Lawrence Livermore National Laboratory

October/November 2017

Science & Technology Review

Nuclear Test Film REVITALIZATION

Also in this issue:

About the Cover

The scientific record of the nation’s atmospheric nuclear tests—conducted between 1945 and 1963—survives in aging, deteriorating film reels. These historic films provided important data to contemporaneous scientists and today are offering new insights that support stockpile stewardship. As the article beginning on p. 4 describes, members of Lawrence Livermore’s Film Scanning and Reanalysis Project are part of a team dedicated to preserving these films through innovative scanning technology, customized image-processing techniques, and high-performance computing. As a result, Laboratory scientists have been able to obtain millions of data points. On the cover, images from two tests are superimposed on artist-generated film reels. A typical film reel captured a single test over thousands of frames.

Cover design: Acen Datuin

About *S&TR*

At Lawrence Livermore National Laboratory, we focus on science and technology research to ensure our nation’s security. We also apply that expertise to solve other important national problems in energy, bioscience, and the environment. *Science & Technology Review* is published eight times a year to communicate, to a broad audience, the Laboratory’s scientific and technological accomplishments in fulfilling its primary missions. The publication’s goal is to help readers understand these accomplishments and appreciate their value to the individual citizen, the nation, and the world.

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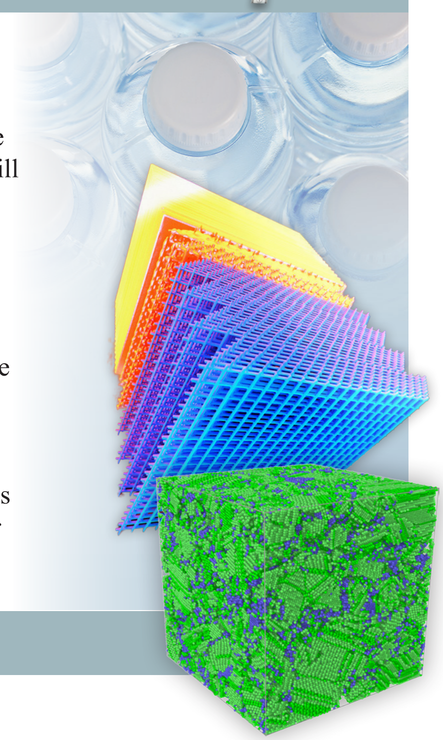
Contents

Feature

Research Highlights



Departments

S&TR Staff

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Getting to the Core of Plate Tectonics

Lawrence Livermore scientist Nathan Simmons and collaborators from the University of Chicago, Université du Québec à Montréal, the University of Florida, Kent State University, Syracuse University, and the University of Texas at Austin have found that heat from Earth's core has a significant effect on tectonic plate movement. This research challenges the theory that movement of Earth's tectonic plates is driven largely by negative buoyancy created as the tectonic plates cool. It also suggests that the underwater mountain ranges, known as mid-ocean ridges (MORs), are not passive boundaries between moving plates, as previously thought.

"Heat from the deep Earth likely plays a significant role in global plate tectonics," says Simmons, a coauthor of the research published in the December 23, 2016, issue of *Science Advances*. "The cooling and sinking of plates is not the only significant plate-driving force." Global-scale tomographic models of Earth's mantle—the zone between Earth's core and crust—were used to simulate mantle convection forward and backward in time. The simulations helped demonstrate the persistence of large-scale mantle upwelling beneath the East Pacific Rise region.

The East Pacific Rise has not significantly moved east–west for 50 to 80 million years, even as parts of it have spread asymmetrically. The researchers say these dynamics cannot be explained solely by subduction. "Through modeling and observations, we found that over the past 80 million years, plate separation along the East Pacific Rise is driven significantly by heat drawn from Earth's core, which is uncharacteristic relative to other MORs," adds Simmons.

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Probing Radiation Defect Dynamics

Researchers at Lawrence Livermore and Texas A&M University used a novel experimental method to study thermally activated defect interaction processes in silicon. Pulsed-ion beams were used to probe defect interaction dynamics. By measuring temperature dependencies of the dynamic annealing rate of defects, the researchers found two distinct regimes of defect interaction—at temperatures above and below 60ºC, respectively. Rate theory modeling, benchmarked against pulsed-beam data, pointed to a crucial role of both vacancy and interstitial diffusion, with the defect production rate limited by the migration and interaction of vacancies.

Understanding radiation defects in crystals has been a major materials challenge for decades. Stable defect formation often involves dynamic processes of migration and interaction of point defects generated by energetic particles. However, the exact pathways of defect formation have remained elusive, and most current predictions of radiation damage are essentially empirical fits to experimental data. This approach applies even to the best studied and arguably simplest material, crystalline silicon, which is the backbone of modern electronics. Until recently, scientists lacked experimental methods that could directly probe the dynamics of defect creation and annealing.

The research, appearing in the January 6, 2017, online edition of *Scientific Reports*, could lead to improvements in modern electronics performance. Sergei Kucheyev, Livermore project lead and coauthor for the paper, says, "This work provides a blueprint for future pulsed-beam studies of radiation defect dynamics in other technologically relevant materials."

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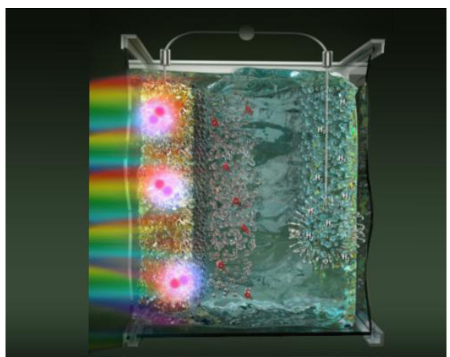
Hydrogen from Photoelectrochemical Cells

Hydrogen production offers a promising approach for producing scalable and sustainable carbon-free energy. The key to a successful solar-to-fuel technology is the design of efficient, long-lasting, and low-cost photoelectrochemical cells (PECs), which are responsible for absorbing sunlight and driving water-splitting reactions (see image at left). Lawrence Livermore scientist Tuan Anh Pham and collaborators from the University of California at Santa Cruz and the University of Chicago are fine-tuning the mechanisms to generate hydrogen from water and sunlight by investigating the interfaces between photoabsorbers, electrolytes, and catalysts in PECs.


(Image courtesy of Peter Allen, University of Chicago.)

Efficient PECs rely on the availability of abundant semiconducting photoelectrode materials that are responsible for absorbing sunlight and driving water-splitting reactions. "Despite steady efforts and some breakthroughs, no single material has yet been found that simultaneously satisfies the efficiency and stability required for the commercialization of PEC hydrogen-production technology," says Pham, lead author of the research appearing in the January 9, 2017, online edition of *Nature Materials*.

With the growing complexity of PEC architectures, understanding the properties of the interfaces is crucial to predicting novel, better performing materials that can eventually lead to optimal device performance. The study addresses the challenges in describing PEC interfaces using first-principles techniques that focus on the interplay between their structural and electronic properties. The researchers also reviewed first-principles techniques relevant to solid and liquid interfaces.

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Commentary by Charles Verdon

Innovative Problem Solving Strengthens Stockpile Stewardship

As *Science & Technology Review* often showcases, Lawrence Livermore's national security mission is deeply rooted in our commitment to the Stockpile Stewardship Program. The National Nuclear Security Administration (NNSA) charges three national laboratories—Lawrence Livermore, Los Alamos, and Sandia—with ensuring the safety, security, and effectiveness of the nation's nuclear stockpile. Our responsibilities include developing and deploying capabilities that predict, assess, and certify nuclear weapons performance in the absence of nuclear testing. Stockpile stewardship efforts have led to important scientific breakthroughs and increased our understanding of thermonuclear processes and high-energy-density (HED) physics. In addition, Livermore's ongoing investments in this area often result in discoveries and developments that benefit other Laboratory programs.

Although stockpile stewardship is a forward-looking endeavor, it requires understanding the past. Founded in 1952, Lawrence Livermore National Laboratory was established during a critical period in U.S. history, when aboveground nuclear weapons testing was under way. The films that captured these experiments have mainly served as archival footage of a bygone era but are now being recognized for their scientific worth. When the films were made, the technology did not exist for completely and accurately measuring blast characteristics such as optical density, fireball size, and other factors necessary for calculating energy yield. Fast-forward to the 21st century, and Livermore's Film Scanning and Reanalysis Project is applying advanced capabilities to extract and analyze data before these films decay beyond usefulness.

As the article beginning on p. 4 describes, the project team had to determine the right combination of technologies and expertise for film scanning, digitization, and analysis. Some solutions (such as open-source software) required customization, while others (such as a specialized film scanner) were completely new to the Laboratory. Livermore's high-performance computing resources have also been applied to manage the millions of high-fidelity data points now available. The team takes a thoughtful, inventive approach to answering time-sensitive questions posed by the films, which contain the only measurable evidence of an isolated experimental period. This information helps improve three-dimensional simulations that predict a device's effects, which in turn increases our confidence in assessing the legacy stockpile and weapons life-extension programs. In addition, the films are captivating reminders of the power of nuclear devices, underscoring the importance of our mission in service to NNSA.

More broadly, the film project epitomizes the Laboratory's approach to any national security challenge—assemble a multidisciplinary team, leverage the best technology, and deliver the right solutions. The highlights in this month's issue illustrate this strategy across a range of fields. The highlight beginning on p. 12 describes a newly developed plutonium reference material, which will enhance nuclear safeguards worldwide. The international collaboration draws on Livermore's analytical expertise and mass spectrometry technologies to ensure isotopic purity. Livermore's work in advancing nanoporous materials science with the development of low-density foams is described in the highlight beginning on p. 16. The Laboratory's additive manufacturing capabilities play a key role in fabricating new parts for HED experiments. The final highlight beginning on p. 20 discusses the Laboratory's work to improve our understanding of physics with experiments that subject matter to extreme conditions. Computer simulations of millions of atoms combined with accelerator experiments help Livermore scientists develop a predictive theory of nonequilibrium transformations.

This year, the Laboratory celebrates its 65th anniversary. As in the past, our current and future missions will require the kind of innovative problem solving that has become a hallmark of this Laboratory. Regardless of the challenge—whether it is studying deteriorating film strips, preparing radioactive isotopes, refining complex simulations, or developing new materials—Livermore scientists and engineers will find a way to address it in service to the nation.

■ Charles Verdon is principal associate director for Weapons and Complex Integration.

PRESERVING the PAST to PROTECT THE FUTURE

Advanced technology revives Cold War–era film reels, providing new data to validate modern nuclear weapons science.

ATMOSPHERIC nuclear weapons testing has an important place in history for its scientific, political, and cultural legacies. Between 1945 and 1963, the United States conducted 210 such tests and captured the events on dozens of cameras. Performed mainly at the Nevada Test Site (now called the Nevada National Security Site) and the Pacific Proving Grounds—a collection of remote locations in the Pacific Ocean, including the Marshall Islands—the tests were designed to explore a range of tactical scenarios, such as detonations at high altitude, on the ground, or over water. Some bombs were dropped directly from aircraft or via parachute. Others were placed atop ground-based towers. Thermonuclear explosions depended on the types of explosives, fuels, and detonation methods used. Testing goals included experimenting with new weapons designs, evaluating weapons reliability and performance, and measuring explosives effects.

The scientific record of the nation's atmospheric nuclear tests survives in aging, deteriorating film reels. Data such as fireball size, shock wave position, and cloud dimensions—all of which can be gleaned through careful frame-by-frame analysis—are critical to Lawrence Livermore and other institutions tasked with stockpile stewardship—ensuring the safety, security, and effectiveness of the U.S. nuclear stockpile. Now, in the post-nuclear-testing era, preserving these decades-old artifacts is a matter of national security. The Film Scanning and Reanalysis Project, a joint effort between Lawrence Livermore and Los Alamos national laboratories, is dedicated to this endeavor.

U.S. atmospheric nuclear tests conducted in the mid-20th century, such as the one shown here from 1955 at the Nevada Test Site, were captured on various film formats. The Film Scanning and Reanalysis Project is using modern scanning technology to digitize these aging films and extract key data with unprecedented accuracy.

