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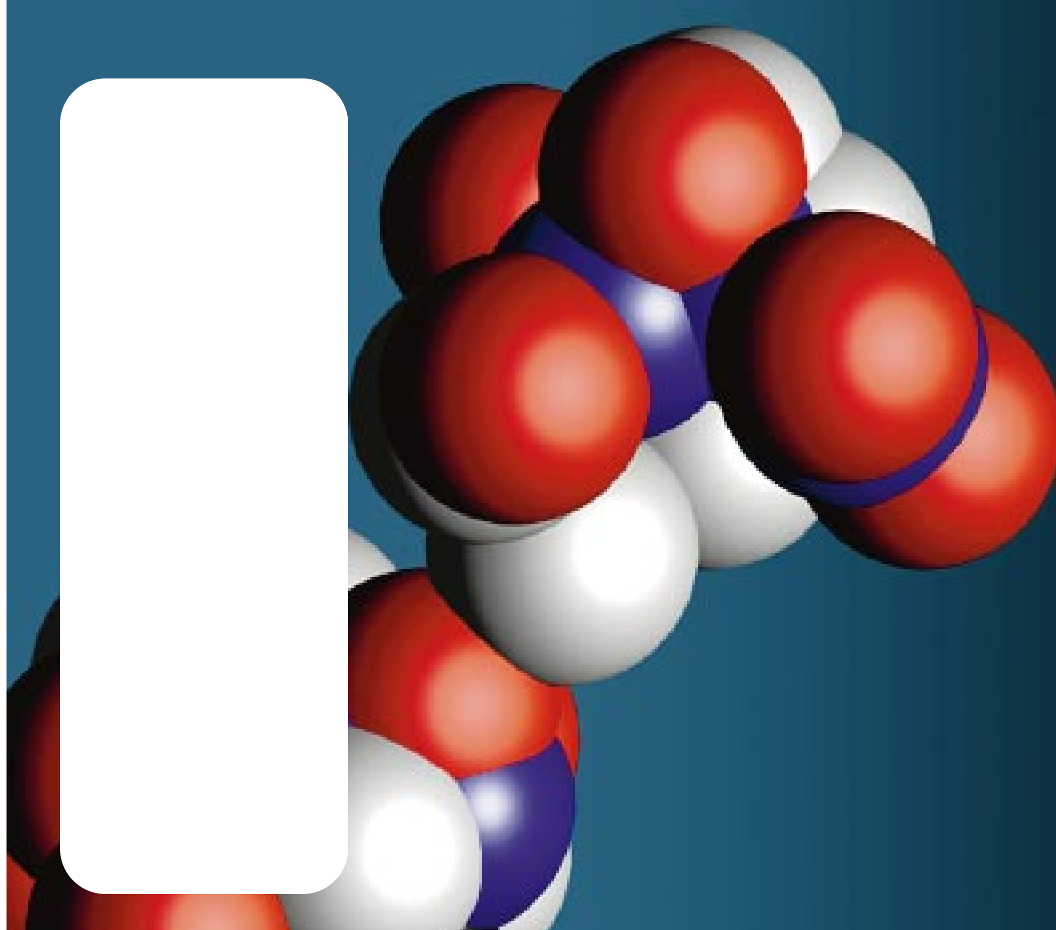

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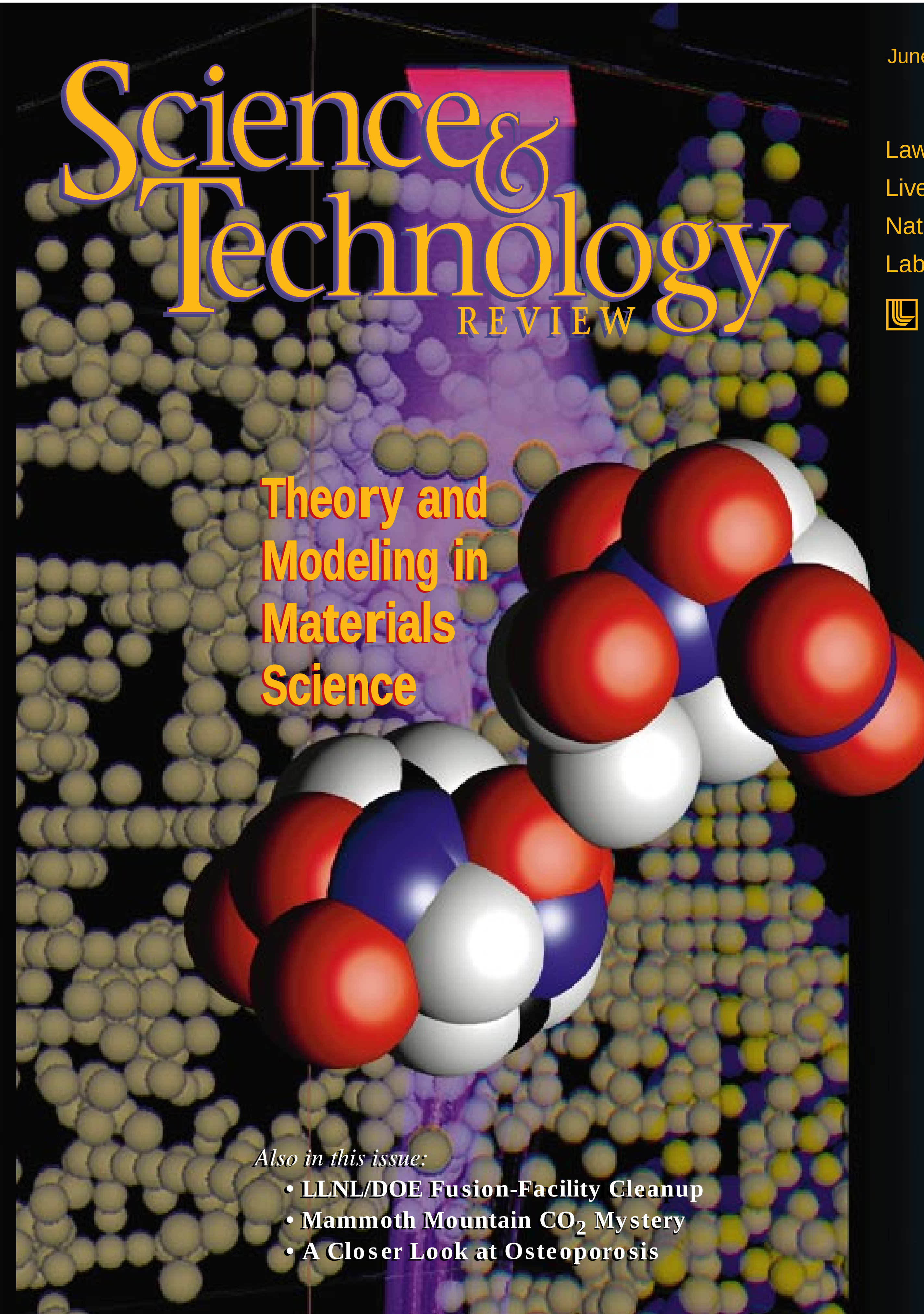

Theory and Modeling in Materials Science

Also in this issue:

- LLNL/DOE Fusion-Facility Cleanup
- Mammoth Mountain CO_2 Mystery
- A Closer Look at Osteoporosis

About the Cover

In the foreground we show a Livermore model in a computer visualization of the molecular behavior (molecules in red. blue, and gray) of a high explosive during a simulated pressure impluse (as in a shock wave). It is superimposed over our structural model of purple smoke flowing through gray aerogel pores. These and other models plus theory and computational technology are providing solutions to support the Laboratory's mission, particularly in science-based stockpile stewardship, energy, and the environment. Our report begins on p. 6.

