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SCIENCE-BASED STOCKPILE STEWARDSHIP

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For the past half century, the ultimate military deterrent for the US has depended on its stockpile of nuclear weapons. Because of technical, military, and political considerations, the decision was made nearly one decade ago, during the administration of President George Bush, that no new designs of nuclear weapons are needed and no nuclear-explosion testing is required for the foreseeable future. (Such nuclear tests do not include experiments that are either nonexplosive—using a pulsed reactor, for example—or subcritical, involving no self-sustaining nuclear reaction.)

The 1992 moratorium was reinforced by President Clinton's signing of the Comprehensive Test Ban Treaty (CTBT) in 1996 (see the article by Jeremiah D. Sullivan in PHYSICS TODAY, March 1998, page 24). The CTBT expands the testing moratorium into an international, legal, and verifiable ban on testing, and serves as part of the US commitment toward the international Non-Proliferation Treaty.[1] However, the CTBT has not yet been ratified by the US (or by some of the other recognized nuclear-capable states)[2] and a central issue in the US ratification debates has been how well the nuclear-weapons stockpile can be sustained without testing.

With no new designs planned, the overriding concern is the state of the aging weapons in the enduring stockpile. Aging of materials can cause changes in properties or in critical dimensions, such that a nuclear weapon would no longer function as designed or may not function at all. Figure 1 shows the number of defects per weapon as a function of weapon age.[3] A purely statistical approach shows that defects in nuclear weapons are historically found to accumulate at a rate of less than 1% per quarter century, implying a characteristic lifetime of more than two millennia for these weapons. The low rate at which defects appear suggests that a focused program of surveillance and refurbishment ought to be adequate for maintaining the stockpile throughout the foreseeable future.

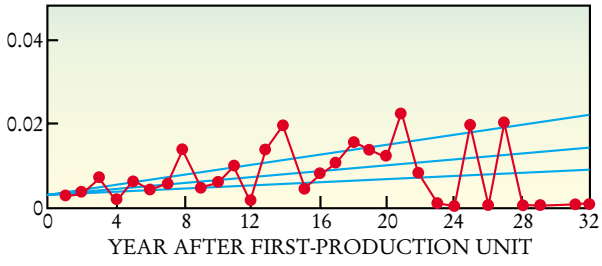

FIGURE 1. STATISTICAL FAILURE RATE of aging US nuclear weapons, shown as number of defects found per weapon investigated, plotted as a function of weapon age. The central blue line represents a fit to the data, and the lines above and below it show 90% confidence limits for the fit. The linear fit indicates that the statistical probability of finding a flaw is 0.26% in new weapons, and rises to 1% after 20 years. Defects due to all causes (not just aging) are included, so the data give a conservative view of how the stockpile ages. Analysis of R. Schwitters and R. L. Garwin, reported in ref. 3.

However, such an approach contains no scientific information about the weapons themselves, their materials or processes, and amounts to assuming that the future is a direct extrapolation of past history. It does not account for the possibility that incremental changes might accumulate nonlinearly, and that failure can occur suddenly. Consequently, it must be augmented with a scientifically-based approach, by which the aging and degradation processes are sufficiently understood to be sensitively monitored. The key requirement is that signs of degradation must be found with enough forewarning that the necessary repair or remanufacturing can be done without jeopardizing the deterrent.

To put the challenge into perspective, a modern thermonuclear weapon contains approximately 6000 parts, including electrical, mechanical, and explosive components as well as the "nuclear package" that creates the fission and fusion explosion. (See the figure on page 26.) It has been said that stewardship with no nuclear testing is similar to guaranteeing that an automobile left in storage for decades would be fully operable, in a suitable emergency, at a moment's notice. Still, most of a weapon's components reside outside the nuclear package, and can therefore be fully checked without nuclear testing; indeed, they are, for the most part, not exercised in underground tests.

Underground testing is intended to verify that the nuclear package works as designed, and is not really a test of an entire weapon system. The great majority of the 1000-plus US nuclear tests were focused on developing new designs, including studies of weapons physics and documentation of military effects; only a small fraction were stockpile confidence tests, meant to test the performance of deployed weapons.[4] The necessarily low number of tests and their destructive nature limit the value of underground testing for monitoring the ongoing safety and reliability of the nuclear arsenal. Based solely on technical considerations, a better way to monitor the state of the enduring stockpile is through a program that combines basic research to identify how the components of a weapon can age with sophisticated surveillance of the weapons.