Livermore nuclear weapons physicist Greg Spriggs helms a team experienced in film preservation, archiving, image processing, shock wave physics, software development, data analysis, and declassification protocol. Since 2011, the team has combed through secure government vaults to inventory and salvage thousands of film rolls. Under the aegis of the Laboratory's Weapons and Complex Integration Principal Directorate, the team uses modern scanning technology to digitize the films while developing image-processing techniques to extract key data with unprecedented accuracy.

Of the estimated 10,000 atmospheric nuclear test films, Spriggs and colleagues have identified approximately 6,500, scanned 4,200, declassified 750, and analyzed 500. In 2017, for the first time, the public was given access to a series of these films. "We hope viewers appreciate the immense power of these weapons," says Spriggs. "Further, in the absence of live testing, the information from these films helps validate our computer simulations needed for stockpile stewardship."

Specialists to the Rescue

Early in the project, Spriggs knew he needed help sorting through the various film stocks. Film expert Jim Moye, whose résumé includes preserving the Zapruder film (footage of President John F. Kennedy's assassination), joined the team to evaluate the condition of the film reels and digitize the images. Livermore's Maxine Trost and Los Alamos's Alan Carr also stepped in to manage film retrieval and identification in the laboratories' archives. Academy Award–winning filmmaker and documentarian Peter Kuran came aboard to advise on historical film stocks and camera equipment. Understanding film technology of the testing era is crucial to the team's progress toward preservation and analysis. (See the box on p.10.)

Although images captured on black-and-white film tend to remain stable throughout the years, various problems can befall an aging film reel. Decades of handling and transport can cause scratches. Alternatively, some reels were treated with a scratch-protective lacquer that over time can alter the original optical density—that is, the measurement of light absorbance and blockage—resulting in poor data quality. Removing the lacquer can cause further damage. In addition, cellulose acetate film is vulnerable to a condition known as vinegar syndrome, in which the breakdown of acetyl molecular chains produces acetic acid and a strong vinegar smell. Other complications include brittleness, oxidation, and curling.

Films of this age and composition also tend to shrink an average of 1.5 to 2.5 percent, and the team must compensate for this issue to increase the reliability of extracted data. "We measure the horizontal distance between the perforations to calculate the amount of shrinkage," notes Spriggs. Uneven shrinkage can sometimes cause the edges of the film to buckle or flute—conditions in which the edges and center of the film are no longer the same length.

Cleanliness is another important aspect of film preservation. Moye usually cleans individual frames to remove or minimize the effect of dirt, debris, ink, and tape adhesive. "Tape of all kinds was used to mark frames for manual analysis. I've seen tape placed every five frames for a thousand frames," he states. When he encounters torn or folded perforations along the edge of a reel, Moye determines the best repair strategy for each type of defect.

Besides age-related problems, the team must account for anomalies caused by conditions at ground zero. For instance, foggy images could be evidence of radiation exposure or a camera's light leak. Kuran examines each reel to diagnose

Film expert Jim Moye selects one of the thousands of film canisters queued for scanning and analysis. (Photo by Lee Baker.)

In addition to the ravages of time, mid-20th century film reels used for capturing events from atmospheric nuclear tests were subjected to radiation damage. Film crews attempted to protect cameras from radiation exposure with lead and concrete shielding and by positioning them at lower risk distances from ground zero. This reel from the first nuclear weapons test (in New Mexico in 1945) is beyond repair. (Photo by Lee Baker.)

lens flares, refraction effects, static electricity, flaws caused by camera operator error, and degradation from repeated copying. Altogether, the team's preservation activities are necessarily thorough. Kuran states, "Unlike Hollywood movies, you can't remake these films."

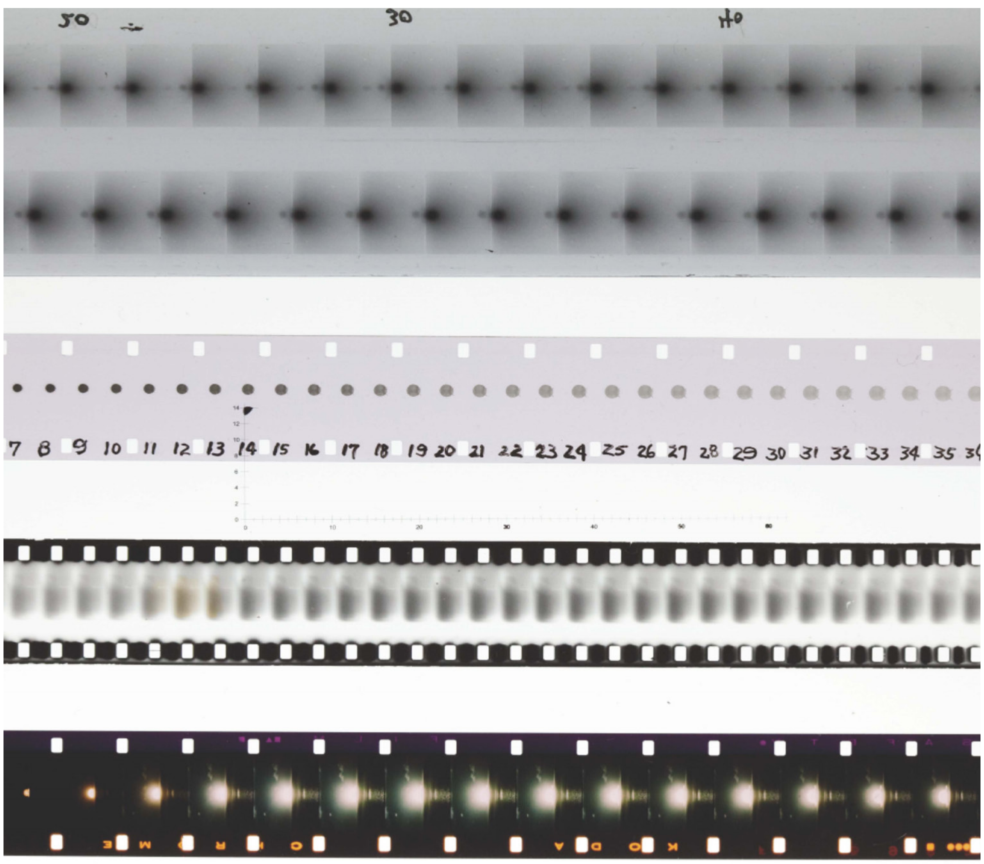
Nuclear Age Meets Digital Age

The most diligent preservation efforts can only slow, not stop, decomposition. "All organic substances, including film, will eventually decompose no matter how well they are cared for," notes Moye. Most of the atmospheric test films are two-thirds of the way into the 100-year expected shelf life of black-and-white film, though Trost cautions, "It's impossible to know exactly how long they will be usable." Spriggs explains that in Livermore's film laboratory, which was specially constructed for the project, the team works efficiently with modern tools to scan millions of frames. He says, "Our scanning technique allows a film to be digitized as a near-perfect copy of the original, thereby preserving this rare scientific information for future use."

One challenge to consistent digitization comes from the range of film formats used by different cameras. The widest film strips, up to 9.5 inches (241.3 millimeters), were used in aircraft to record the blast field from above or to capture the mushroom cloud from ground-based photo stations located as far as 48 kilometers away. Some of the 70-millimeter reels do not have perforations, which indicates that they were shot by a camera taking still images at several-second intervals, rather than as a continuous movie. This format was used to document cloud movement and dissipation over relatively longer time periods. Unlike standard 70-millimeter-wide film, some reels contain vertically oriented images. For example, in a vertical orientation, a mushroom cloud's top and stalk grow toward the short ends of the rectangular frame instead of the long sides. Another variation is frame height. Moye states, "Some frames are five perforations high, while others are seven. I was surprised to see so many odd formats."

A key acquisition for the team was the Golden Eye II scanner from Digital Vision in Sweden, whose versatile aperture and light source can accommodate film formats from 8 to 70 millimeters. (The team uses a flatbed scanner for larger widths.) While most scanners grip a film strip's perforations to feed it around flanges and through rollers, the Golden Eye II is sprocketless. "I don't have to restore all the damaged perforations. The scanner handles old, shrunken film well," says Moye.

The scanner's dual-camera system produces high-resolution images up to 8,000 pixels across with as many as 4,096 different tones (shades of gray). To preserve as much of the original optical density as possible, the team scans the film strips first to obtain lower densities, then again for higher densities. Moye explains, "A digital scanner cannot process everything at once, darkest to lightest, from the films. We would lose shadows or highlights. We need more range, so we scan twice." A computer program combines the two scans into a single digital image that includes the full optical range of the original frame. Even with the extra scan, the speed and resolution of the scanner combined with reduced repair time enable the team to digitize several films per day.



Atmospheric nuclear tests were shot on several film stocks. Pictured here are (from top) offset images on 35-millimeter film without perforations, 8-millimeter images on 16-millimeter film, double 8-millimeter images, and standard 16-millimeter images. Digitizing the multiple formats of test films is a challenge for the preservation team.

Faster, Better, Newer Data

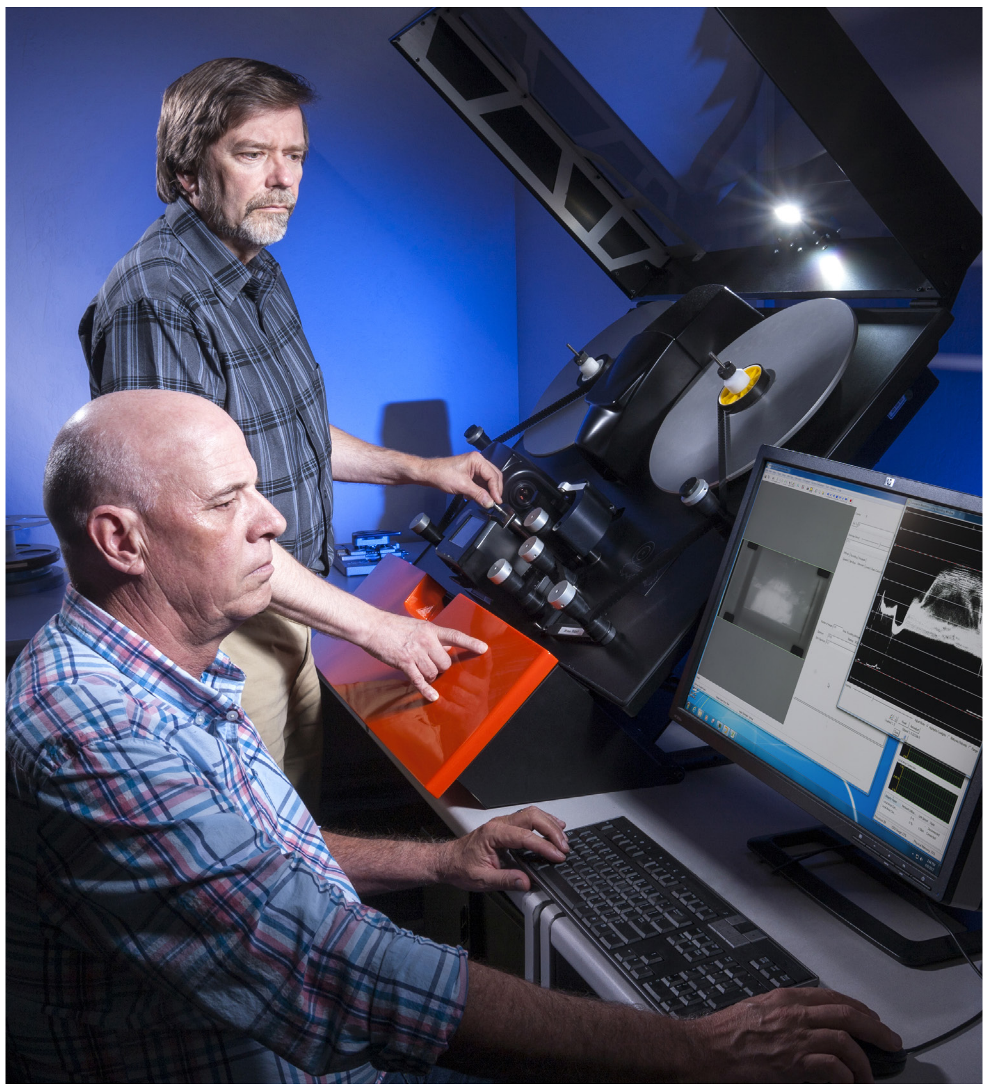
Members of the Film Scanning and Reanalysis Project can analyze film faster, with fewer people, and with more accuracy than previously possible. The team's approach is threefold: speed up the process through automation, collect better data for films analyzed with older methods, and gather data from films not previously examined. The team is conducting a complete analysis of the approximately 50 films shot for each atmospheric test. According to physicist Jason Bender, "Our level of analysis is unprecedented."