Cover design and illustration: George Kitrinos and John Madsell

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About the Review

The Lawrence Livermore National Laboratory, operated by the University of California for the United States Department of Energy, was established in 1952 to do research on nuclear weapons and magnetic fusion energy. *Science & Technology Review* (formerly *Energy & Technology Review*) is published ten times a year to communicate, to a broad audience, the Laboratory's scientific and technological accomplishments, particularly in the Laboratory's core mission areas—global security, energy and the environment, and bioscience and biotechnology. 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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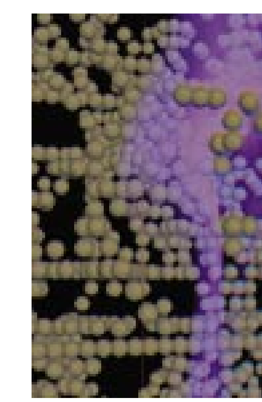

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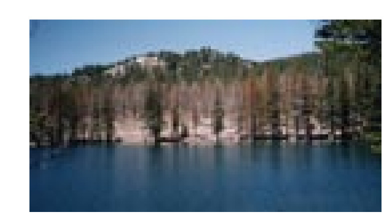

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Lab, Minnesota firm to develop micro x-ray catheter

Interventional Innovations Corporation of Minnesota has entered into a partnership with Lawrence Livermore to produce an x-ray catheter that will prevent formation of scar tissue following angioplasty treatment to clear blocked arteries. Such scarring of artery walls, called restenosis, forms in 35 to 50% of angioplasty patients. The result is reblockage of the artery, which necessitates further treatment.

Experiments have shown that mild radiation treatment of arterial walls immediately following angioplasty is 100% effective in preventing restenosis. The IIC–Lawrence Livermore x-ray catheter would be introduced into an artery following angioplasty to deliver a tailored amount of radiation to artery walls. The new system will eliminate radiation hazards associated with using radioactive isotopes with catheters. Instead, this system generates low-level x rays electrically and only strong enough to treat the arterial walls.

The Laboratory will combine its expertise in x-ray physics and microfabrication with IIC's experience in catheter design and restenosis treatment to develop x-ray catheters as small as 1.5 millimeters in diameter. IIC will provide $1.3 million for the Lab's portion of the work, as well as some $1.5 million to its own effort in the project.

Contact: Dennis Matthews (510) 422-5360 (matthews1@llnl.gov), or Jim Trebes (510) 423-7413 (trebes1@llnl.gov).

Lab, Russians sign fiber-optics contract

In an example of Post-Cold War cooperation, Gennady Yanpolsky, Russia's deputy minister for defense industries, signed a one-year, $100,000 contract with Lawrence Livermore earlier this year for Russian development and fabrication of an advanced fiber-optic system for Livermore.

The low-dispersion, low-loss optical fibers will allow laser beams to be easily transmitted from the laser source to the point of application, where the beams can be used, for example, in computer chip manufacturing or laser diagnostics. Livermore Lab laser scientist Howard Lowdermilk calls the ultraviolet transmitting optic fiber developed by Russia's Vavilov State Optical Institute "a unique technology."

During a visit in March, Yanpolsky and his eight-member Russian delegation held talks with Livermore laser scientists and toured LLNL's Nova and Beamlet lasers, the crystal growth lab, and the Large Optics Diamond Turning Machine.

Contact: Howard Lowdermilk (510) 422-5498 (lowdermilk1@llnl.gov).

Lab teams with Silicon Video on flat-panel displays

Working closely with San Jose-based Silicon Video Corp., Lab scientists have developed a field-emission cathode for low-cost, high-performance flat-panel displays for portable computers. Silicon Video aims to produce a 12-inch field-emission display screen for Hewlett-Packard's OmniBook portable computer in 1997, according to *Electronic Engineering Times*.

The new flat-panel field emission display technology is projected to cost as little as one-half that of active matrix liquid crystal display (LCD) systems now used in laptop computers. The technology also offers better resolution and performance while consuming a fraction of the battery power required by LCD systems.

Tony Bernhardt, program leader of the Lab group working on flat-panel display technologies, says the field-emission technology has the potential to "regain a domestic manufacturing base" in an industry currently dominated by Japan.

Contact: Tony Bernhardt (510) 423-7801 (bernhardt1@llnl.gov).

Weapon dismantlement process reduces waste

Livermore researchers created key components for the Advanced Recovery and Integrated Extraction System (ARIES) that will be shared with Russia to expedite mutual nuclear stockpile reductions. The basic system consists of five modules that remove plutonium from "pits" (the trigger of a nuclear weapon) and repackage it as a metal ingot or oxide powder for eventual disposition. The Lab is responsible for two modules, pit bisection and hydride/oxidation (HYDOX). This process destroys the pit and therefore reduces the number of stockpiled nuclear weapon pits. In addition, the process allows dismantlement to be achieved with less waste.

Contact: Mark Bronson (510) 422-3061 (bronson1@llnl.gov).

Anastasio named Associate Director

Laboratory Director Bruce Tarter selected Michael Anastasio as Associate Director for Defense and Nuclear Technologies. In this position, which he has held in an acting capacity since January, Anastasio will be responsible for leading the Lab's efforts in the nuclear weapons program to assure that the stockpile is safe and reliable. He will also be responsible for ensuring that Livermore has an outstanding staff and a set of core capabilities in nuclear weapons science and technology. In addition, as a member of the Laboratory's Council on National Security, he will work with the Associate Director for National Security and other council members in developing and implementing the Laboratory-wide national security program.

As a major spokesman for the LLNL weapons program, Anastasio has interacted with numerous external senior advisory groups and high-level members of the executive branch, DoD, and DOE. Last year he served a five-month appointment as scientific advisor to the Assistant Secretary of Energy for Defense Programs, assisting in the development of the DOE Stockpile Stewardship and Management Program.

Anastasio joined the Laboratory in 1980 as a physicist in B Division, where he became involved in the design, evaluation, and understanding of systems both in the stockpile and under development. In 1991 he became B Division Leader, a role in which he has served with distinction for the last five years, Tarter noted.

FBI, Navy use Lab's "iWatch" in hacker hunt

FBI and Navy investigators used iWatch, a Lawrence Livermore-developed network monitoring program, in their search for a computer hacker who was attacking government computers. iWatch is part of the Network Intrusion Detector, which began as a group of computer programs to protect Department of Energy computers. It was developed for the FBI and the Navy at the beginning of their investigation last summer.