The Department of Energy's Stockpile Stewardship Program (SSP)[5] is the scientific and engineering effort to

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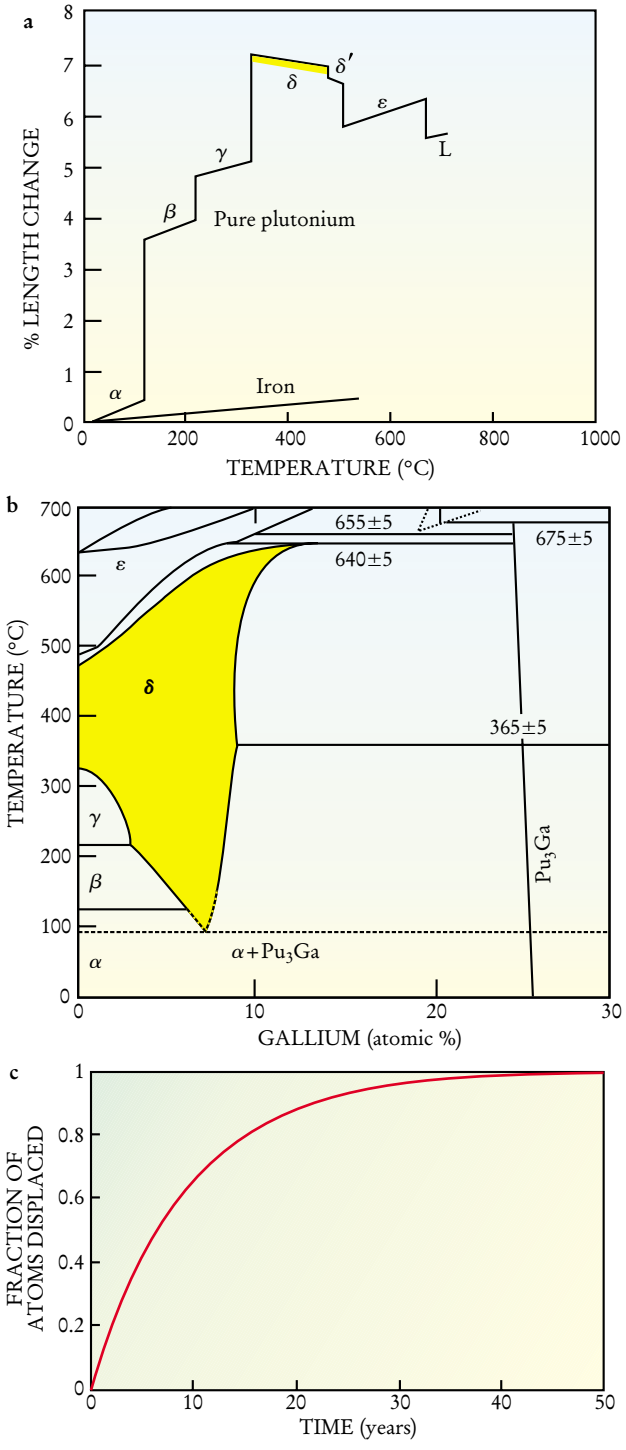

FIGURE 2. COMPLEX PROPERTIES OF PLUTONIUM. (**a**) Large expansion of pure Pu as a function of temperature, with several corresponding phase changes.[7] The more typical expansion of iron is also shown. (**b**) Strong influence of alloying (here with gallium) on phase stability.[8] Compared to pure Pu, a modest increase in Ga fraction results in much wider temperature range for the stable δ phase (the region shaded in yellow). (**c**) Effects of self-irradiation, shown by the expected fraction of atoms displaced as a function of time for a Pu-239-rich sample, assuming no annealing or other crystal-restoring processes.

maintain the US nuclear deterrent in the present era of no underground testing. The program has three essential parts: 1) monitoring of the weapons in the enduring stockpile, 2) repair and remanufacture of components to remedy any degradation observed in surveillance, and 3) basic research to identify what happens in the aging process and to ensure that any refurbishments are adequate and appropriate. For more details about the structure and operation of the SSP, see the box on page 48.

Nuclear weapon architecture

A modern thermonuclear weapon consists of the nuclear package, made up of primary and secondary stages, and such nonnuclear components as the electrical system; the arming, fuzing, and firing system; and a variety of safety interlocks.[4] (The full weapon system includes additional parts, such as the guidance systems, that lie outside the warhead.) Some of the greatest concerns about aging of the stockpile involve the fate of the components of the primary stage.