Livermore's innovative computerized image-processing technology eliminates most of the manual work of analyzing every frame. Bender explains, "We have many modern tools we can bring to bear on this problem, making it much easier to analyze digital content than in the past." The first hurdle was establishing the best method for extracting data from the scanned films. Bender and colleagues turned to the open-source software community to leverage industry-standard visual-processing functions. The project team uses the Python programming language and the OpenCV (Open Source Computer Vision) library to measure blast dimensions and timescales present in the films. The team relies on the Livermore Computing (LC) Division's parallel computing capabilities for large-scale, batch-processed analysis and fast iteration. "LC's standardized environment is game changing," states Bender. "Studies that once took hours or days to complete on a typical film can now be done in minutes. Through LC, we are also able to bring new interns and scientists into the project."

Analysis begins with measuring a blast's fireball radius and growth rate. Similar to facial recognition technology, the team's software algorithms include shape and pattern matching and noise reduction to find the edges and center of the fireball. By enhancing the background to delineate light from dark, features can be better identified and more data points become available. Machine-learning algorithms weed out visible defects, film manufacturers' marks, and false positives, while customized graphical user interfaces enable scientists to compare fireball contours to circular and elliptical overlays. Pixel-level analysis is possible, and results are highly accurate and consistent. Bender says, "Our tools impose quantitative standards and remove human bias. Data are therefore more reproducible."

Measuring Energy Yield

Crucial to a weapon's effectiveness is knowing a device will hit the right target and produce the intended effects without inadvertent outcomes. At a time when nuclear deterrence policy prohibits nuclear weapons testing, Livermore scientists rely on three-dimensional computer models to predict shock, thermal blast, and fallout effects occurring in various environments. For instance, a mushroom

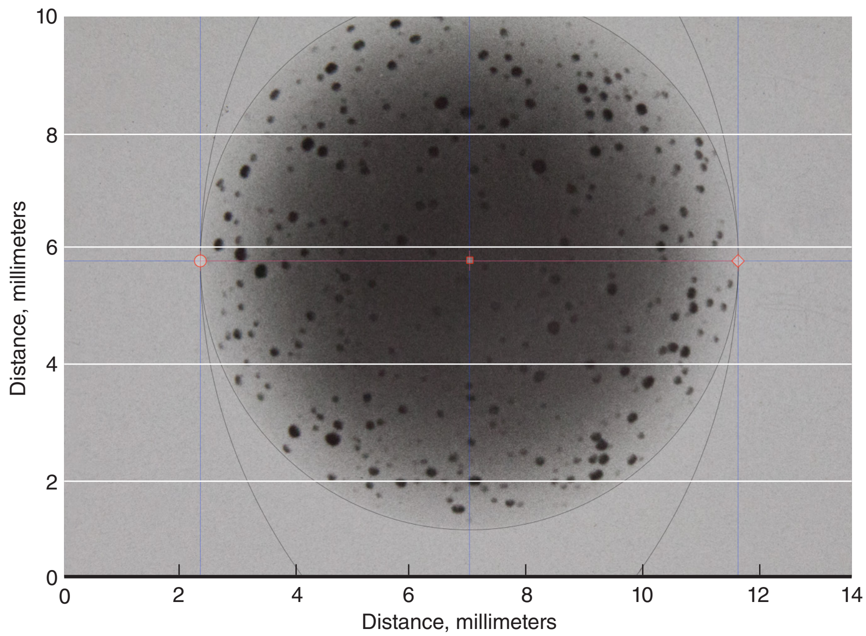


Filmmaker and documentarian Peter Kuran (standing) operates the Golden Eye II scanner, while Moye reviews a scanned image's digital fidelity and optical density distribution. (Photo by Randy Wong.)

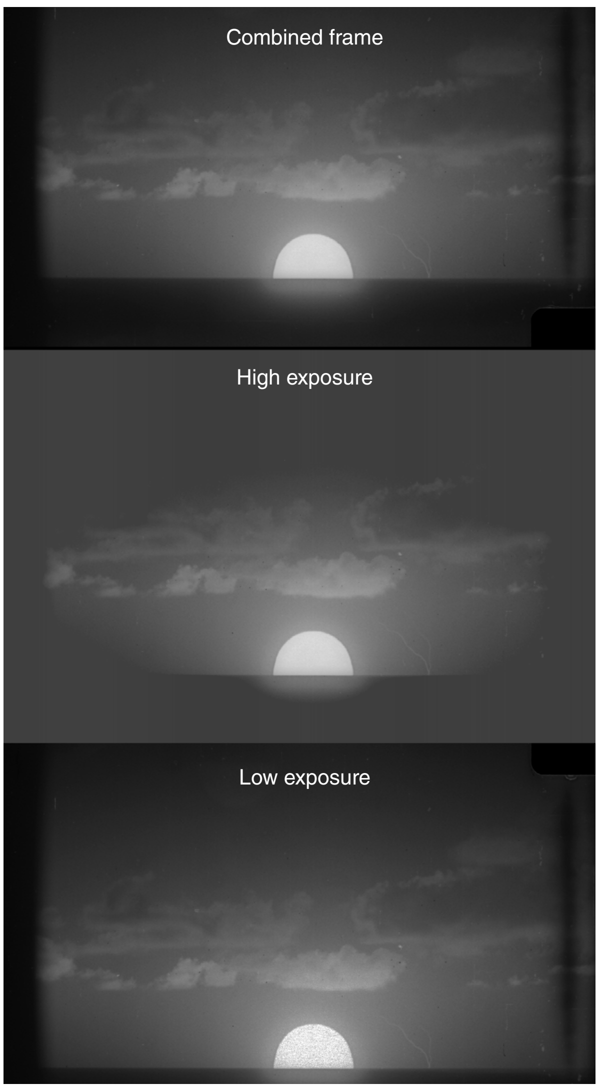
cloud's size helps determine the amount, concentration, and dispersal behavior of fallout particles. This information, in turn, affects calculations of radiation dose rate. Spriggs explains, "As the stockpile ages, we must be able to accurately predict any changes in weapons performance. Since we can no longer physically test our weapons, computer simulations are an essential tool for assessing the health of the stockpile. We need reliable data to validate that our simulations are trustworthy." (See *S&TR*, March 2012, pp. 6–13; and *S&TR*, July/August 2015, pp. 6–14.)

A key data point in these simulations is a weapon's yield. Much like an engine's work rate is measured in terms of horsepower, the energy yield of a nuclear weapon is expressed in units of TNT (trinitrotoluene). For example, an explosion with a yield of 17 kilotons produces energy equivalent to 34 million pounds of TNT—roughly the energy produced by the nuclear devices dropped on Hiroshima and Nagasaki, Japan, in 1945. The weapons immortalized in the atmospheric nuclear test films often produced energy yields several hundred times more powerful.

Yield can be estimated as a function of time based on light output. A nuclear detonation produces two light pulses. The first pulse is associated with the shock wave's formation and its subsequent cooldown during expansion. The second pulse is produced by the light from heated gases in the atmosphere (the fireball). The brightest points of these two pulses and the minimum light output occurring between them helps scientists determine a weapon's yield. Other parameters include the duration of the fireball and the initial rise velocity of the mushroom cloud.



(above) To determine a fireball's dimensions, the project team uses a measured calibration factor that converts pixels to millimeters and accounts for the camera's focal length and distance from the detonation point. This fireball was produced by an air-dropped device in 1953. Livermore scientists can confirm its diameter to within 1 pixel (0.4 meters), which represents an energy yield uncertainty of approximately 0.2 percent—significantly more precise than the contemporaneous calculation of 6.0 percent.



Each reel passes through the scanner twice to improve image fidelity. (bottom) The low-exposure range provides higher fidelity shadows outside the fireball, but the fireball itself appears grainy. (middle) The high-exposure range provides more nuanced highlights yet lacks depth in darker tones. (top) The combined frame contains this 1956 test's full optical range.

Necessity and Invention in Mid-20th Century Film Technology

The era of atmospheric nuclear testing ushered in advancements in film technology for better capturing testing events. During that time, a pioneering photography company called Edgerton, Germeshausen, and Grier, Inc. (EG&G), provided camera timing and weapons firing systems as well as high-speed photography for the tests. In addition to test films, other artifacts—camera rigs anchored to palm trees or stowed in concrete bunkers, towers assembled for observation, before-and-after images of target structures in the blast zone—were featured in documentary footage and still photography. Many models of high-speed motion picture cameras and film stocks were used, ranging from 8-millimeter to 9.5-inch (241.3-millimeter) widths. Special cameras were designed for studies of motion, velocity, and light intensity. By 1962, cameras had become faster and produced sharper images than were possible just a decade earlier.

According to Peter Kuran, author of *How to Photograph an Atomic Bomb*, EG&G was navigating a learning curve. As Hollywood filmmakers shifted from the highly flammable cellulose nitrate film to cellulose acetate, EG&G technicians also transitioned to ensure higher fidelity recordings. Acetate-based film, known as safety film, is less flammable than cellulose nitrate and more sensitive to light, which affects its optical density and exposure. However, safety film has limitations when it comes to ultrabright images of explosions. EG&G worked with film manufacturers to create emulsions that increased the light sensitivity of black-and-white safety film beyond what was commercially available.

Partway through the testing years, EG&G began using a new product called microfile film, with a fine-grained silver-halide emulsion. Smaller grains reduced the film speed, helping prevent overexposure from intense light. This film composition enhanced the quality of high-resolution nuclear detonation photography. “Nuclear explosions are brighter than the Sun. Most film stocks back then were not made for that range of exposure, but microfile film could handle it,” notes Kuran.

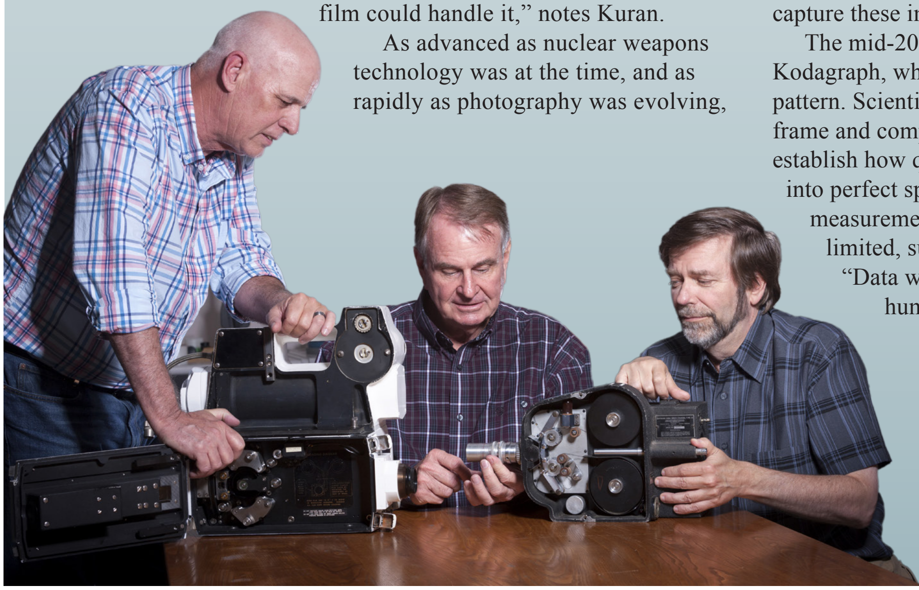
As advanced as nuclear weapons technology was at the time, and as rapidly as photography was evolving, photographic analysis tools had yet to catch up. Using a device called a densitometer, analysts painstakingly measured optical density at fixed points, frame by frame, to measure light output over time. The densitometer works by aiming a light source at a photoelectric cell, which converts the light into electricity. When a film strip is placed between the light source and the cell, the amount of light transmitted through the images is measured by the change in electricity produced. The densitometer displays this change as an optical density reading.