Following the model of a telephone wiretap, iWatch captures computer-to-computer network exchanges containing specific passwords and programs an intruder uses. After obtaining permission to use iWatch for network "wiretaps," Navy and FBI investigators viewed only short contexts during the hacker intrusions, allowing adjacent unrelated communications to pass unreviewed. As a result of the investigation, a warrant was issued in early spring for the arrest of an Argentine hacker.

Contact: Scott M. Denton (510) 423-7425 (denton2@llnl.gov).

E. European nations receive seismic safety tips

Nuclear power plant regulators and energy personnel in Armenia and the Czech Republic received advice on how to improve the safety of nuclear power plants in seismically active areas during a Lab-organized workshop and training course this spring. The activities were put together by Bob Murray, leader of the Geologic and Atmospheric Hazards Project in LLNL's Environmental Programs Directorate.

In 1995, Murray visited Hungary and the Slovak Republic to speak at special gatherings of Eastern European state regulators on various methods of seismic safety. His most recent trip was the result of a request by Czech representatives that he visit their country to update regulators and researchers on lessons learned in the U.S. from recent earthquakes. Accompanying Murray were several Lawrence Livermore subcontractors, all from California consulting firms. At least one team member is expected to return in July to offer further suggestions for seismic retrofits.

Contact: Bob Murray (510) 422-0308 (murray6@llnl.gov).

Patents and Awards

Each month in this space we report on the patents issued to and/or the 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.

Patents

Patent issued to	Patent title, number, and date of issue	Summary of disclosure
Richard R. Leach Farid U. Dowla	**Real-Time Neural Network Earthquake Profile Predictor** U.S. Patent 5,490,062 February 6, 1996	A network that uses first-arrival energy to predict the characteristics of impending earthquake seismograph signals. The neural network produces a profile of the complete earthquake signal using data from the first seconds of the signal.
Robert D. Paris Richard P. Hackel	**Method and Apparatus for Monitoring the Power of a Laser Beam** U.S. Patent 5,490,157 February 6, 1996	A method that discloses how to insert an optical fiber in the path of a laser beam so that laser radiation impinging on the lengthwise outer surface is coupled into the inner core.
Daniel M. Makowiecki Joseph B. Holt	**Reactive Multilayer Synthesis of Hard Ceramic Foils and Films** U.S. Patent 5,490,911 February 13, 1996	A method involving the sputter deposition of alternating layers of reactive metals with layers of carbon, boron, or aluminum and the subsequent reaction of the multilayered structure to produce a dense crystalline ceramic.
Bruce E. Warner William McLean, II	**Apparatus for Laser Assisted Thin Film Deposition** U.S. Patent 5,490,912 February 13, 1996	An apparatus that uses fiber optics to deliver visible output beams. Optical fibers are coupled to one or more laser sources and deliver visible output beams to a single chamber, to multiple targets in the chamber, or to multiple chambers.
William A. Brummond Ravindra S. Upadhye	**Injector Nozzle for Molten Salt Destruction of Energetic Waste Materials** U.S. Patent 5,491,280 February 13, 1996	A nozzle that rapidly injects a safe mixture of energetic material with a carrier gas into a molten salt reactor to prevent premature detonation.
Stanley W. Thomas	**Collimator Application for Microchannel Plate Image Intensifier Resolution Improvement** U.S. Patent 5,495,141 February 27, 1996	An angle-adjustable collimator inserted in contact with or slightly above the phosphor screen to cause electrons entering at an angle greater than the collimator acceptance angle to strike the collimator walls and be prevented from reaching the phosphor screen.
Abelardo L. Ramirez John F. Cooper William D. Daily	**Using Electrokinetic Phenomena and Electrical Resistance Tomography to Characterize the Movement of Subsurface Fluids** U.S. Patent 5,495,175 February 27, 1996	A method using electrokinetic transport to enhance the ability of electrical resistance tomography (ERT) to detect position and movement of subsurface contaminant liquids, particles, or ions, and for subsurface imaging of soil and rock properties.
Richard F. Post	**Dynamically Stable Magnetic Suspension/Bearing System** U.S. Patent 5,495,221 February 27, 1996	A system that achieves a state of stable equilibrium above a critical speed by using passive elements with permanent magnets to provide their magnetomotive excitation.

Awards

The Federal Laboratory Consortium presented four awards for **excellence in technology transfer** to LLNL researchers. **Dan Thompson** received one for machining and engineering with U.S. companies and directing a series of machine tool metrology workshops. Chemist **Dan Makowiecki** received one for designing a new type of magnetron sputtering source that has been licensed to a Bay Area company. **Alfred Goldberg, Don Lesuer, Mike Strum, Stephen Root, Dick Landingham,** and **Paul Curtis** received one for transferring a superplastic steel technology to two companies. A fourth award was given to former LLNL engineers **Robert Whirley** and **Bruce Engelmann**, who helped transfer the computer program DYNA3D (used in analyzing impacts upon structures) to private industry.

In recognition of his sustained superior performance in the area of nonproliferation, **Delbert F. Wright** was awarded the **Intelligence Community Seal Medallion** by John Deutsch, Director of U.S. Central Intelligence. The February presentation honored Wright's work in nonproliferation policy and operations.

Commentary on Materials Modeling and Stockpile Stewardship

Michael R. Anastasio
Associate Director
Defense and Nuclear Technologies

IN late September 1995, President Clinton issued a directive that provided Lawrence Livermore and its fellow nuclear weapons research laboratories, Los Alamos and Sandia, with a new and extremely challenging mission—science-based stockpile stewardship.* The DOE, in cooperation with the national security laboratories, initiated the Stockpile Stewardship and Management Program to meet nuclear weapons responsibilities when our nation halted both nuclear weapons testing and new weapons development and production as it pursued negotiation of the Comprehensive Test Ban Treaty. Stockpile stewardship is the assurance that in the absence of nuclear weapons testing and new weapons production and development—and in the presence of continued stockpile downsizing, dismantlement, and aging—the U.S. can retain confidence in the safety and reliability of its nuclear arsenal.

Since the President's announcement, scientists and top managers at Livermore, Los Alamos, and Sandia have been hard at work doing the long-term strategic planning necessary to meet the challenge of implementing the stockpile stewardship program. It has become abundantly clear during our planning that materials issues must be at the heart of our efforts if the stockpile stewardship program is to succeed.