During weapon operation, a chemical explosion triggers the primary stage by compressing a core, or pit, containing plutonium-239 until it reaches critical density. The resulting fission yield is "boosted" by the presence of a deuterium–tritium mixture that is driven to fusion by the imploding Pu.[6] The combined fission and fusion yield of the primary stage initiates the secondary, or main, stage, which may consist of fissionable materials, such as uranium-235, as well as fusionable materials. It is the secondary stage that provides the bulk of the weapon's military yield, but the secondary requires a minimum energy from the primary to ignite. Thus, a vital index of the reliability of the nuclear package is the performance margin, which is the difference between the minimum yield obtained from the primary and the yield required to drive the secondary. One concern about aging is that the materials within the primary stage might deteriorate with time, so that the performance margin could vanish or even become negative.

Limited-life components such as those containing tritium (β-decay halflife of 12.3 years) must be replaced at regular intervals, but this replacement is planned for in the weapon design, and such components are typically located outside the sealed radiation case that encloses the nuclear package. Of greater concern is possible long-term deterioration of permanent components such as Pu and chemical high explosives, because the primary yield can depend sensitively on the performance and the physical properties of these materials.

Primary-stage materials

One of the greatest concerns about aging of the stockpile regards the fate of the Pu that is a key component of the primary stage. A significant factor is that Pu exhibits complex and highly variable properties. This is largely because its 5f electrons are delicately balanced between being itinerant and localized, as can be inferred by examining the variations in atomic radii and other properties of elements near Pu in the periodic table (compare the elements between thorium and neptunium, lighter than Pu, to the heavier elements between americium and einsteinium).[7] As shown in figure 2a, for example, solid Pu undergoes several phase transitions and expands by more than 20% in volume when heated from 0°C to 400°C. This anomalous sensitivity to temperature is matched by a sensitivity to alloying with certain elements, including gallium, aluminum, and Am; figure 2b shows how small amounts of Ga can stabilize the low-density δ phase so that it appears at near-ambient temperatures.[7,8]

If the complexities of the material weren't enough,

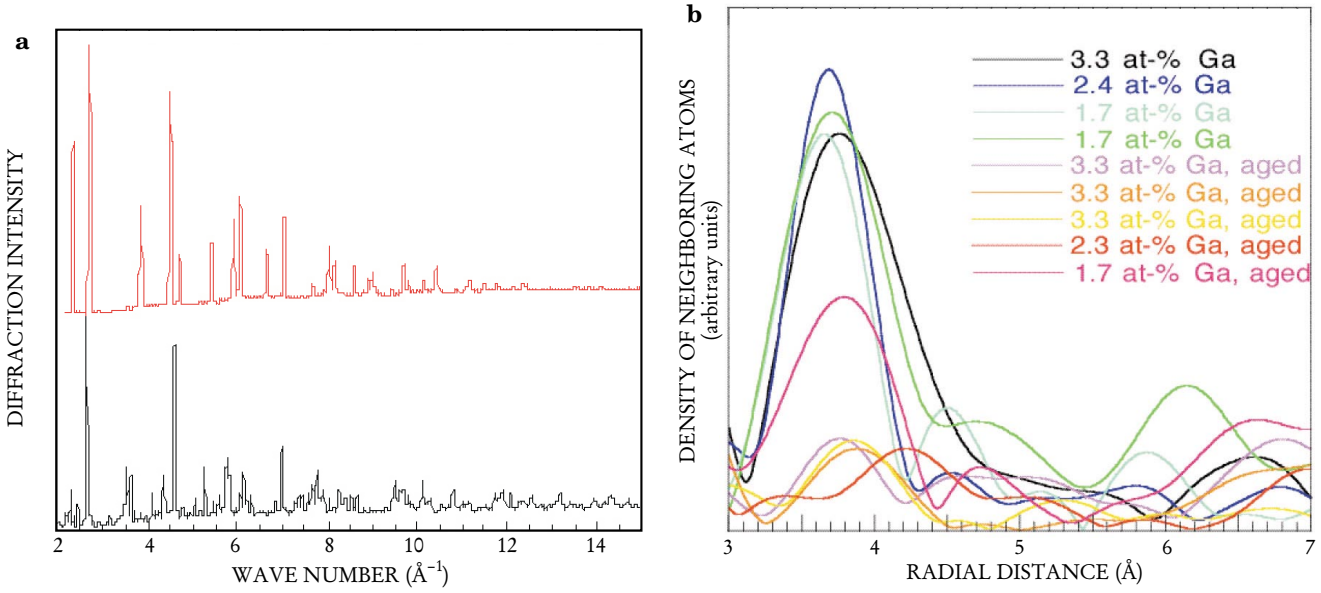

FIGURE 3. PLUTONIUM CRYSTAL STRUCTURE persists and actually gets more regular with aging, as shown by various measurements. **(a)** Synchrotron-based x-ray diffraction patterns of the α (black) and δ (red) phases in Pu samples that are more than 30 years old have peaks that indicate an ordered structure.[10] **(b)** Radial distribution functions of new and aged δ-phase Pu alloys obtained from Pu L_{III} x-ray absorption fine structure (XAFS) measurements show an anomalous peak at 3.8 Å in the new samples that shrinks with age. (For clarity, the contribution from the ideal face-centered cubic structure was subtracted from the plots.) In some cases, data are shown for multiple samples with the same gallium fraction. Evidently, the aged samples have a more nearly ideal crystal structure than the new sample.[10]