Although the densitometer allowed analysts to measure light output, they were unable to discern minor changes within a given region of the film. For instance, a shock wave boundary could escape detection and simply blend into the sky. “Scientists of that era did not have the technology to look at the shockwave boundary once it was far removed from the fireball,” observes Livermore physicist Greg Spriggs.

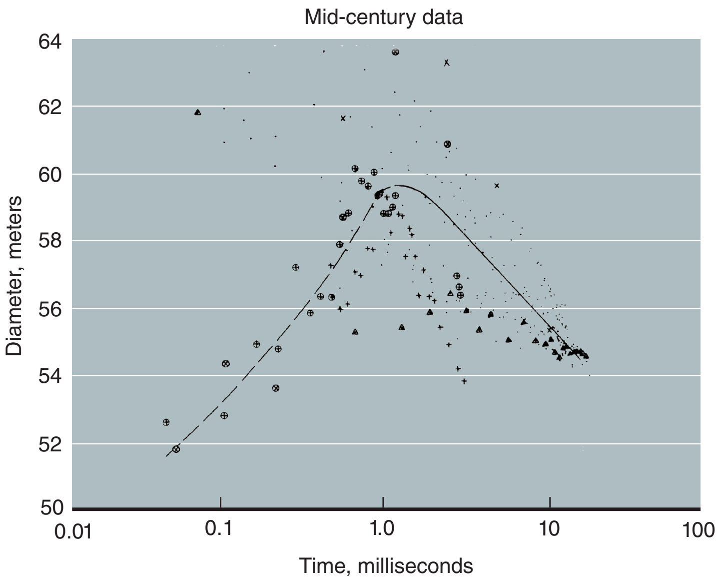
A new type of camera attempted to overcome some of these limitations. Called a streak camera, this device allows light to enter through four to six horizontal slits in the lens cap. The light is captured on a film strip moving at a constant speed, producing continuous streaks along the length of the film. To ensure that at least one streak will not be saturated, neutral-density filters of various optical densities were placed in front of the slits to attenuate the light.

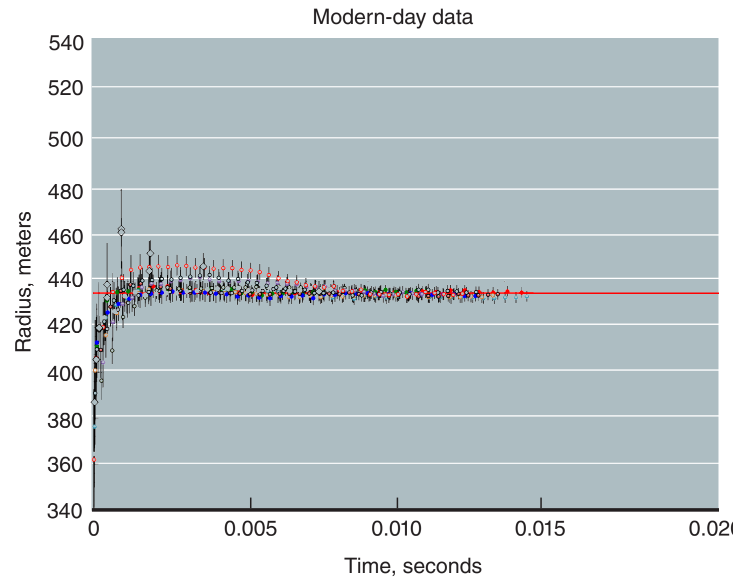
Streak cameras usually ran at 100 frames per second, which was fast enough to capture both light pulses produced by the detonation, but too slow to be accurately resolved by a densitometer. Spriggs explains, “At the time, EG&G knew light output could be captured with streak cameras, but no practical way existed to analyze the results in detail.” EG&G stopped using the streak camera on atmospheric nuclear tests in the late 1950s. Sixty years later, the camera can be appreciated as ahead of its time. “Technology has come a long way since then. Now, for the first time, we can analyze in great detail the entire light output from a nuclear detonation,” continues Spriggs. “We are thankful EG&G had the foresight to capture these images.”

The mid-20th century film analysis toolbox also included a Kodagraph, which enlarged an image and projected it onto a grid pattern. Scientists would measure the size of a fireball in a single frame and compare it to measurements in subsequent frames to establish how quickly it expanded. However, fireballs do not form into perfect spheres, and multiple analysts ended up with different measurements of the same blast. The overall process was limited, subjective, and inconsistent. According to Spriggs, “Data were all over the map, leaving a lot of potential for human error.”



(from left) Jim Moye, Greg Spriggs, and Peter Kuran examine two high-speed, rotating-prism cameras similar to those used for recording mid-20th century atmospheric nuclear tests. The Fastax (left) and Eastman both shoot 16-millimeter film. (Photo by Randy Wong.)





Plots derived from contemporaneous and modern-day data analysis depict fireball measurements from a 1955 atmospheric test. Although the graphs use different scales, uncertainty is approximately a factor of 10 smaller with the newer data points, which align more closely with an asymptotic value. Asymptotes are shown as a dashed curve (left) and red line (right). Colors and symbols represent different camera shots of the same test.

Shock wave position can also contribute to yield estimates. However, accurately measuring a shock wave's radius can be difficult when the shock wave is irregularly shaped, and measurements off by only a small amount can adversely affect yield calculations. For instance, if the radius is incorrectly measured by 1 percent, the yield of the blast could be skewed by 5 percent. Spriggs states, "Using modern image-processing techniques, we can make more precise measurements of the shock wave radius. In most cases, we have reduced the uncertainty of the yield estimates by nearly an order of magnitude."

Silent Films Speak Volumes

By mining the valuable data found in historic atmospheric nuclear test films, the Film Scanning and Reanalysis Project strengthens the Laboratory's stockpile stewardship capabilities for future generations. Throughout the project, Spriggs and colleagues have processed the films with assistance from college and graduate students studying math, computer science, and physics. These summer interns have helped develop data extraction tools, work on uncertainty quantification in timing, and build the database of timing data. "We're establishing a complete analysis platform so scientists can uncover other types of data from these films, such as complex features in fireball structure," says Bender. The data may also prove useful for modeling cloud rise in more complex environments, such as urban canyons. The team plans to extend their analysis technology to new global security applications for first responders and radiochemists.

In March 2017, the Laboratory posted the first batch of 63 digitized films to its YouTube channel with an introductory video featuring Spriggs and Moye. The videos range in length from 2 seconds to more than 7 minutes, each with a frame counter in the corner. The YouTube release has generated intense public interest, with collective views topping 5.5 million in just 8 months, and the number continues to grow. The project has gained attention from major media outlets including *The New York Times*, CNN, CBS Sunday Morning, National Public Radio's Northern California affiliate (KQED), the *San Francisco Chronicle*, and several San Francisco Bay Area news television stations. *Scientific American, WIRED, Esquire*, and other national magazines have also covered the release.

Spriggs has received positive feedback from many YouTube viewers. Some offer their services in film archiving or preservation, while others share personal stories of the nuclear testing era. "The videos are resonating with the public, which is what we want," remarks Spriggs. "Our efforts to maintain a safe, secure, and effective nuclear deterrent are paramount to our national security."

—Holly Auten

Key Words: atmospheric nuclear test, computer simulation, energy yield, film preservation, Film Scanning and Reanalysis Project, Livermore Computing (LC), Nevada National Security Site, Nevada Test Site, nuclear weapons, OpenCV (Open Source Computer Vision), open-source software, optical density, Pacific Proving Grounds, Python, stockpile stewardship.

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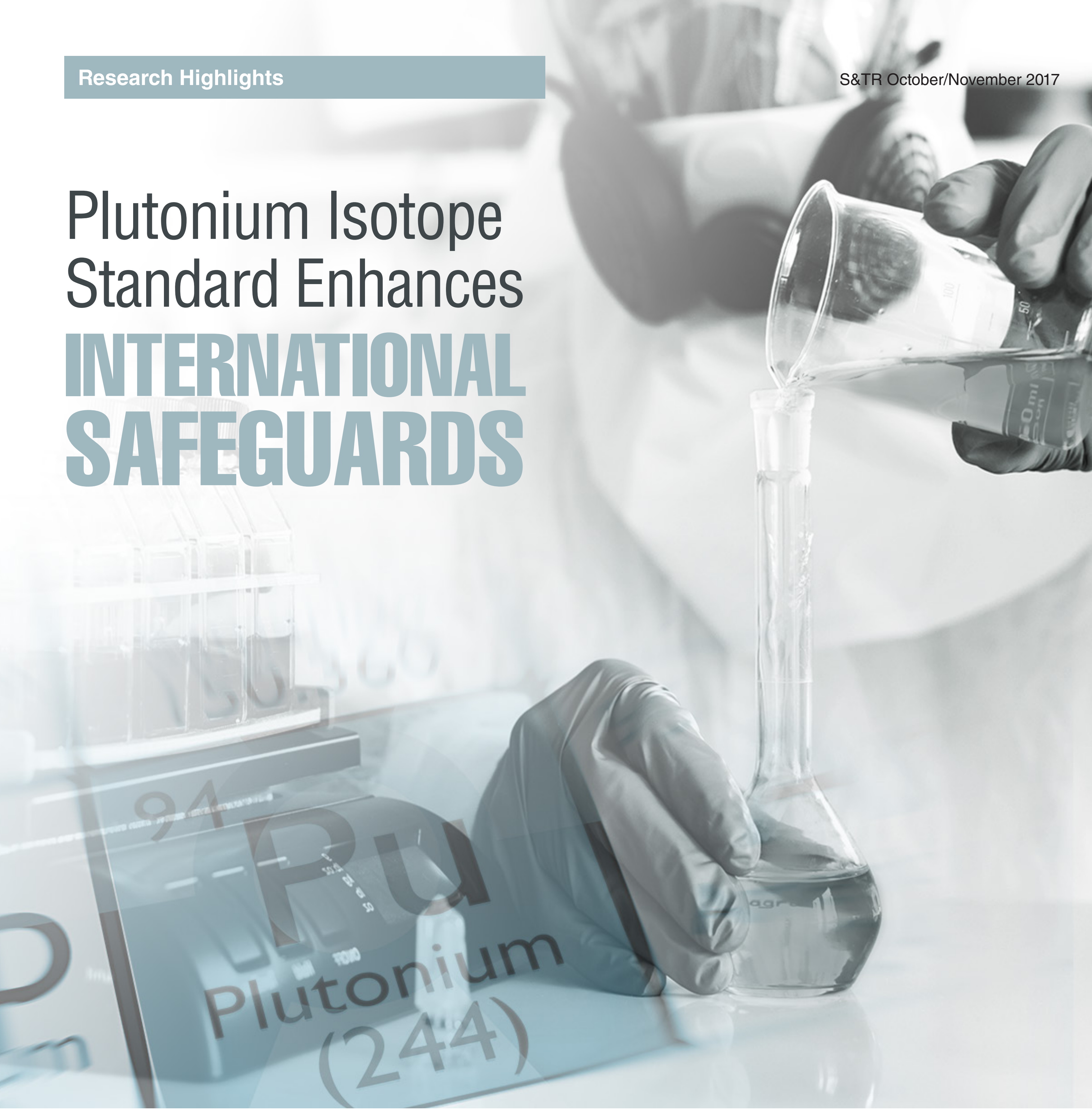
Plutonium Isotope Standard Enhances INTERNATIONAL SAFEGUARDS



FOR decades, Lawrence Livermore researchers have developed new technologies to help detect undeclared nuclear materials and associated activities. Many of Livermore's ongoing efforts have been funded by the Department of Energy's National Nuclear Security Administration (NNSA) through its Office of Nonproliferation and Arms Control (NPAC) and the Office of International Nuclear Safeguards. A continuing aim of NPAC is strengthening the capabilities of the International Atomic Energy Agency (IAEA) and its member states to implement and meet obligations regarding international safeguards. Based in Vienna, Austria, IAEA is responsible for deterring the proliferation of nuclear weapons through early detection of possible misuses of nuclear materials or technology.

IAEA's Network of Analytical Laboratories (NWAL) analyzes environmental samples taken by IAEA inspectors at nuclear facilities worldwide. Careful analysis demands extraordinarily sensitive equipment and high-purity reference materials that are used to measure the amount and isotopic composition of a sample. Over the last three years, a Livermore team led by isotope geochemist Ross Williams has been fabricating a high-purity plutonium-244 (^{244}Pu) reference material for use in IAEA analyses of environmental samples. The new material is expected to enable more precise measurements and at lower concentrations.