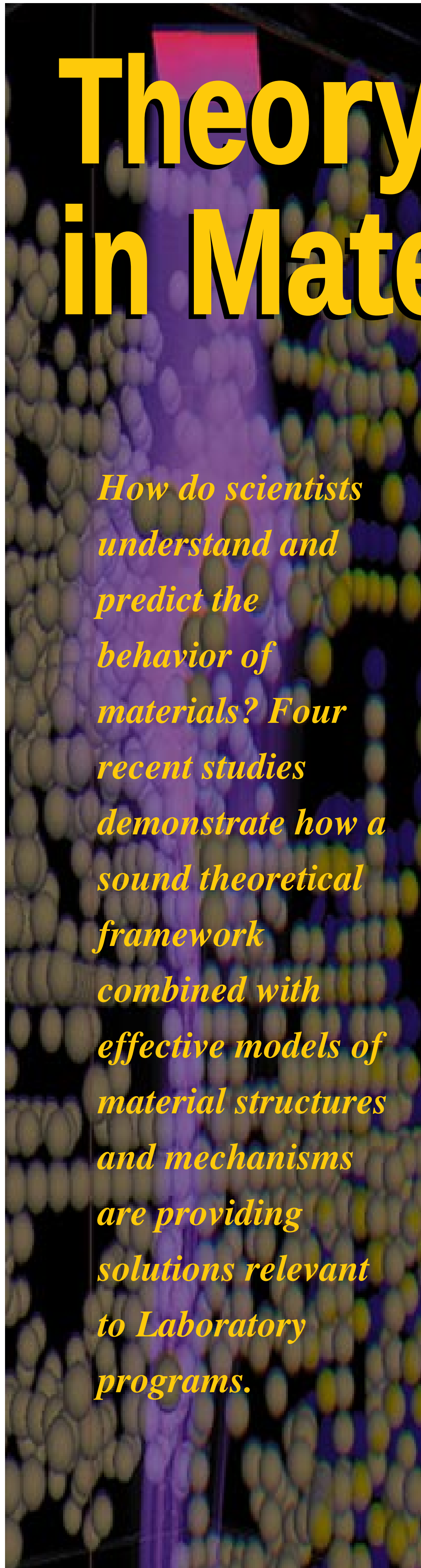
The stockpile of which Livermore, Los Alamos, and Sandia are the stewards is older than at any time in our nation's history—and it ages daily. In the absence of testing, scientists still need answers to numerous questions about the aging materials in stockpiled weapons if they are to predict performance and assure safety and reliability. Questions include: How will radiation damage affect material strength? How will temperature variations affect an explosive's sensitivity? Does the material used in a replacement component have the same properties as the material in the original component?

In the past, when scientists needed to know the effects of age on the inorganic (metal) and organic (plastic) materials that make up a nuclear device, they tested a weapon with aged components and used complex computer codes and their own experience and scientific knowledge to interpret the results. In the future, they will reach judgments about the effects of aging on the performance and safety of these devices through a more detailed understanding of weapons materials garnered from sophisticated computational modeling capabilities and non-nuclear experiments, plus their own knowledge of weapons physics and their experience with weapons materials.

It is in this context that we begin to understand the real challenge of stockpile stewardship and how materials modeling will be invaluable to us in meeting this challenge. The article on materials modeling beginning on p. 6 discusses recent advances in materials modeling and their relevance to Laboratory programs, particularly stockpile stewardship. The combination of sound theory, effective models of weapons materials, and experimental validation of those models will enable us, in the absence of nuclear testing, to understand and predict the effects of aging on weapons performance and to develop new or replacement materials. These advanced computer simulations become important contributors to judgments assuring the safety and reliability of our nuclear stockpile.

The Laboratory is indeed well positioned to do the materials modeling fundamental to stockpile stewardship. For many years—and in a variety of applications—we have been using materials modeling from the atomic level to the system, or continuum, level to develop new materials, like aerogels, and to determine the effects of aging and exposure to hostile environments on materials such as silicon, metals, and explosives. It is these highly sophisticated and well-honed materials modeling capabilities that will serve us well as we explore the effects of aging on the materials that comprise integrated weapons systems, which must perform predictably in perhaps the most hostile environment imaginable—a nuclear weapon. Materials modeling provides this nation's stockpile stewardship scientists with one of their most useful and important capabilities for validating the safety and reliability of our nuclear deterrent in the absence of testing.

**Science & Technology Review*, November/December 1995, p. 2.



Theory and Modeling in Materials Science

How do scientists understand and predict the behavior of materials? Four recent studies demonstrate how a sound theoretical framework combined with effective models of material structures and mechanisms are providing solutions relevant to Laboratory programs.

EVER since our ancestors first used tools to make tasks easier, understanding the properties of materials has been a practical concern. The challenge of explaining how modern materials behave is driven by the vast range of new materials and processing methods that are available and by the demands placed on performance, sometimes in harsh or unusual environments.

The cessation of nuclear testing and the advent of science-based stockpile stewardship as a primary Laboratory mission increase the challenge. Today, we need to predict changes in the structure and properties of materials in stockpiled warheads and the effects of these changes on how weapons perform. Success in fulfilling the stockpile stewardship mission will also provide far-reaching benefits to other Laboratory programs and the commercial sector.

One way scientists study material properties is by applying fundamental physical and mathematical principles to form the basis of models. By combining models with spectacular advances in computational technology, we can often shed light on the mechanisms that determine how a material behaves. Furthermore, theory and modeling in materials science are often directed toward predicting, not just describing, the properties of materials. Models have progressed to a point that they can often tell us not only what happens, but how or why it happens.

Lawrence Livermore scientists have an arsenal of tools and devices to model the behavior of materials without always resorting to experiments that can be expensive. On the other hand, experiments are usually used to validate models, so theorists and experimenters often work together.

Today in the Chemistry and Materials Science Directorate we are addressing increasingly complex phenomena and a broad range of problems in materials science relevant to Laboratory programs. Examples of our current modeling capabilities include:

- The evolution of microstructures, such as the formation and growth of voids produced by radioactive decay or irradiation of materials.
- The performance and degradation of high explosives and polymers.
- Alloy properties, such as phase diagrams.
- Analysis of spectroscopic scattering data.
- Metals processing.
- Corrosion damage.

These topics and many others also have important applications in defense, industry, and other sectors. The diverse materials we model include aerogels, alloys, ceramics, high explosives, metals, and polymers, to name only a few. The breadth of our modeling capabilities means that we cover length scales starting from atoms and electrons at the submicroscopic level, to grains and grain boundaries at intermediate

These materials scientists use a variety of approaches to solve materials problems described in this article. (Left to right) Standing: Daniel Calef, modeling of aerogels; Lloyd Chase, division leader; and William Gourdin, physically based models of tantalum deformation. Sitting: Larry Fried, molecular dynamics and phenomenological modeling of high explosives; and Tomas de la Rubia, kinetic Monte Carlo modeling of ion implantation and defects in silicon.

lengths, to finished components at the opposite end of the spectrum.

About Length Scales

The concept of modeling on all relevant length and time scales is fundamental in our research; Table 1 illustrates the concept. Materials generally have a wide range of internal structures that determine their behavior and performance. Our objective is to predict, explain, and sometimes control properties across the full range of material structures, which span spatial dimensions from a fraction of a nanometer to meters. (A nanometer is one billionth of a meter; a typical atom is about 0.3 nm in diameter.)

At the shortest lengths and times relevant to materials properties, atoms and electrons determine characteristics such as a material's hardness, conductivity, and optical properties. Sometimes we are able to calculate the behavior of a material based on quantum-mechanical theory alone. In that case, we call the process a "first-principles" calculation because we essentially do not use or need any experimental input. About all we need to know is the atomic numbers of the atoms involved and sometimes their positions. First-principles calculations increase our understanding of materials by allowing us to make predictions, reveal trends, test hypotheses, and analyze experimental data.