the Pu-239 used in weapons also undergoes α decay with a halflife of 24 400 years. In addition to the accumulation of helium from the α particles stopped within the lattice, an estimated 2500 Frenkel-pair defects are generated by the uranium nucleus emitted in each decay. As shown in figure 2c, the crystal structure is thus disordered at a rate of about 10% of the atoms being displaced per year.[9] Is it possible that self-irradiation could transform the Pu within aging weapons, rendering the weapons ineffective (and also potentially unsafe for storage and handling)? At present, this question is best answered experimentally. Various probes are available for assessing whether and to what degree the Pu crystal structure may be disordered, and these have been used to compare Pu samples from old weapons with new Pu samples. There are also methods available for experimentally simulating the aging process over a shortened time scale; one way is to spike samples with Pu-238 (halflife 86 years).

Perhaps the most important result from measurements is that Pu exhibits good crystalline order even after decades of aging. A good illustration of the long-term preservation of order comes from high-resolution x-ray studies of Pu more than 30 years old, illustrated in figure 3a. These studies have shown peaks in the diffraction intensity, indicating the presence of periodic structure, to high magnitudes of the scattering vector. The preservation of crystal structure despite displacements and other damage reflects an apparent self-annealing of the Pu. In the x-ray diffraction studies, both the α and δ phases of Pu show this self-annealing behavior, suggesting that it is intrinsic to Pu rather than being limited to a single structural configuration.

Nevertheless, changes have been observed in the crystal structure ordering of Pu after aging for periods of a decade or longer. One such change is, surprisingly, an increase in crystal ordering with time. Observations using x-ray absorption fine structure (XAFS), illustrated in figure 3b, show that newly-made Pu in the δ phase exhibits deviations from the ideal face-centered cubic (fcc) structure, as indicated by an additional peak in the radial distribution function at about 3.8 Å. The extra peak might be attributed to local body-centered cubic arrangements within the fcc lattice, but high-resolution x-ray diffraction measurements by Steven Conradson and colleagues at Los Alamos National Laboratory have indicated a long-range order suggesting that a better explanation would be an fcc modulation with a different periodicity from the lattice.[10] XAFS observations are sensitive to the atomic packing configuration because core electrons liberated by the absorbed photon are scattered by neighboring atoms, modulating the absorption coefficient as a function of energy. In this case, observations were made using x-ray energies sufficient to liberate 2p core electrons.

The most striking result of these studies is that the local deviation from the ideal fcc structure vanishes with aging, disappearing sooner in samples having a higher Ga content. It is not surprising that the most gallium-poor samples would retain the most structural nonideality over time, because Ga is known to stabilize the δ phase (figure 2b); on the nanometer scale, aging appears to have the same effect as a greater Ga concentration, in that it shifts the Pu to a more stable configuration.

Another diagnostic tool is high-resolution transmission electron microscopy, which has been used by Jeffrey Kass and colleagues at Lawrence Livermore National Laboratory to observe the evolution of microstructures and the accumulation of He within the Pu.[11] The overall finding from a variety of observations, including detailed electrical-resistivity measurements as a function of temperature (which are sensitive to the density and distribution of defects present), positron annihilation spectroscopy, and other studies, is that the Pu samples not only retain long-range order but actually get closer to the ideal crystal structure with increasing age. Annealing processes, perhaps related to those countering the crystal-structure disordering, appear to counteract radiation-