An Ideal Reference

Williams explains that a critical task of IAEA is verifying nations' assurances about their use or production of plutonium isotopes. Verification requires regular environmental sampling of nuclear facilities and their products, followed by analysis at one of IAEA's laboratories, which include Livermore. High-purity reference materials are used to measure the amount and isotopic composition of a sample through a technique called isotope dilution mass spectroscopy (IDMS). This technique offers detection limits at femtogram (10^{-15} grams) or attogram (10^{-18} grams) levels of plutonium isotopes. The analyses typically focus on obtaining the exact ratio of plutonium-239 (^{239}Pu) to plutonium-240 (^{240}Pu), which can be correlated with specific nuclear processes, ranging from nuclear weapons research and production to nuclear power–related activities.

IDMS precision depends on the quality of the isotopic reference material used for analysis. The isotope presumed to be the least abundant in the sample is preferable as an isotopic tracer, called a spike. Isotope ^{244}Pu is extremely rare in nature and is not produced in quantity by the nuclear fuel cycle. With a half-life of 80 million years, ^{244}Pu is also the most stable of plutonium's six isotopes. These characteristics make it ideal as a spike for quantifying plutonium isotopic content and their relative concentrations.

"Adding a high-purity rare isotope reduces the uncertainty of analysis when you are looking for more prevalent isotopes," says Williams. He notes that a typical environmental sample contains zero (or an extremely small amount) of ^{244}Pu but unknown amounts of ^{239}Pu, ^{240}Pu, plutonium-241, and plutonium-242. The new Livermore plutonium spike contains precisely measured concentrations of ^{244}Pu with trace (but known) amounts of the other plutonium isotopes. The extremely low abundance of these other isotopes enables higher precision measurements of the ratio of ^{239}Pu to ^{240}Pu in environmental samples.

Williams notes that the worldwide stocks of the current ^{244}Pu spike are nearly exhausted. In addition, the existing reference material lacks the desired isotopic purity and contains relatively high levels of the other plutonium isotopes, the correction for which increases the uncertainty of measurements. The new high-purity ^{244}Pu certified reference material (CRM) should increase the confidence in NWAL-reported results.

Spike Origins Date from the 1970s

The high-purity (99.98 percent purity) ^{244}Pu CRM produced by Williams, colleague Kerri Treinen, and other Livermore scientists, technicians, and nuclear chemistry students caps a 25-year saga involving international negotiations as well as

In 2015, the All-Russian Scientific Research Institute of Experimental Physics shipped highly purified plutonium-244 (shown here) to Lawrence Livermore, where it was further purified, analyzed, and dispensed into aliquots. The plutonium was finally made into a high-purity certified reference material for use in analyses of environmental samples from the International Atomic Energy Agency.

purification and certification work. Collaborators have included the IAEA's Department of Safeguards; the All-Russian Scientific Research Institute of Experimental Physics (VNIIEF); Savannah River, Oak Ridge, and Los Alamos national laboratories; the Commissariat à l'Énergie Atomique (CEA) in France; and the National Institute of Standards and Technology (NIST), part of the U.S. Department of Commerce.

During the early 1970s, 86 plutonium targets were irradiated at the Department of Energy's Savannah River Site to produce gram quantities of californium-252 (^{252}Cf). Twenty-one irradiated targets were processed at Oak Ridge to recover the ^{252}Cf and other elements. A 37-gram plutonium fraction containing 25 percent (or 8.8. grams) of ^{244}Pu was also recovered. The plutonium was enriched to provide the nearly exhausted, high-purity ^{244}Pu currently in use. In 1991, IAEA began to explore options to produce high-purity ^{244}Pu needed by its analytical laboratories using tails (at about 17 percent ^{244}Pu) formed during the earlier enrichment. VNIIEF announced its capability to perform the required separation processes using its electromagnetic mass separator. However, technical preparations did not begin between NNSA, VNIIEF, and IAEA until more than a decade later. In 2005, NNSA supplied IAEA with a 0.5-gram "test" portion of the Oak Ridge source material. Following many years of negotiations, the test material was delivered to VNIIEF in February 2012, and work began on electromagnetic separation of the material to increase its isotopic purity. As part of the international agreement, VNIIEF had to demonstrate its capability with the test portion, and IAEA would verify measurements of the separation products.

VNIIEF scientists performed two stages of electromagnetic separation for the supplied material. The first round of separation in 2012 yielded approximately 10 milligrams of intermediate product. A small quantity was shipped to Livermore in April 2013 for analysis. Both the U.S. and Russian measurements of the plutonium isotopic composition were in agreement—the purity had increased to 98.86 percent. The following year, VNIIEF conducted the second and final round of separation.

Laboratory isotope geochemist Ross Williams inspects bottles that will be used for preparing a new certified reference material. (Photo by Randy Wong.)

Following several stages of purification, the team prepared a master solution of plutonium-244. Five-milliliter aliquots of the solution were dispensed into 190 (30-milliliter capacity) fluorinated ethylene propylene bottles, which will eventually be sent to IAEA's Network of Analytical Laboratories.

In early 2015, IAEA oversaw the conversion of the final liquid solution into dry salts of plutonium nitrate to ensure safe transportation. The entire volume of the final product (approximately 800 micrograms) was sent to Livermore in May 2015 to be fabricated as high-purity CRM. The bulk of the high-purity material was contained in two 25-milliliter glass volumetric flasks. Analysis performed by Williams's team confirmed the value reported by VNIIEF—99.98 percent, slightly below the target value of 99.99 percent.

A Rigorous Preparation Process

Upon receipt of the purified material, the Livermore team embarked on a painstaking, multistep effort to prepare the CRM. The researchers worked in a laboratory that was refurbished in 2014 solely for the effort. The laboratory features advanced analytical instrumentation and state-of the-art air filtration technology found in many semiconductor fabrication facilities. The special facility was required to eliminate any possibility that residual trace amounts of plutonium isotopes left in other laboratories from research conducted many years ago might contaminate preparation of the CRM.

The team transferred the material from the two glass flasks, verified that the isotopic composition of each was identical, and purified the plutonium to separate any uranium and low concentrations of trace elements, such as lead and iron found in the final product from VNIIEF. “Our focus has been on preserving the isotopic purity of the plutonium and ensuring that it does not contain elements that might interfere with analyses,” says Williams. A master solution was then prepared from the purified plutonium.

Five-milliliter aliquots of the master solution were dispensed into 190 (30-milliliter capacity) fluorinated ethylene propylene bottles for long-term storage. Simply cleaning the bottles required several months of work to ensure their sterility and several more months to determine the tare (empty) weights of the individual bottles. The filled bottles were weighed and their contents verified so that each contained about 110 nanograms of ^{244}Pu. The solutions were then dried to ensure a long-term shelf life of at least 20 years.

(from left) Livermore’s Kerri Treinen, Richard Essex of the National Institute of Standards and Technology, and Williams review the procedures for dispensing a certified reference material from a master solution. (Photo by Randy Wong.)

The isotopic purity of each CRM is greater than 99.98 percent ^{244}Pu, with 0.0040 percent ^{240}Pu and 0.0012 percent ^{239}Pu. This purity is significantly greater than the ^{244}Pu spike currently used by NWAL. Assuming that analysis of one environmental sample requires up to 10 picograms of ^{244}Pu, and with a tenfold margin to account for various quality control measurements, one CRM unit containing 110 nanograms of ^{244}Pu will be sufficient for analyzing 1,000 samples, which covers IAEA’s annual needs.

In early 2017, CRM preparation entered its final phase prior to NIST certification. This phase involves rigorous verification of the composition and concentration of the CRM using IDMS. In October 2016, the team prepared samples of the ^{244}Pu CRM mixed with a ^{239}Pu spike obtained from Los Alamos, while others were mixed with a ^{239}Pu spike from the European Union’s Institute of Reference Materials and Measurements. These IDMS mixtures were sent to Los Alamos and CEA for independent analysis, while the Livermore team also conducts its own analysis. The team will compile the three laboratories’ findings and report these results to NIST for evaluation. Upon completion of the evaluation, NIST will determine in early 2018 the suitability of the ^{244}Pu CRM for certification. Following certification, the units will be packaged for shipment and long-term storage. NWAL will be the first recipient.

These high-purity ^{244}Pu spikes will become extremely valuable to the safeguards community for improving analyses of very low-level plutonium in environmental samples. Williams emphasizes that the spike was made possible through more than a quarter-century of international collaboration. He says, “It’s been a long and tortuous path, but we’re almost there.”

—*Arnie Heller*

Key Words: All-Russian Scientific Research Institute of Experimental Physics (VNIIEF), certified reference material (CRM), Commissariat à l’Énergie Atomique (CEA), Institute of Reference Materials and Measurements, International Atomic Energy Agency (IAEA), isotope dilution mass spectroscopy (IDMS), National Institute of Standards and Technology (NIST), Network of Analytical Laboratories (NWAL), Office of International Nuclear Safeguards, Office of Nonproliferation and Arms Control, plutonium-244 (^{244}Pu).

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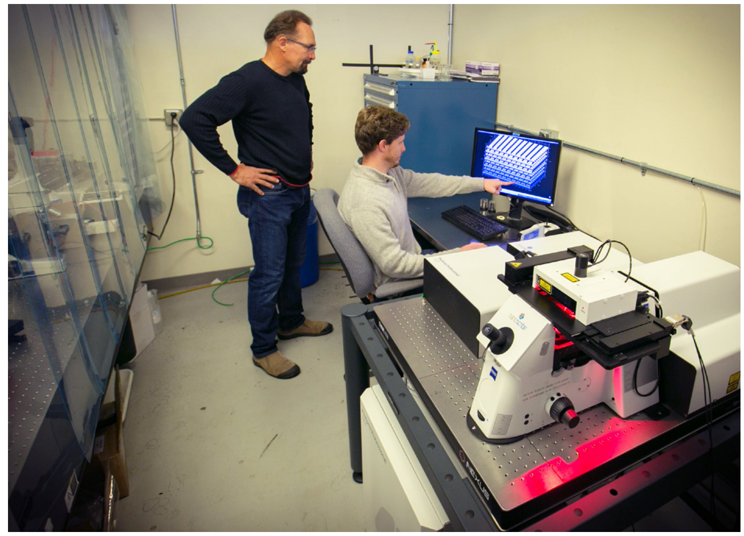
Additive Manufacturing Helps Reinvent NANOPOROUS MATERIALS

LOW-DENSITY bulk materials, foams in particular, are a ubiquitous ingredient in everyday life. Used in various products from kitchen sponges to insulation in spacesuits, these materials have both basic and exotic applications. At Livermore, a type of bulk material called nanoporous foam plays a role in high-energy-density (HED) experiments at the National Ignition Facility (NIF).

Lawrence Livermore has a long history of developing a class of foams known as aerogels, which are useful for scientific research because their high surface area gives them unique physical, chemical, and mechanical properties. Aerogels are formulated through a simple wet-chemistry process that exploits nature's methods of self-assembly, resulting in a random network of interconnected, nanometer-sized particles. However, the wet-chemistry process limits a scientist's control over the aerogel's multiscale morphology and thus its structural architecture across various length scales.

To address the need for low-density materials with engineered morphologies, shapes, and densities, Lawrence Livermore researchers Juergen Biener and James Oakdale, along with a team of Laboratory scientists, are applying additive manufacturing (AM) processes to generate specific nanoporous structures. (See *S&TR*, March 2012, pp. 14–20.) Funded by the Laboratory Directed Research and Development (LDRD) Program, the scientists have created low-density foam parts with submicrometer features that can be tailored to precisely controlled macroscopic dimensions and nonuniform architectures—perfect for HED experiments and other scientific and commercial applications.