First-principles calculations form the basis for many of our modeling activities at Livermore.[1] Examples include the properties of metals and alloys, the behavior of surfaces and interfaces, and the modeling of experimental measurements. Because first-principles theory and modeling were discussed extensively in the August/September 1994 issue of *Energy & Technology Review*,[2] this article emphasizes the other approaches.

At increasing length scales in Table 1, we study the properties associated with larger structures by using approaches such as molecular dynamics (MD), kinetic Monte Carlo, or phenomenological models. Models associated with greater lengths are increasingly based on the empirical or measured responses of materials to stress, deformation, temperature, and other factors. By combining several approaches, we can deal with the wide variety of physical properties we need to assess. Illustrating diverse approaches to modeling across a range of material structures and properties, the following four examples of recent accomplishments are only a few of our many modeling efforts in progress.

Defects in Silicon

Over the last 30 years, exponential growth of the semiconductor industry has been driven toward denser packing of smaller components that make up a silicon chip. To develop the silicon chips required for microelectronics components in the 21st century, we need to understand more about how defects are produced and how dopants diffuse in silicon.

Dopant atoms are required to make silicon usable for manufacturing semiconductor devices. During manufacturing, dopants are routinely implanted (using ion accelerators) into very precise regions of a silicon wafer. This process damages the silicon wafer by introducing defects that must be removed. At the high temperatures used for the removal process, the defects and dopant atoms interact and diffuse over long distances. Dopants therefore can end up at destinations different from their intended location in a wafer. When that happens, the defective devices are not suitable for the marketplace.

We are creating a "virtual laboratory" to study this problem and to model other types of radiation effects in materials. Our strategy is to use an experimentally validated hierarchy of theoretical and computer simulation tools to span many length and time scales, from picoseconds to minutes. At the shortest lengths (at atom level) and times (up to about a nanosecond), we use MD simulations based on forces between atoms that accurately reproduce relevant properties of the material. Over time, defects in silicon can aggregate to form larger structures, like dislocations. To study how such structures evolve over longer times (minutes or hours), we use kinetic Monte Carlo simulations. In this work, we have a collaboration with scientists at AT&T Bell Laboratories, which allows us to develop a new capability to support other Laboratory programs.

Table 1. Theory and modeling activities at Livermore cover all of the length scales associated with material structures.

Material structure	Length scale	Primary theory or model used
Atomic or electronic	Angstroms ($1\ \text{Å} = 10^{-10}$ m)	QM
Simple defects (vacancies, point defects, interstitials)	Angstroms to nanometers ($1\ \text{nm} = 10^{-9}$ m)	QM MD KMC
Extended defects (dislocation, cores, small voids, clusters, and precipitates)	10 to 100 nm	QM MD PM
Nanoscale to microscale structures (grain boundaries, grains, precipitates)	10 nm to 100 µm ($1\ \mu\text{m} = 10^{-6}$ m)	QM MD PM Phen
Polycrystallines, composites, and interfaces	Micrometers to meters	PM Phen
Continuum (i.e., auto or bridge)	Varies	CM

Quantum mechanics (QM) forms the rigorous theoretical basis for studies of electrons and atoms, chemical bonds, molecular structures, interfaces, and defects—the smallest structures that determine how a material behaves.
Molecular dynamics (MD) calculates the motions of atoms or molecules combining Newton's laws of motion with quantum-mechanical understanding, e.g., modeling the collisions of high-energy particles with the atoms of a solid undergoing radiation damage.
Kinetic Monte Carlo (KMC) models are used to study how atoms and defects in a material diffuse spatially by discrete jumps. The probability of a jump is determined by temperature and energy barriers for the movement.
Physically based models (PM) are based on physical concepts that emulate the behavior of material structures, e.g., dislocation movement, grain-boundary sliding, crystallographic twinning, and material movement.
Phenomenological models (Phen) use mathematical relations without any known physical basis to describe experimental observations.
Continuum models (CM) treat structures, such as a car frame or beams of a bridge, as a continuous or homogeneous material, e.g., the process of forging an automobile bumper.

Recent computer simulations based on our models are giving us a clear and consistent physical picture of the production and evolution of damage in silicon under energetic-beam bombardment. A typical simulation begins with a cube of silicon made of about one million atoms in a normal lattice arrangement. Then we simulate the bombardment of the top of the cube with high-energy ions to implant arsenic, boron, or other dopant atoms. Figure 1 shows the defects—that is, displaced atoms—in a silicon cube. We can simulate the full range of beam energies that are typically used to process silicon devices, from about one-tenth of an electron volt to several thousand electron volts. As the energy increases, the amount of total damage increases, as expected, but we also find that the size of the largest defect clusters increases.

Our simulations produce images that look as though they come from a high-resolution microscope. We validate the simulations by comparing them with damage observed in actual materials, as shown in Figure 2. Comparisons like this confirm that our computer-aided design package accurately predicts experimental results.

Our work on semiconductor devices also applies to a range of other problems. For example, the walls in nuclear power plants undergo radiation damage from neutron bombardment. Similar processes may occur in nuclear weapons components. Our simulations can help predict the performance of materials used in weapons, existing fission power plants, and fusion plants that may be developed in the future. Because void formation is also seen in

metallic nuclear fuel rods and other structures, the modeling of defects and voids has applications to these problems as well.

Deformation in Tantalum

Anyone who has attempted household plumbing knows that copper tubing becomes more difficult to work by hand after repeated bends. This phenomenon, known as work hardening, occurs in many metals. The increase in strength is caused by interactions between lattice defects called dislocations.

Dislocations consist of extra or unequal planes of atoms, like an extra sheet of paper slipped part way into a stack of sheets. Another handy way to imagine dislocations is to think of them as "wrinkles" in the regular arrangement of atoms in a metal crystal—much like wrinkles in a rug. Imagine creating a small wrinkle at one end of a rug and then pushing the wrinkle along to the other end. In a similar manner, atoms in a metal lattice can be moved relative to each other by creating a dislocation and then moving it through the crystal. Like a wrinkle in a rug, dislocations are long, string-like defects. When many are present, they tangle like spaghetti. In metal, the more dense the tangles, the more energy is needed to deform it.

Copper belongs to a class of common metals with a simple structure known as face-centered cubic—a cube of atoms with an additional atom on each face. Another group of metals, including iron and tantalum, has a body-centered cubic (bcc) structure with atoms at the corners of a cube and one atom in the center. Because these metals are technologically important, their mechanical behavior is of considerable interest.