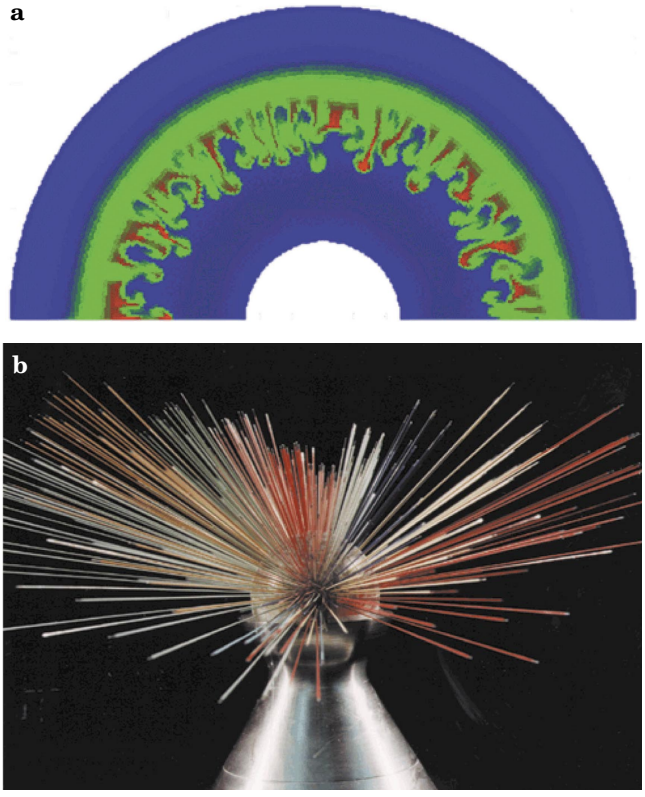

FIGURE 4. HYDRODYNAMICS STUDIES combine numerical simulation with detailed experimental measurements. **(a)** Results of a numerical simulation of an implosion of an inertial-confinement fusion capsule show the development of a Richtmeyer–Meshkov instability.[12] **(b)** The "pin dome" array of fiber-optic detectors used to experimentally measure surface velocity and symmetry of an explosive-driven implosion.[13]

induced damage and mitigate the initial buildup of He quite effectively, at least for Pu in the US stockpile.

In addition to Pu, there have been concerns about aging of the high explosive that initiates the compression of the primary stage. The organic polymers making up the high explosive are known to change over time, with measurable evolution of properties over the first several months followed by slower changes in succeeding years. Over time, volatile constituents are lost and changes have been observed in high-explosive crystallinity and molecular weight. Surprisingly, however, the high explosive used in US weapons has been found to improve systematically with age in key measures of performance, such as yielding characteristics and detonation-front velocities.

Thus, crucial primary-stage components that were initially subject to concern have been shown through the SSP to be robust as they age. Indeed, there is now consensus among specialists that the Pu pits in the US stockpile are stable over periods of at least 50–60 years, with the most recent studies suggesting a far longer period. More important than the indications of benign aging is the demonstration that the materials are now becoming understood in sufficient detail, and surveillance methods are becoming sensitive enough, to ensure that any signs of degradation will be observed in time to apply the necessary repairs or refurbishment.

Numerical and experimental simulation

Subtle changes in material properties can, in principle, affect the dynamics of the primary-stage implosion. The boosting process must be efficient enough to create the yield required to drive the secondary stage to ignition; without proper functioning of the primary stage, the weapon can fail altogether. As long as the primary releases sufficient energy, however, the secondary is driven to full or nearly full yield under a wide range of conditions.

Because of its importance, much attention has been paid to understanding the detailed hydrodynamics associated with the initiation, implosion, and boosting of the primary stage. These processes can be sensitive to changes in material density, acoustic and shockwave velocities, dynamic yielding, and ejecta formation. Ejecta formation, in particular, can be influenced by such minor-seeming alterations as changes in grain size and texture or variations in trace-element concentrations. It is essential to identify those material characteristics that can have the most dramatic effects on the primary-stage dynamics and investigate how they are likely to change, either because of aging or because a component is refurbished in a slightly different manner from the original.

The study of weapons dynamics involves a tight linkage between laboratory experiments and computer simulations. The installation of multi-teraflop computers at the weapons laboratories has allowed the development of far more sophisticated numerical codes than had previously been possible. Codes under development include reliable, three-dimensional simulations of hydrodynamic instabilities that aim to give insight into why a small variation in material properties might, or might not, make a difference to performance. Validation of the numerical simulations includes comparisons with experiments involving physical processes similar to those found in the weapons, although sometimes for very different geometries or time scales (figure 4a).[12]

Laboratory studies of elastic properties, dynamic equations of state, and ejecta formation provide direct input to the numerical simulations. Some of these studies are subcritical experiments involving fissionable materials and may be conducted underground at the Nevada Test Site. Hydrodynamic and neutronic simulations are performed before the tests, and the predictions are compared with in-situ measurements during each experiment. In addition to validating the simulation predictions, these measurements also verify that the experiments are truly subcritical. Providing more open access to this level of monitoring, at US and other test sites, could enhance confidence among parties regarding subcritical experiments under a CTBT regime.