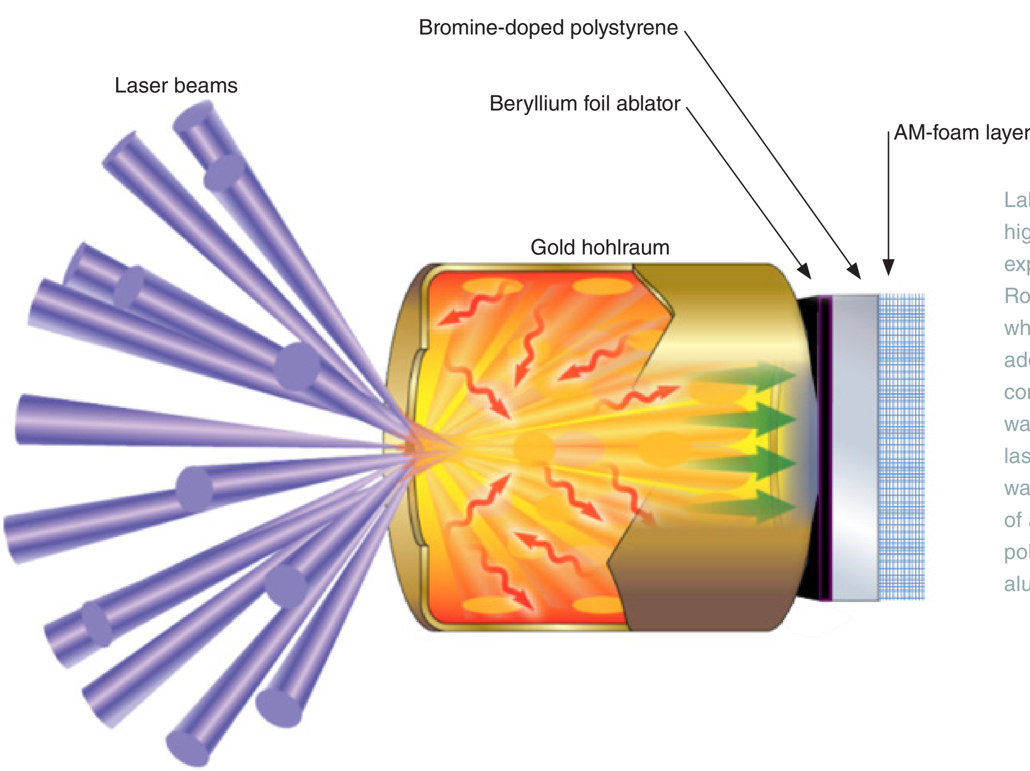


Livermore scientists James Oakdale (seated) and Juergen Biener examine a three-dimensional printed foam design before fabricating it using the Laboratory's Nanoscribe Professional GT printer.

Process Makes It Possible

Low-density foam target components are typically made through precision machining of bulk aerogel samples, and in the case of graded-density target components, by assembling several machined parts made from materials of different densities. Producing such target components is expensive, time consuming, and not easily reproducible. Moreover, the resolution of traditional AM-based fabrication methods limits their use for producing the kind of low-density foams needed for HED experiments. Recently, advancements in instrumentation have improved AM resolution and allowed such methods to reproduce the structural characteristics of bulk aerogels. These improvements allowed Biener and Oakdale to strategically implement an AM technique known as two-photon polymerization direct laser writing (2PP DLW) to print specialized, low-density foams. Biener says, "This technique is uniquely suited to fabricate ultrahigh-resolution, low-density foam components for superior performance."

The foams are created by focusing a femtosecond-pulsed laser onto a photo-responsive liquid material. At the focal point of the laser beam, the light intensity is high enough to drive two-photon absorption processes, which cure the liquid into a solid glassy



Laboratory researchers conducted high-energy-density ramp compression experiments at the University of Rochester's OMEGA Laser Facility in which nanoporous foams, created through additive manufacturing (AM), were used to control the temporal shape of the pressure wave applied to a sample. In these tests, laser beams drive an indirect plasma shock wave through the reservoir consisting of a beryllium ablator, a bromine-doped polystyrene film, and the AM foam onto an aluminum thin film sample (not shown).

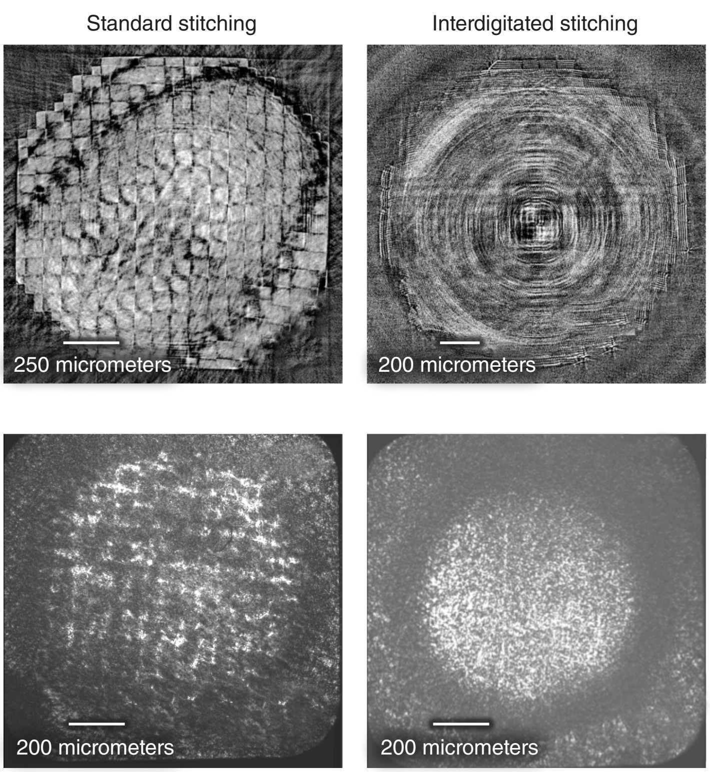
material. The focal spot of the laser is then moved around in space to obtain the desired three-dimensional (3D) structure. Unlike other AM techniques, 2PP DLW allows the scientists to create extremely fine features, such as lines less than 150 nanometers wide (200 times thinner than a human hair). Yet, fine features at high resolution come at a price. Only a finite area (100 by 100 micrometers square) can be scanned at a time because of limitations imposed by the focusing optics. Thus, to build a millimeter-sized structure, smaller structural units must be printed one at a time and arranged over the desired area—a process known as stitching.

A New Type of Building Block

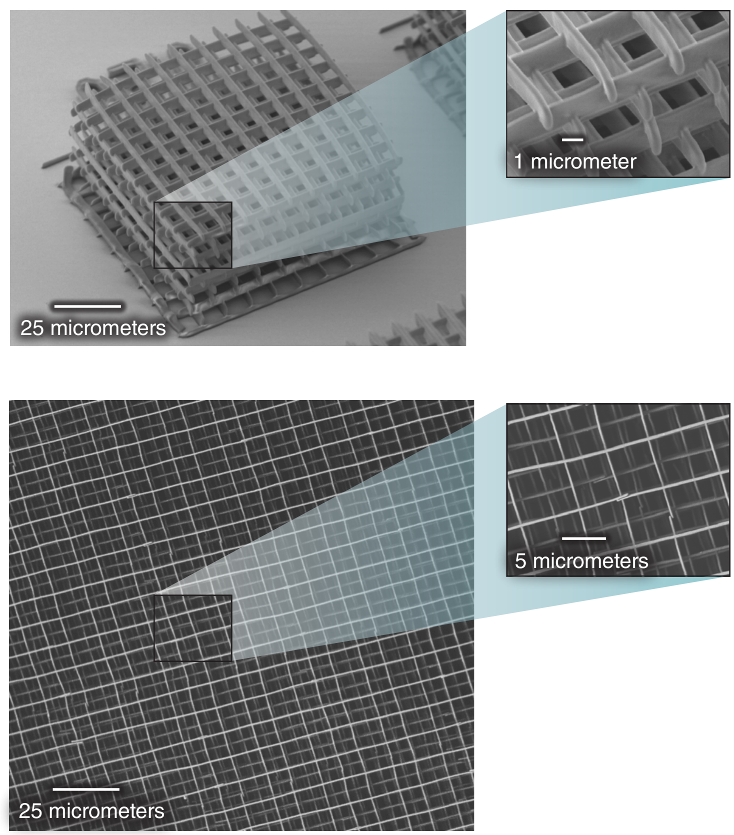
"We often compare the process of stitching to playing with Legos®," explains Oakdale, a postdoctoral researcher in the Materials Science Division of the Laboratory's Physical and Life Sciences Directorate. "We are building microscale blocks and stacking them together to create a larger structure." According to Biener and Oakdale, the tricky part of creating these structures was determining how to fabricate samples large enough for HED experiments.

The scientists used the Laboratory's Nanoscribe Professional GT printer to initially print a millimeter-sized sample. However, they soon discovered the system software could not handle the large file size. As a result, team member Will Smith rewrote how the writing process was controlled. "By creating our own writing process, we have full control over the structure and how that structure is made," explains Smith. "This direct design approach allows us to fully optimize our production and bypass any delays embedded within the software." After Smith's breakthrough, printing a millimeter-sized foam sample went from taking two months to less than eight hours.

Lawrence Livermore experimental physicist Ray Smith and team members tested the newly printed foams at the University of Rochester's OMEGA Laser Facility. For each experiment, a target containing a disk of the 2PP DLW–created foam was mounted behind a beryllium foil ablator and a layer of x-ray absorbing bromine-doped



Cross-sectional tomography images (top) and two-dimensional velocity maps from ramp compression tests (bottom) using 0.07 grams-per-cubic-centimeter AM-manufactured foam samples illustrate how standard (left) and interdigitated (right) stitching affect the uniformity of the pressure drive. Interdigitated stitching strongly reduced local density fluctuations along the stitching seams and thus improved the drive's uniformity, as evidenced by the more even distribution of gray tones within the velocity map.



Using interdigitated stitching, foam blocks are interwoven to produce a stronger, more homogenous structure. Scanning electron microscopy images show the individual foam blocks within a "log pile" architecture (top) and the seams of multiple interdigitated blocks within the stack (bottom).

Hohlraum liners are just one example of the potential applications for additively manufactured foams. The research team's new interdigitated stitching technique enables tooling-free fabrication of liners with tunable feature sizes and high reproducibility. (inset) A sacrificial AM-foam template lines a gold sleeve.

polystyrene. Laser beams drive an indirect plasma shock wave through the AM foam and onto an aluminum target. The density and thickness of the foam controls the temporal shape of the pressure wave applied to the aluminum layer.

"The porous foams help avoid shocks and tailor the pressure ramp," explains Smith. "Nonuniform material density introduces instabilities that affect the uniformity of the pressure front." Indeed, the team's first test shots revealed that the standard stitching process introduced density fluctuations along the seams. These results led the team to develop a new interdigitated stitching technique for building a more seamless 3D structure. Oakdale says, "We essentially weave individual printed blocks together, burying the stitch seams inside the structure of adjacent blocks."

Follow-on experiments using the interdigitated foams revealed significantly more uniform pressure drives. In addition, the team found that the foam briefly holds the sample pressure constant at 0.5 gigapascals (nearly half a million atmospheres of pressure) for 5 nanoseconds—a discovery with huge implications for other types of research. "Those 5 nanoseconds may be enough to test material properties at constant high pressures," says Smith. "We could potentially develop this technique into a platform to study matter deep within planetary interiors."

Additional Discoveries

The combination of 2PP DLW and interdigitated stitching has allowed the team to fabricate reproducible foams with engineered density gradients and the exact macroscopic dimensions required for tooling-free integration into test targets. This process is much faster and more streamlined than traditional approaches using bulk carbon aerogels. Biener says, "It opens the door to many more applications."

One potential application is supporting the spherical ablator shell inside targets for inertial confinement fusion experiments at NIF. The ablator shells are usually supported by ultrathin polymer tents, which, although thin, can still affect implosion. Low-density foams are a promising alternative because their extremely low density effectively renders the foams invisible. Another application could be as a sacrificial template for hohlraum liners to increase the efficiency of the hohlraums during experiments.

Foams could also be used for energy storage purposes. Most energy applications require directional mass transport, and its effectiveness determines how fast a battery can be charged or discharged. "The fabrication of foam materials with engineered morphologies for directional mass transport is at the forefront of materials research in the area of energy storage," says Biener. AM methods are ideal for energy storage because they have the capability to define the 3D architecture of a material. Another LDRD-funded project led by Livermore scientist Marcus Worsley is already in the early stages of investigating this application. "The project is aimed at further improving the power performance of flow batteries through structural optimization," says Biener.