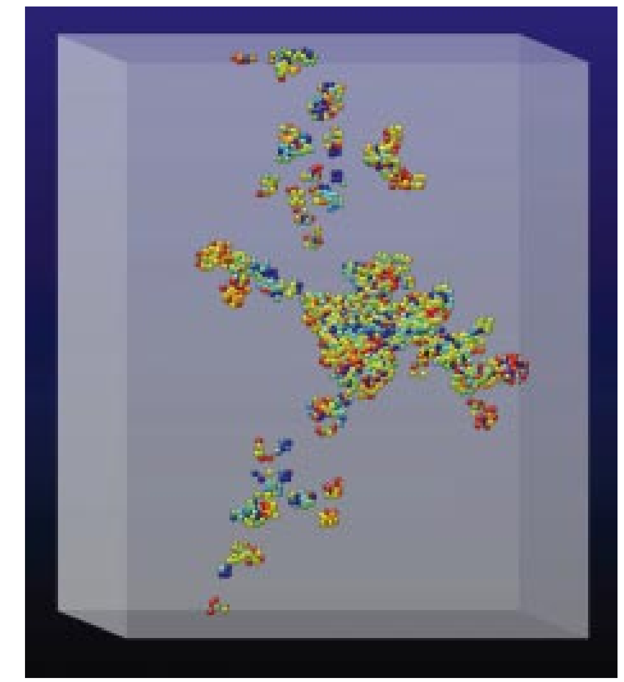


Figure 1. Monte Carlo computer simulation of displaced atoms in a cube of translucent silicon after implantation with 15-keV arsenic ions. The unaffected silicon atoms are not shown here. The atoms in blue are under tensile stress and represent areas with vacancies; the atoms in red are in compressive stress and indicate the presence of interstitials. The large mass in the middle is an amorphous zone; i.e., the crystalline order has been destroyed.

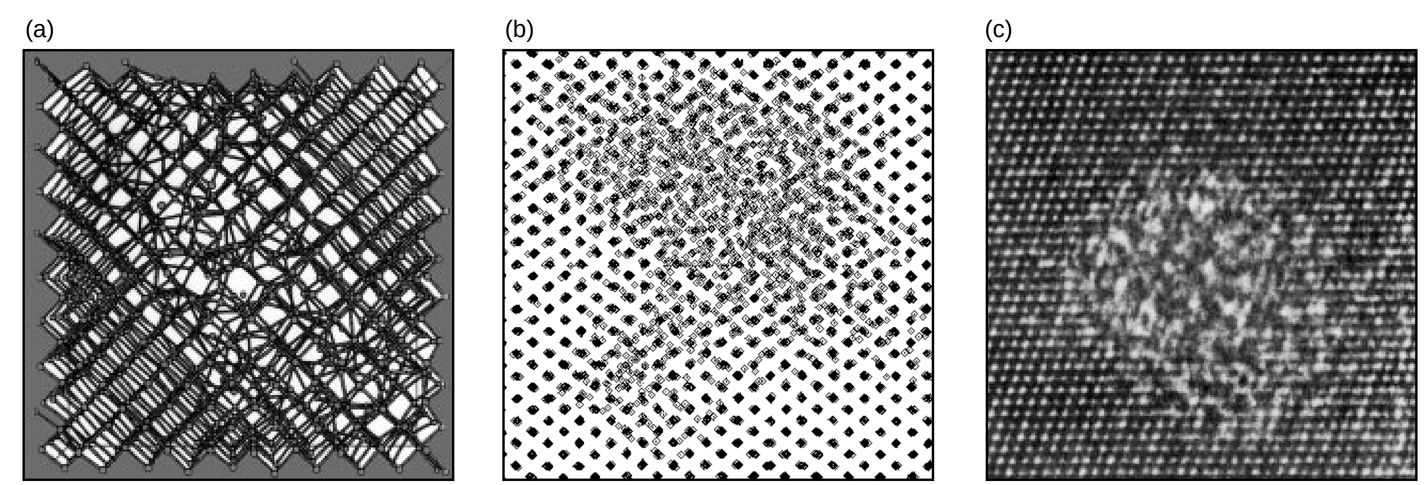


Figure 2. (a) Three-dimensional plot of damage created by a 25-keV platinum ion in silicon. A highly disordered, amorphous region is surrounded by crystalline material. (b) A two-dimensional projection of the atoms in (a). (c) An experimental high-resolution electron micrograph done elsewhere[3] of the impact of a 100-keV bismuth ion in silicon. Although the exact conditions of the simulation and experiment are not identical, this type of comparison helps us to validate the simulation and to interpret the experimental observation.

Our goal is to understand the mechanical behavior of bcc metals and to include enough physics concepts in the model so that calculations can be meaningfully extrapolated to new conditions. The problem is beyond the reach of quantum mechanical calculations. Instead, we are using physically based models that are realistic in representing the actual processes that control deformation. Tantalum is a good test case for this work because it is ductile, shows substantial work hardening, and has important defense applications.

Our model for tantalum accounts for both yield stress (force per unit area at which it begins to permanently deform) and work hardening. Previous explanations said nothing about work hardening and did not explain it for this class of materials. We suggest that there are two (or possibly more) barriers to moving a dislocation, as illustrated by the humps in Figure 3. At first, a dislocation in tantalum must move as if it were isolated, and enough force must be applied to overcome a series of small barriers. In the analogy of wrinkles in a rug, even if no other wrinkles block the path, some force is still required to move an isolated wrinkle. (The material in front of a wrinkle must be lifted as it moves forward.) After moving a certain distance, however, a dislocation may encounter a barrier produced by other dislocations. The force to overcome this barrier increases with deformation and accounts for work hardening in a natural way.

Our model combines the two mechanisms, yield stress and work hardening, and is able to describe which one dominates at different stages and under different conditions of deformation. Figure 4 shows how well the model can reproduce the observed mechanical behavior of tantalum at room temperature. We find similar agreement when temperature is varied.

What is the model good for? With the increased power of modern computers, companies like automobile manufacturers can now simulate the forming and performance of key structural components. However, computer simulations are only as good as the underlying models used to describe the behavior of materials under conditions that are often severe (for example, crashes). Physically based models more realistically describe material properties, yield more meaningful results, and can be reliably extended beyond the scope of experimental data. Whereas the current Livermore model for the deformation of tantalum was conceived for bcc metals, it provides a framework for face-centered cubic metals as well.

Modeling High Explosives

Energetic materials, which include high explosives, are widely used in both military and civilian applications. Livermore has studied high explosives for decades because they are crucial to the performance of nuclear weapons. In the area of stockpile stewardship, we studied how shock dynamics change in older, degraded materials. In another recent