Direct measurements using mechanical diagnostics (figure 4b)[13] as well as radiographic diagnostics (such as at the Dual-Axis Radiographic Hydrodynamic Test facility described in the box on page 48) provide detailed tracking of implosion dynamics right up to the onset of criticality, and are important checks on the reliability of the numerical simulations. Archival records from past nuclear-explosion tests are also being reanalyzed with modern computational resources. Clearly, computer simulation plays a special role in integrating the wide array of experimental, surveillance, archival-analysis, and modeling activities of the SSP.

Chemical corrosion

Given the complexity of design and the corrosive materials involved with nuclear weapons, it is not surprising that the surveillance and research programs have indeed uncovered defects. In particular, corrosion can take place inside the nuclear package, most typically involving the reaction of hydrogen with various metals. Other than in

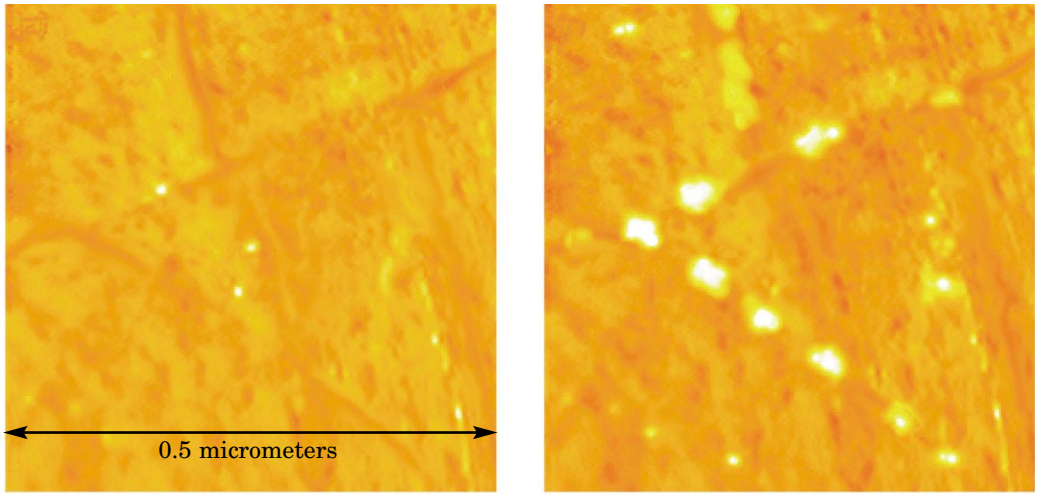

FIGURE 5. CORROSION of uranium surfaces can be seen in atomic force microscopy images both **(a)** before and **(b)** after exposure to hydrogen. The right image shows pitting (light-colored regions) with the formation of uranium hydride along the grain boundaries.[14]

the boost-gas system, hydrogen and its isotopes should not be present as free species within the nuclear package. Nevertheless, hydrogen can evolve from the various organic compounds and hydrogen-bearing salts found within the radiation case. In addition, improper heat treatment or poor sealing of components can leave enough humidity to cause problems years later. Unlike the examples of deterioration previously described, these issues have arisen in the secondary as well as primary stages.

As shown in figure 5, the formation of metal hydrides typically begins at surfaces or interfaces.[14] A certain level of corrosion can be tolerated, but in extreme instances some components have required remanufacture. Good design and production practices, and the judicious use of seals and "getters" (compounds that soak up and immobilize hydrogen), have proven to be effective in counteracting this form of chemical corrosion.

Basic science

Although its applied mission of preserving the physical stockpile is necessarily its primary objective, the SSP also produces major contributions to basic research in a wide range of activities. A case in point is recent work on extending the equation of state of hydrogen isotopes into the terapascal regime. (1 TPa is 10 Mbar, or roughly 10^7 atm pressure.) Such work is certainly relevant to understanding weapons physics (the onset of fusion), but there are also other scientific applications, such as modeling the interiors of stars and giant planets (of which hydrogen is the predominant component).[15,16]

The Stockpile Stewardship Program

The SSP is a nearly $5 billion per year program with the goal of maintaining the robustness of the US nuclear deterrent without resorting to underground testing (see PHYSICS TODAY, March 1997, page 63). The program involves the national laboratories—Lawrence Livermore (LLNL) and Los Alamos (LANL) national laboratories have responsibility for the nuclear packages, and Sandia National Laboratory (SNL) has responsibility for the nonnuclear components—as well as nuclear production plants and other sites—notably the Kansas City, Pantex, Savannah River, and Y-12 plants, and the Nevada Test Site. (Manufacturing of key components is also undertaken at LANL and SNL.)