Going forward, Biener and Oakdale's goal will be to apply their techniques to templating. With material templates, scientists could create multiple copies of a material—such as the hohlraum liner—all exhibiting identical properties. The challenge will be determining how to uniformly coat and remove the template without damaging the created structure. "To achieve this objective, we will need to develop application-specific resins that enable efficient and uniform surface deposition as well as damage-free template removal," says Biener. "We now have the technology we need to make further progress."

—Lauren Casonhua

Key Words: additive manufacturing (AM), aerogel, energy storage, high-energy-density (HED) experiment, interdigitated stitching, Laboratory Directed Research and Development (LDRD) Program, low-density bulk materials, low-density foam, National Ignition Facility (NIF), two-photon polymerization direct laser writing (2PP DLW).

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Sudden Changes at ULTRAHIGH PRESSURE

LOWER water's temperature to its freezing point and it turns to ice. Raise it to its boiling point and it becomes water vapor—a gas. At one atmosphere of pressure (101.3 kilopascals), which prevails approximately at sea level, these phenomena are familiar to everyone. However, at much higher pressures and temperatures—up to hundreds of thousands of atmospheres and several hundreds to thousands of degrees—water and other matter behave in complex ways. Under such conditions, a solid of the same composition may assume different crystalline structures, and the molecules that form liquids may order themselves in various configurations. These changes in molecular arrangements are called phase transitions.

Livermore physicist Jon Belof and a team of physicists, engineers, and computational scientists are subjecting matter to extreme conditions and simulating experiments with high-performance computers to study phase transitions at ultrahigh pressures. Examining how matter arranges itself and behaves at these levels may reveal new underlying physics principles. The work also supports the Laboratory's mission in stockpile stewardship by improving models that are used to simulate stockpile components.

Exploring a New Frontier

Belof has been studying phase transitions in elements since 2010, when he was a postdoctoral researcher at Lawrence Livermore. In January 2017, Belof was one of 100 scientists in the nation to receive a Presidential Early Career Award for Scientists and Engineers. This honor is the highest bestowed by the U.S. government on outstanding scientists and engineers who are early in their independent research careers. "The study of material phase transitions at extreme conditions could be a new scientific frontier," he says. "We can now study these phenomena in ways that were not possible five years ago, both experimentally and computationally, allowing us to gain insights into the atomistic physics of material transformations."

Livermore's increasingly powerful high-performance computers and new tools for shocking matter at ultrashort timescales have enabled Belof's team to peer into the physics of phase transitions at an unprecedented scale—occurring as fast as tens of picoseconds (trillionths of a second). With these capabilities, Belof and his team are developing a predictive theory of nonequilibrium phase transitions. In such cases, a material has had its pressure and temperature changed so rapidly that it enters a state far from stability. In an effort to return back to equilibrium, the stressed material changes its phase, but when that process begins and how quickly it occurs are not well understood. By developing a predictive theory for phase evolution, scientists can greatly improve simulation capabilities, and thus achieve more accurate results of a material's behavior and physical properties.

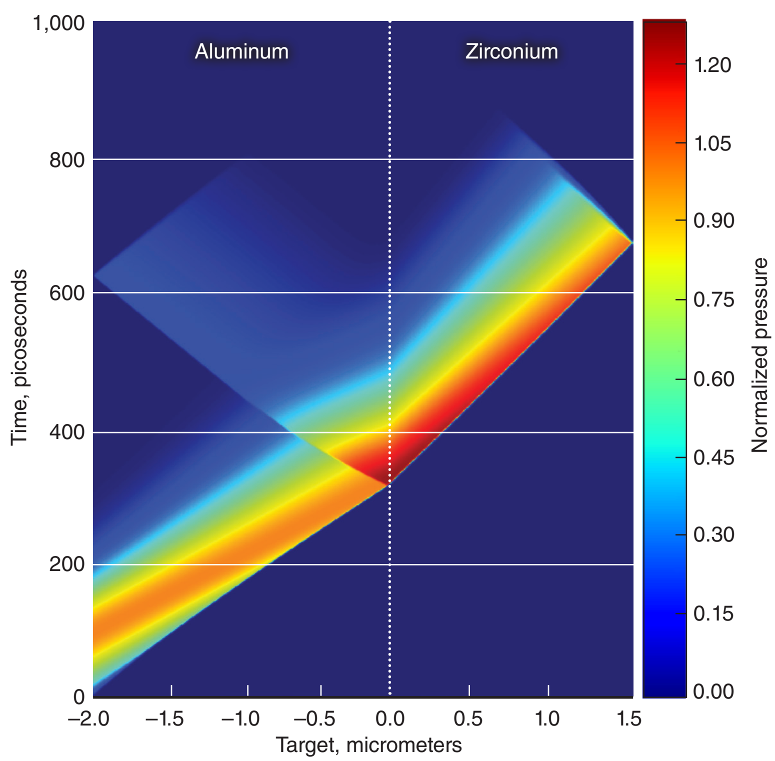
The Secrets of Shocked Zirconium

Recently, Belof's team turned its attention to zirconium (Zr). This metal, which has a high melting point (1,855ºC), is used for heat-resistant applications in the aerospace and nuclear industries. It also has biomedical applications, such as in dental implants and hip replacements.

Funded by Livermore's Laboratory Directed Research and Development Program, Belof and his colleagues partnered with SLAC National Accelerator Laboratory (previously called Stanford Linear Accelerator Center) to run Zr experiments. Using SLAC's Linac Coherent Light Source (LCLS), the team subjected 1.7-micrometer-thick Zr samples (each deposited onto a 2.2-micrometer layer of aluminum) to high-energy laser pulses. During the experiments, the Zr samples underwent more than 100 gigapascals (GPa) of pressure for a duration of less than 1 nanosecond (one-billionth of a second). For that very brief period, while the Zr was compressed, LCLS delivered an intense x-ray pulse to probe the atomic structure of the material by diffraction.

Livermore's Mike Armstrong, co-leader of the experiment, says, "The real power driving this experiment is the LCLS x-ray pulse, one of the brightest sources of x rays in the world." Bombarding the sample with x-ray pulses and recording how the x rays scatter provides a record of the sample's phase transitions. "The distinguishing feature of our experiments is the ability to observe the details of phase transitions at very short timescales," he adds. Since the window of opportunity to examine the shocked material is so small, researchers need to record the diffraction patterns in the briefest intervals possible, less than 100 picoseconds.

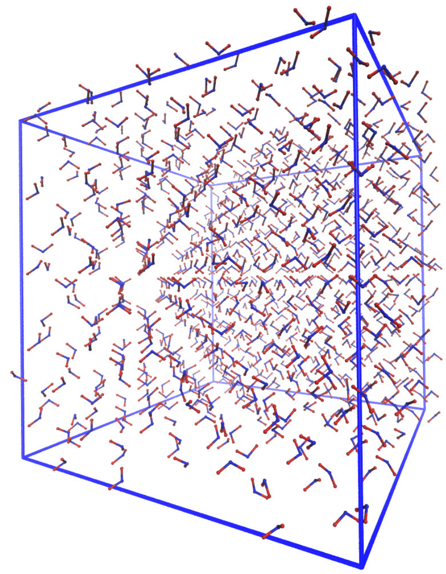
The results of the experiments were striking. The known shock Hugoniot—a set of high-pressure, high-density states



Simulation data from one of the team's experiments using the Linac Coherent Light Source at SLAC National Accelerator Laboratory show how pressure on the target—aluminum ablator (left) and zirconium (right)—changes over time. The x-axis shows the length scale of the target. The laser pulse is passing through the target from left to right, and the color gradient represents the increased pressure from the energy of the pulse. The pressure units are normalized to 27 gigapascals.

achieved when a shock wave is driven at constant velocity through a sample—for Zr indicates that the material passes through multiple solid phases. "However, if the drive is rapid enough, the zirconium can melt directly out of its ambient pressure phase without going through its intermediate phases, suggesting that the transformation pathways are different when the material is subjected to very rapid compression," says Belof. "The kinetics seem to dictate that the relaxation to an equilibrium state is nontrivial."

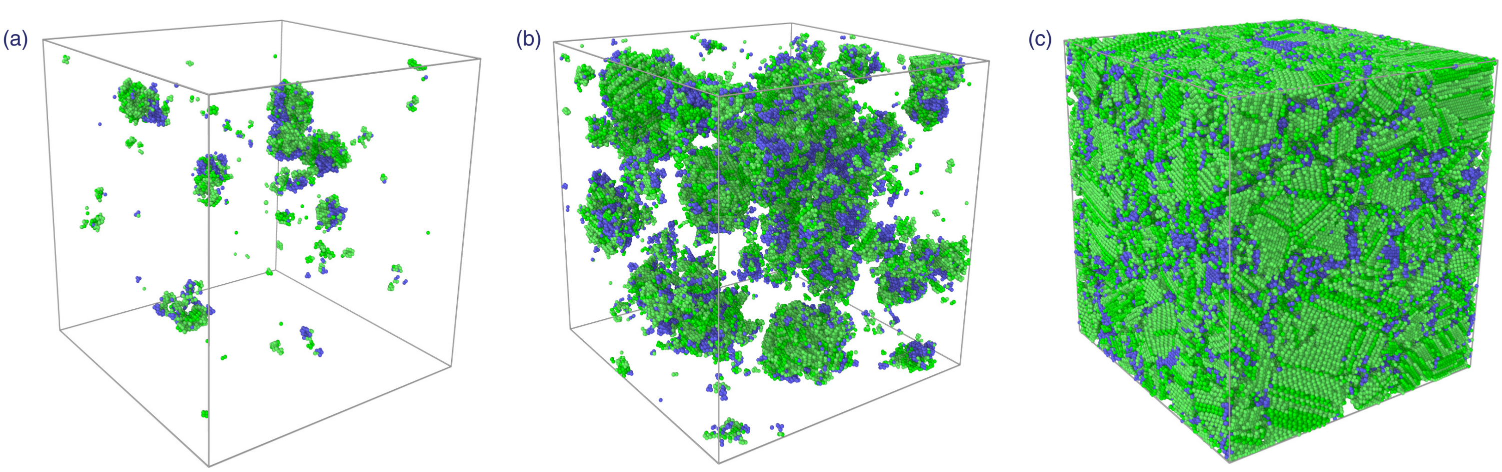
Typically, a material's phase transitions are explained by classical nucleation theory (CNT), first developed in the 1920s. During the formation of a new phase, such as the transition from a solid to a liquid, tens of thousands of molecules organize themselves into the new phase first, forming clusters which then seed the transition. More molecules then aggregate to these nucleation sites until the changeover from liquid to solid is complete. The laws of thermodynamics govern when the change potentially starts and where it ultimately ends (the equilibrium state). At the pressures and temperatures produced during the experiments, CNT begins to break down. Belof says, "At this very short timescale, we could be seeing new physical principles at play that we are just starting to uncover."



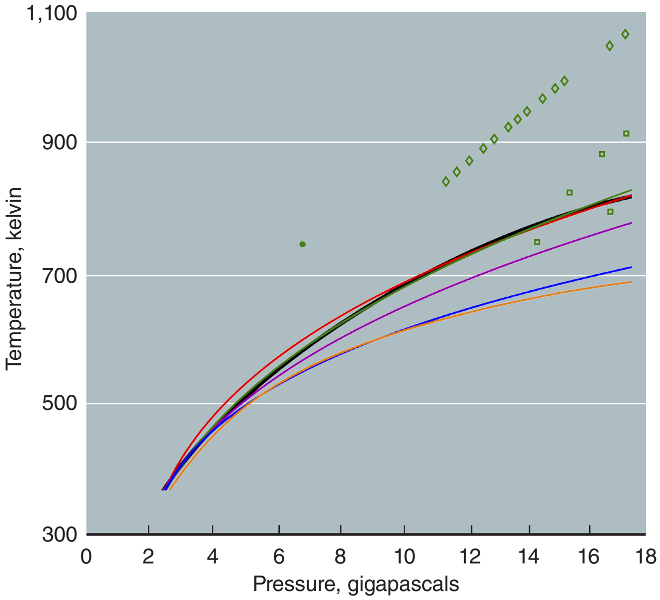
This visualization depicts a molecular dynamics (MD) simulation of the high-pressure water polymorph ice VIII. Oxygen atoms are blue and hydrogen atoms are red. The applied pressure is 10 gigapascals and the temperature is 200 kelvin. MD simulations such as this are making it possible to develop models for phase transformation kinetics in hydrocodes.