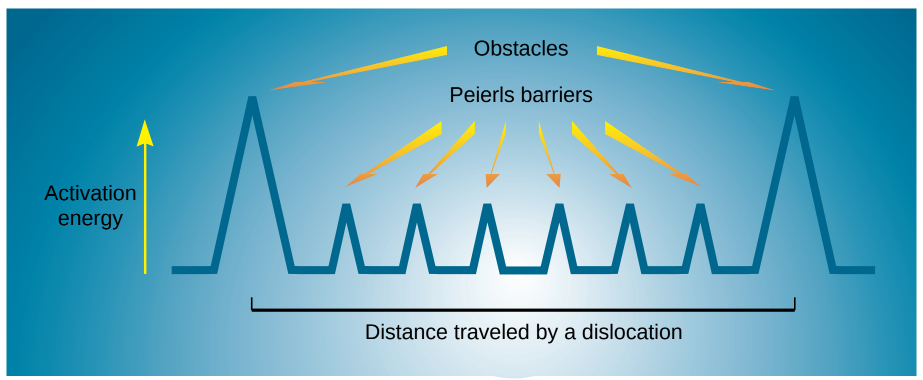
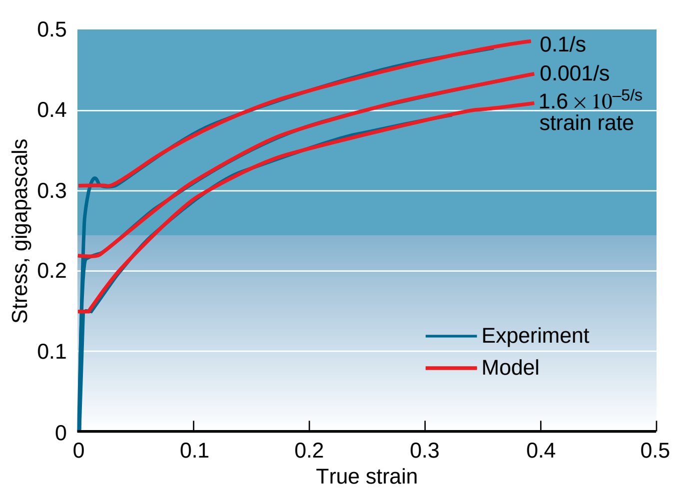


Figure 3. How to envision the two barriers to plastic flow in tantalum: The distance along the bottom refers to the distance traveled by a moving dislocation when a material is deformed. The Peierls barriers are associated with the motion of isolated dislocations (analogous to wrinkles in a rug). The larger obstacles occur where dislocations intersect.

Figure 4. Our model accurately reproduces experimental values of stress (force applied per unit area) and strain (relative change in dimensions) for various strain rates (rates of deformation) in unalloyed tantalum at room temperature.



project, we developed a candidate bunker-busting munition for the Air Force following their experiences in the Gulf War. In the civilian sector, the Bureau of Mines needs to evaluate explosives for mining operations. To better assess environmental concerns, we need to know what reaction products are generated following a detonation.

Typical energetic materials are made of large, floppy molecules with more than 20 atoms, and they can undergo a variety of chemical reactions. Over time, such molecules can degrade and the crystals become more porous, making them dangerous to handle. At the atomistic level, we are simulating how the propagation of a shock wave through high explosives is affected by the degree of degradation. On a macroscopic scale, we can model the performance of existing and novel energetic materials.

As one example of new work on the atomistic scale, we are applying MD simulations to study how the shock properties of the widely used explosive triaminotrinitrobenzene (TATB) change as a function of its degradation and increased porosity. By using this advanced capability, we can assess how an explosion is initiated on a molecular level in aged material found in weapons stockpiles.

As shown in Figure 5, we simulate crystals of about 10,000 TATB molecules and apply a shock wave (a simulated pressure impulse) to crystals with different degrees of defects. We found that the shock wave in degraded material travels much more slowly and spreads out over a much wider area than in pure TATB. At the molecular level, the collapse of voids leads to hot spots in degraded (porous) TATB, and the temperature behind the shock front becomes higher and much more nonuniform.

To understand how molecules like those in TATB react on a much larger scale, we have developed the CHEETAH computer code, a phenomenological thermochemical model to predict the performance of explosives.[4] In contrast to our MD simulations, this more mature modeling effort looks at macroscopic events at lengths of centimeters to meters. The code is empirically based and is derived from more than 40 years of experiments on high explosives at LLNL.

CHEETAH models the interactions (for example, the electrical potentials) of a mix of molecules between them to predict a variety of outcomes, such as those shown in Figure 6. If we think of explosives as a bucket of hot chemical soup, CHEETAH acts like a thermometer and pressure gauge. It predicts the reaction products and the detonation properties, such as pressure, velocity, and energy. The code allows us to vary the recipe (chemistry) and the starting conditions to optimize the properties we want, such as the best early- or late-time energy.

The value of CHEETAH is that it predicts the performance of a given amount of high explosives to within a few percent. With libraries of about

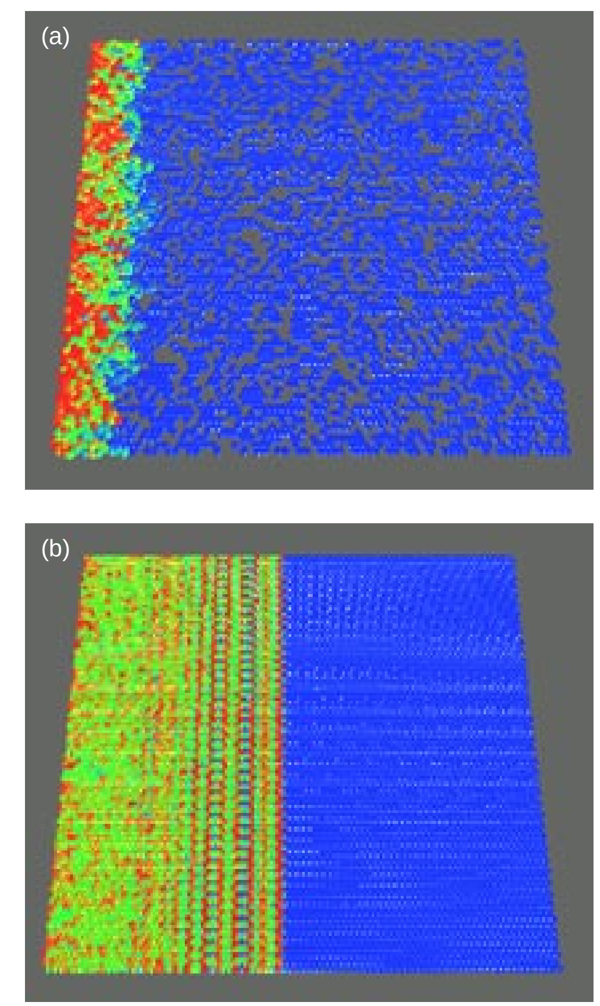


Figure 5. "Snapshots" of molecular dynamics simulations for (a) pure and (b) degraded TATB. The molecules are shaded according to their kinetic energy as a shock front passes through the lattice, with red corresponding to higher temperatures and purple to lower temperatures. In contrast to a sharp, smooth shock front in pure material, porous TATB produces a broader and less uniform shock front with hot spots.

100 reactants and 6,000 products, the program is now used by more than 80 research teams in industry, academia, and the international scientific community, including England, Canada, Japan, Sweden, and France.

The code is both physically simple and user friendly, and it can guide applications ranging from rocket and gun propellants to the formulation of new explosives with improved performance.