With or without a nuclear testing ban, some form of stockpile surveillance and maintenance would be required. The particular task of the SSP is to carry out that mission without testing. On an immediate basis, the SSP is challenged each year by the need to assure that the secretaries of defense and energy can certify to the president that each weapons type in the nuclear stockpile is safe, reliable, and effective.

The activities of the SSP are defined along three broad lines: stewardship campaigns, including the underlying scientific and engineering work; directed stockpile work, which comprises the work being done on each specific weapons type; and infrastructure, which includes major facilities and related efforts. Because research and surveillance make no sense unless there is a means of repairing defects or deterioration, another crucial aspect of the SSP is sustaining a capability to manufacture weapons components.

The major campaigns are of three basic types: primary-stage certification, secondary-stage certification, and certification of nonnuclear components. Surveillance and remanufacturing (or repair) are among the most prominent activities under each of these campaign efforts. Directed stockpile work describes application of these campaigns to the specific weapons in the enduring stockpile. Infrastructure includes at least four major initiatives that are considered long-term investments for the research, surveillance, and refurbishment activities. Of these, the National Ignition Facility (NIF) at LLNL is the facility perhaps most directly tied to basic research. NIF, when operational, will investigate the properties of materials at high compression and study the onset of inertial confinement fusion. The hydrodynamics and radiography initiative is intended to experimentally characterize the mechanical implosion of the primary stage of a nuclear weapon, right up to the onset of criticality; this initiative includes the Dual-Axis Radiographic Hydrodynamic Test (DARHT) Facility at LANL. The Accelerated Strategic Computing Initiative (ASCI) encompasses state-of-the-art computer simulation, and serves as a basis for integrating past experience (such as that from underground tests), surveillance findings, and the results obtained from new research. The Advanced Design and Production Technologies (ADAPT) program initiative modernizes manufacturing and repair capabilities at production plants; it involves both engineering work and computer simulation.

Facilities developed for the infrastructure initiatives (and others being considered for the future) are planned to provide integration across the various campaigns. For example, NIF is applicable to both primary and secondary certification, DARHT is relevant to primary and nonnuclear component certification, and both ASCI and ADAPT apply directly to all three major campaigns. It is their cross-cutting nature, rather than their magnitude (or expense), that distinguishes these efforts for the SSP.