Modeling Nucleation at the Atomic Scale

To better understand the physics required for modeling phase transitions, Belof and colleagues are using massively parallel codes on Livermore supercomputers to model the details of each step. Computational materials scientist Luis Zepeda-Ruiz is applying these codes to model systems involving tens of millions of atoms and to follow individual atoms as they change from one phase to another. "For atomistic-level simulations, we still cannot reach the length and timescales associated with most experiments, with a few exceptions," he says. Molecular dynamics simulations are limited by the time step used to propagate the equations of motions. The faster the physics, the smaller the time steps. "However, at extremely high



A visualization of homogeneous nucleation according to classical nucleation theory shows (a) crystals begin to form clusters in a liquid at nucleation sites. (b) Each site grows as more molecules aggregate to the clusters. (c) Growth stops when the phase change to a solid is complete. (Liquid atoms are removed for clarity.)



A portion of the phase diagram for water shows the approximate boundary between liquid water and ice VII. The many curves illustrate boundaries reported by different research teams, indicating that the boundary is still imperfectly known. The Livermore team's theoretical model of the boundary is the black solid line.

deformation rates, processes happen rapidly. We can acheive greater simulation accuracy at these very short timescales."

Zepeda-Ruiz is modifying a code called the Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS) to routinely simulate 30 to 40 million atoms, and at times more than 200 million, during a phase transition. "We can pinpoint the interface between a solid and a liquid with good accuracy," he says. The atomistic-level LAMMPS code provides parameters, such as nucleation and growth rates, that are used in other hydrodynamics codes—SAMSA, developed by Belof, and ARES—to model the material at the continuum level. Together, these codes provide multiscale coverage. Livermore's high-performance computing (HPC) infrastructure makes the work possible—few laboratories in the world have the computing power needed to pursue this research systematically.

The Wonders of Water's Phases

Improved simulation has also allowed the research team to model the solidification of water at ultrahigh pressures using, for the first time, a physics-based model for the phase transition. "Water is one of the most complex substances in existence, so simply determining its equation of state (EOS) is difficult—and we need the EOS before we can attempt to solve the mysteries of kinetics," says Belof. The EOS describes the phases of a material at varying pressure and temperature. Water has at least 17 solid phases.

One reason why this research is important is that water may exist at ultrahigh pressures on some of the many super-Earths detected by NASA's Kepler Mission, a few of which may harbor life. *Escherichia coli* bacteria have been shown to survive at pressures as high as 2 GPa. Furthermore, many scientists believe the majority of the universe's water exists as an ultracold glassy state, a particular phase of water that is of intense interest.

Postdoctoral researcher Philip Myint, working with Belof and colleagues, has explored the solidification of water into ice phases at ultrahigh pressures. "We are trying to implement CNT in a code and use that to understand dynamic compression experiments as well as make predictions. Past models of phase transitions in shock physics have been empirical," says Myint. After successfully modeling a number of shock physics experiments on water, several long-standing controversies on the compressive freezing of water are closer to being resolved.

At pressures above 2.2 GPa, water freezes into ice VII, a state in which the molecules order themselves into a cubic crystalline form (unlike the hexagonal form of ice typical at Earth's surface). Ice VII forms through nucleation clusters that grow until all the material has solidified. This form of ice could be common at the high pressures of extrasolar super-Earths and can exist at temperatures well above 600 kelvin.

Through HPC simulations, Belof, Myint, and colleagues have observed a breaking point at about 7 GPa. "At this pressure, ice VII may form through a homogeneous nucleation process where the clusters contain fewer than a dozen molecules—perhaps as few as two or three—instead of thousands," says Myint. In contrast to freezing initiated by heterogeneous nucleation, which occurs along the plate walls and may take hundreds of nanoseconds to complete, freezing initiated by homogeneous nucleation occurs within the bulk of the water and is completed in less than 10 nanoseconds. "The liquid is being driven so rapidly that it's approaching instability," says Belof. "The dynamic freezing of water to ice VII is shedding light on the fundamental physics of nucleation, since this magnitude of undercooling cannot be achieved without rapid compression." Exploring this new frontier of phase transitions will reveal how nature transforms matter away from equilibrium."

—Allan Chen

Key Words: ARES, classical nucleation theory (CNT), equation of state (EOS), heterogeneous nucleation, high-performance computing (HPC), homogeneous nucleation, ice VII, Laboratory Directed Research and Development Program, Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS), Linac Coherent Light Source (LCLS), phase transition, SAMSA, SLAC National Accelerator Laboratory, super-Earths, ultrahigh pressure, water, zirconium.

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Patents and Awards

In this section, we list recent patents issued to and awards received by Laboratory employees. Our goal is to showcase the distinguished scientific and technical achievements of our employees as well as to indicate the scale and scope of the work done at the Laboratory. For the full text of a patent, enter the seven-digit number in the search box at the U.S. Patent and Trademark Office's website (http://www.uspto.gov).

Patents

Anti-Clogging Filter System
Erik P. Brown
U.S. Patent 9,630,137 B2
April 25, 2017

High Surface Area Graphene-Supported Metal Chalcogenide Assembly
Marcus A. Worsley, Joshua D. Kuntz, Christine A. Orme
U.S. Patent 9,631,148 B2
April 25, 2017

Apolipoprotein Nanodiscs with Telodendrimer
Juntao Luo, Wei He, Kit S. Lam, Paul Henderson, Matthew Coleman, R. Holland Cheng, Li Xing
U.S. Patent 9,644,038 B2
May 9, 2017

Spectroscopic Quantification of Extremely Rare Molecular Species in the Presence of Interfering Optical Absorption
Ted Ognibene, Graham Bench, Alan Daniel McCartt, Kenneth Turteltaub, Chris W. Rella, Sze Tan, John A. Hoffnagle, Nabil Saad, Eric Crosson
U.S. Patent 9,645,077 B2
May 9, 2017

Conformal, Wearable, Thin Microwave Antenna for Sub-Skin and Skin Surface Monitoring
Mark C. Converse, John T. Chang, Eric B. Duoss
U.S. Patent 9,653,784 B2
May 16, 2017

Shape-Memory Polymer Foam Device for Treating Aneurysms
Jason M. Ortega, William J. Benett, Ward Small, Thomas S. Wilson, Duncan J. Maitland, Jonathan Hartman
U.S. Patent 9,662,119 B2
May 30, 2017

Awards

Three Lawrence Livermore teams received **Secretary's Appreciation Awards** from the **Department of Energy** (DOE) for providing assistance to the Ebola task force, the Cancer Moonshot team, and the technology convergence working group. The awards were presented by Dimitri Kusnezov, chief scientist for the National Nuclear Security Administration and DOE.

As part of the Ebola task force, Livermore researchers used their expertise in protein structure prediction along with viral genomic sequence data derived from patients to forecast how the evolution of the viral genome might alter the efficacy of treatment options that were in development. Members were **Jonathan Allen, Tom Bates, Reg Beer, Monica Borucki, Alda Carrillo, Shea Gardner, Erret Hobbs, Pejman Naraghi-Arani, Jason Paragas, Jason Perry, David Rakestraw, Gabriele Rennie, Tom Slezak, Elizabeth Vitalis, Michael Woods, Adam Zemla, and Carol Zhou**.

The Cancer Moonshot team worked together with researchers from Argonne, Los Alamos, and Oak Ridge national laboratories to push the frontiers of computing and align them with the research priorities of the National Cancer Institute. Members were **Ghaleb Abdulla, Jim Brase, Bill Goldstein, Amy Gryshuk, Jason Paragas, David Rakestraw, and Fred Streitz.**

The technology convergence working group was formed because advances in biosciences and biotechnology have resulted in changes to this area's threat space and received high-level attention in the U.S. national security community. The work was conducted along with Sandia and Los Alamos national laboratories. Livermore members were **Tom Bates, Jim Brase, Patricia Falcone, Bill Goldstein, Jason Paragas, Jason Perry, and David Rakestraw**.

Four Livermore programs have been awarded **research and development grants** by the **High Energy Laser Joint Technology Office** (HEL-JTO) within the Department of Defense to launch key partnerships with leading defense, industry, and academic institutions. These research partnerships carry a prospective combined value of more than $2.2 million for the Laboratory through the JTO, which coordinates efforts to develop foundational science for directed-energy laser programs.

The **"Multi-Core Fiber for Automation of Fiber Laser Fabrication" program** is a collaboration with Lincoln Laboratory at the Massachusetts Institute of Technology. Livermore researchers will apply their ability to fabricate fiber optics with ultrapure materials in customized precision forms and bundles in diameters thinner than a human hair.

In collaboration with the Air Force Research Laboratory, the National Ignition Facility's Fiber Laser Group is leading development of fiber optics capable of advancing the science of directed energy. The work is being conducted for the **"Anti-Resonant Tube Fibers for Directed Energy Lasers" program**.

The goal of the "**All-Fiber Raman-Beam-Combined High-Repetition-Rate Eye-Safe Tracking Illuminator Lasers" program** is to develop directed-energy lasers by utilizing wavelengths that will not blind the users. The program collaborates with the Army Research Laboratory.

The **"Additively Manufactured Waveguiding Gain Elements" program** pairs up the Laboratory's N Program within the Global Security Principal Directorate and the Additive Manufacturing Initiative to build in optical control of a laser beam by tailoring the structure of waveguiding ceramic gain media.

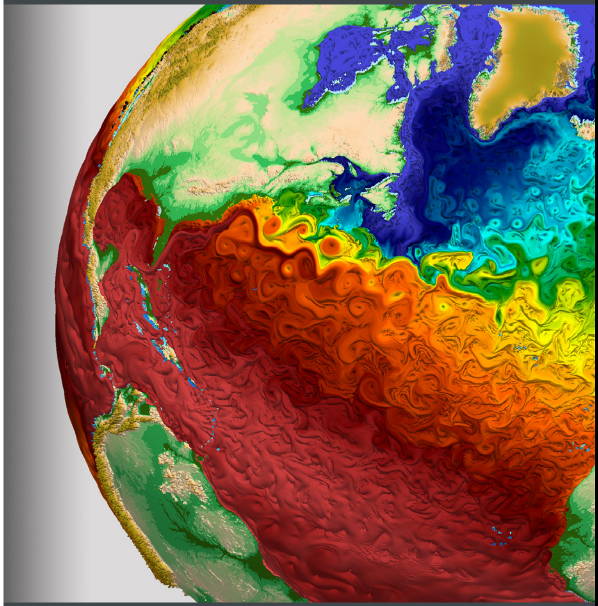
Abstract

Preserving the Past to Protect the Future

Atmospheric nuclear weapons testing has an important place in history for its scientific, political, and cultural legacies. Between 1945 and 1963, the United States conducted 210 such tests and captured the events on dozens of cameras. Testing goals included experimenting with new weapons designs, evaluating weapons reliability and performance, and measuring explosive effects. Now, in the post-nuclear-testing era, preserving these decades-old films is a matter of national security. The Film Scanning and Reanalysis Project, a joint effort between Lawrence Livermore and Los Alamos national laboratories, focuses on digitizing and analyzing the aging, deteriorating films. The team has combed through secure government vaults to inventory and salvage thousands of film rolls. The researchers use modern scanning technology to digitize the films while developing image-processing techniques to extract key data with unprecedented accuracy. Livermore scientists rely on three-dimensional computer models to predict blast effects and energy yield, and the newly digitized films have produced millions of valuable data points. In 2017, for the first time, the public was given access to a batch of these films via YouTube.
Contact: Greg Spriggs (925) 423-8862 (spriggs1@llnl.gov).

U.S. Government Printing Office: 2018/570-075-10030

The Evolution of Livermore's Climate Models



Through Livermore's computing capabilities, originally developed for the weapons program, the Laboratory has become a leader in climate modeling.

Also in December

• Combining optics design and additive manufacturing, Livermore creates a new class of functionally graded optical materials.

• The discovery of reversible carbon-dioxide capture leads to a novel separation technology.

• Experiments indicate that large amounts of hydrogen may exist inside Earth's mantle.

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