Voids or bubbles could be created by recoiling uranium nuclei and gaseous helium from alpha particles that result from plutonium decay. Here, an aged sample has been intentionally annealed to create bubbles.



A dislocation—an extra half plane of atoms—in the plutonium structure can create sinks or sources for radiation damage.

The primary strength of the instrument is that it can provide detailed characterization simultaneously over many length scales and at high resolution—from hundreds of micrometers to nanometers—and do this in either imaging, spectroscopic, or diffraction modes (see box on p. 24). “In principle, we can observe and measure the defects and composition of microstructural features in these materials down to the nanometer level,” says Wall.

Schwartz and Wall start with plutonium samples measuring less than 3 millimeters in diameter and 150 micrometers thick. They then use special sample preparation techniques to thin each sample until it is transparent to high-energy electrons, that is, to between 10 to 100 nanometers in thickness. The specimens are then vacuum-transferred to the TEM for characterization experiments. The resulting electron micrographs reveal in unprecedented detail the nature of the material and any defects in it. During this work, Schwartz and Wall produced the first-ever image of plutonium at the atomic level.

Using samples of plutonium from old, disassembled nuclear warheads and comparing their resulting micrographs to those from newly cast plutonium, the researchers can better determine the kinds and amounts of defects and changes that occur over time. In particular, they look for voids or bubbles created by recoiling uranium nuclei and the gaseous helium from alpha particles. An example from an old material annealed to intentionally form bubbles is shown in the image directly above. Dislocations—which can be described as an extra half plane of atoms—can create sinks or sources for radiation damage (see image above right).

So Far, So Good

To date, the news for the stockpile is encouraging. Schwartz sums up the results as “So far, so good. We haven’t seen any issues or surprises with the pit samples we’ve viewed.” Last year, the team began another project, looking at special plutonium alloys that have been prepared to accelerate the rate of aging. For Livermore’s Enhanced Surveillance Program (see *S&TR*, September 1999, pp. 3–11), scientists have made several alloys spiked with plutonium-238, which decays much faster than plutonium-239, to try to understand what will happen with stockpiled plutonium as it ages.

Schwartz and Wall also plan to conduct in situ microscopy of plutonium. Heating plutonium samples up to 400°C will allow researchers to see helium bubbles nucleate and for the first time see the early stages of nucleation. “In essence, we’ll be speeding up the kinetics of the material and increasing the diffusion rate,” said Schwartz.

—Ann Parker

Key Words: plutonium research, stockpile stewardship, transmission electron microscope.

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