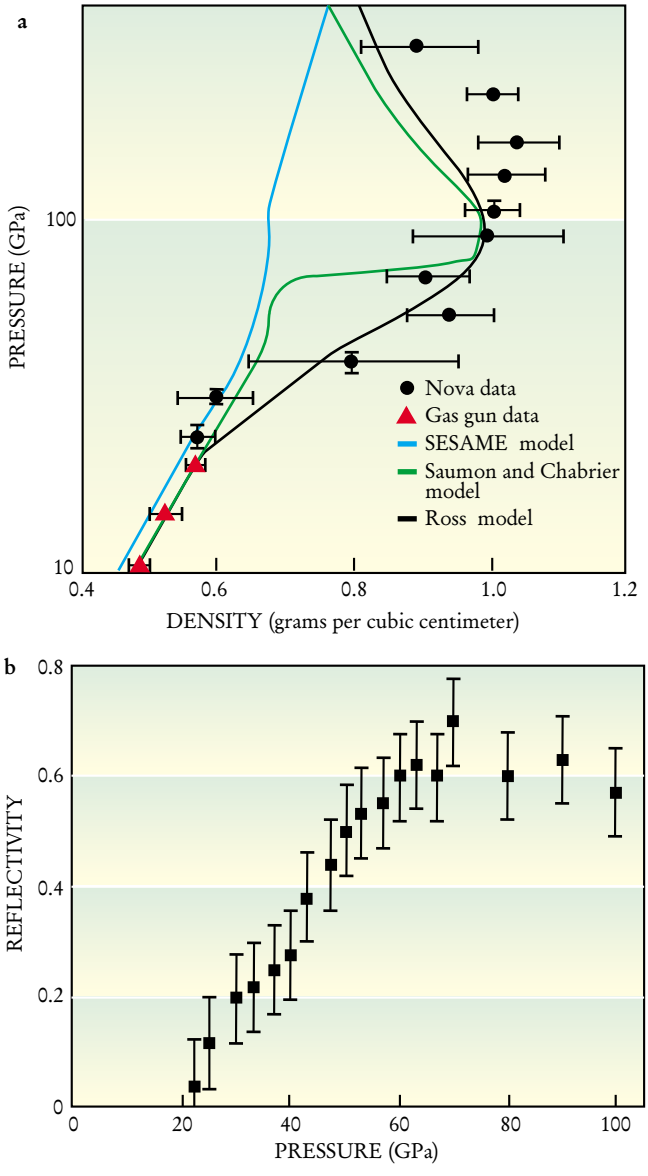

FIGURE 6. HIGH-PRESSURE DEUTERIUM MEASUREMENTS may reveal the emergence of a long-predicted metallic state. Laser-driven shock experiments conducted in conjunction with National Ignition Facility development have obtained **(a)** new equation-of-state data (black circles) and **(b)** new reflectivity measurements. Shown for comparison are predictions of several equation-of-state models (solid lines); the SESAME model (blue line) represents the best prediction based on earlier data. Also shown are mechanical shock measurements (red triangles), which are in good agreement with comparable laser measurements. The large increase in density at high pressure and the corresponding enhanced reflectivity above 60 GPa together point to the onset of a dense metallic state in fluid deuterium at temperatures of several thousand kelvin.[18]

Nominally the simplest element in the periodic table, hydrogen provides a rich testing ground for both theory and experiment at high pressures (see the article by Russell J. Hemley and Neil W. Ashcroft in PHYSICS TODAY, August 1998, page 26). For example, the 1935 prediction of Eugene Wigner and Hillard B. Huntington that hydrogen becomes metallic under compression has provoked much interest, but there has been remarkable difficulty in confirming either the conditions or the mechanism associated with metallization. Static compression experiments with diamond anvil cells have yet to document a metallic state at pressures above 200 GPa, which is well within the range of several theoretical predictions (see the Hemley and Ashcroft article). However, recent dynamic-compression studies on hydrogen do appear to have demonstrated the creation of a dense metallic state.

As part of the preparation for the National Ignition Facility, and as a continuation of pioneering light-gas gun measurements,[17] laser-driven shock experiments were used to obtain the high-temperature equation of state of deuterium; some results are shown in figure 6a.[18] The experiments indicate that deuterium at 100 GPa is more than 50% denser than had previously been thought. Because the technique is relatively new, the measurement uncertainties are large, but the discrepancy between the new observations and earlier predictions is still significant. Other measurements made in similar experiments reveal a sharp increase in optical reflectivity at pressures approaching 60 GPa (see figure 6b), giving further evidence for a metallic state. The laser-shock equation-of-state results agree well with prior measurements using mechanical shock loading (gas-gun data, shown as red triangles in figure 6a), where the conditions of measurement overlap.[16,17]

Why has the metallic state been detected with the dynamic shock-compression experiments and not with the static diamond anvil experiments? The difference between the results can most likely be ascribed to temperature: The static measurements have been conducted almost exclusively at and below room temperature, whereas the dynamic measurements were made at temperatures of several thousand kelvin. Apparently, both temperature and density (or pressure) play important roles in determining the onset of hydrogen metallization.

Much of the basic science conducted within the SSP is completely unclassified. For this reason, SSP basic research programs have benefited from close collaborations between researchers inside the national laboratories and those in academia, both within the US and abroad. Laser-based research provides good examples of international cooperation, and the computer-simulation effort (the Accelerated Strategic Computing Initiative, or ASCI) explicitly involves collaborations between universities and the national laboratories. Only through such close links with the open scientific community can the technical excellence of the SSP, and its participants, be ensured over the long term.

Broader issues

The SSP is not only challenging, it also represents a departure from the way that the US previously maintained its nuclear weapons. When new designs were continuously being developed, existing weapons (including those that might be flawed) could simply be replaced by new designs. It is therefore understandable that those having responsibility for defending the nation could initially feel concern about the reliability of the stockpile under a testing moratorium (such as currently exists) or a CTBT.

Certain critics have expressed the view that the SSP is a means of perpetuating the nuclear danger, and that it would be best to let the stockpile degrade and its stewards turn to other activities. However, as long as the military and political decision within the US is to rely on a nuclear deterrent, the worst option is to allow the stockpile to become unsafe or unreliable. This is especially so in the light of arms-control initiatives intended to reduce the sizes of nuclear arsenals (such as START II, now ratified by the US and Russia, which is to significantly reduce the

two nations' arsenals, with further prospects for diminished nuclear-weapon stockpiles being contemplated under START III). Maintaining the stockpile also requires maintaining the requisite technical expertise—high-quality scientists and engineers.

In fact, the necessary research and surveillance activities are now well under way and have proven to be an effective means of monitoring and maintaining the nuclear deterrent. The program has built a good foundation for achieving its broader objective—to provide the technical basis that allows the US to pursue the most visionary policies in global and national security, including maintaining the balance between defense and arms control.

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