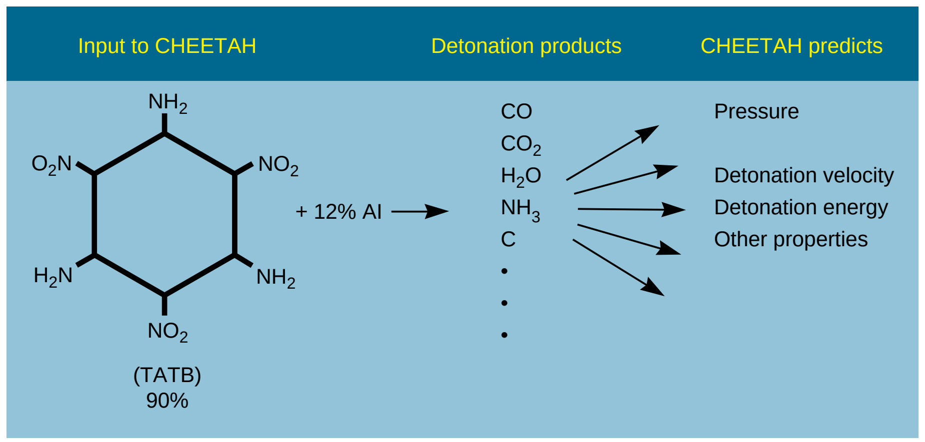


Figure 6. To predict the performance of explosives, CHEETAH starts with one or more base reactants, such as TATB and metallic aluminum. It then solves thermodynamic equations to predict the detonation products and their properties, such as temperature and volume. From these values, CHEETAH predicts the detonation properties, including pressure, velocity, and energy.

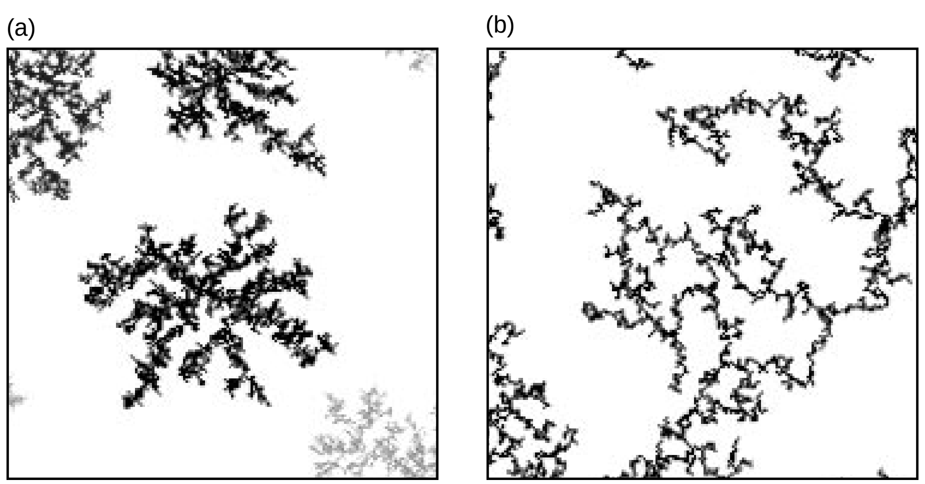


Figure 7. We model aerogel structures by varying the number of starting particles and the rules by which they move and adhere to one another. Compared to (a) clusters grown from fixed seeds, (b) cluster–cluster aggregates more accurately mimic real aerogels.

Transport Through Aerogels

Aerogels have exceptional strength and enormous surface area and are among the lightest solids known.[5] Some varieties are 100 times less dense than water. LLNL first studied aerogels for a national defense application, but their use is being proposed as electrical, thermal, and sound insulators; optics, space, and catalyst devices; capacitive deionization units for water purification; aerocapacitors for energy storage; and various novelties and toys. Many aerogel applications remain relevant to Laboratory programs focused on national defense, the environment, and energy. Their use as filters and catalyst supports take advantage of their tremendous surface area.

At the microscopic scale, these highly unconventional solids are made of "beads" that are some tens of nanometers in size. At an intermediate length scale (in the range of micrometers), groups of beads are clustered to form an open network with large and small voids or pores in the network. To understand how molecules flow through an aerogel, as they would in a filter, we need accurate structural models and flow codes for highly irregularly shaped networks.

Developing these models was a considerable challenge because the absence of any characteristic pore size in an aerogel complicates the treatment of fluid flow. We have replicated the structure of aerogels at the intermediate scale by simulating the growth of clusters.[6]

In the models, particles on the order of 10 nanometers wide represent the beads. These particles or "walkers" randomly move through a three-dimensional lattice and stick to each other. Both the number of walkers and the sticking rules are varied in different simulations. For example, if walkers only cluster around a set of fixed particles, then structures like those in Figure 7a are created. Alternatively, if walkers adhere to each other and the clusters continue to diffuse, then we generate structures like those in Figure 7b, called cluster–cluster aggregates.

When we make the structures more like those in Figure 7b, they act more like a simple, random distribution of obstacles, and they more accurately mimic the structure and behavior of real aerogels. A commonly measured quantity for flow through porous materials is permeability. In comparisons of calculated permeabilities based on our models, the cluster–cluster aggregates closely match the observed experimental behavior for the flow of a gas through aerogels.

Figure 8 shows a puff of smoke flowing through one of our modeled aerogels. This visualization, developed by the Livermore Computer Center graphics laboratory, clearly shows that the flow patterns are dominated by the largest pores. Such results reinforce the view that our approach successfully models these highly irregular and unconventional solids.

Work to Come

What does the future hold for theory and modeling of materials properties at LLNL? To accomplish our stockpile stewardship mission, we must improve our ability to predict how the structures of metals, high explosives, and polymers change with time or vary with manufacturing methods. Then, we need to assess the effects of these changes under the extreme conditions relevant to weapons performance. For this purpose, we need robust models that can be used reliably. We are collaborating with Laboratory colleagues in the Physics and Space Technology and Engineering Directorates, as well as with researchers at many universities, to develop the required approaches.

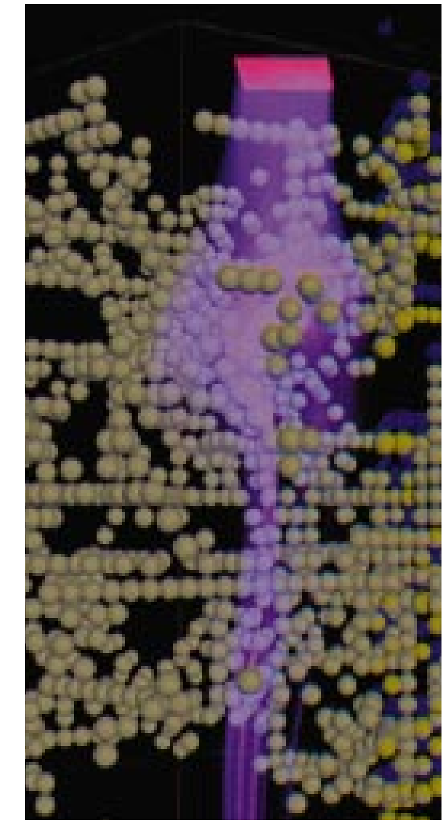


Figure 8. A puff of smoke flowing through an aerogel shows that the flow patterns are dominated by the largest pores.

Key Words: computer modeling, materials science, material structure, microstructures, molecular dynamics.

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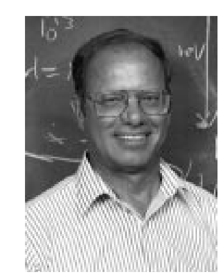
About the Scientist



LLOYD L. CHASE is the division leader for Materials Science and Technology in the Chemistry and Materials Science Directorate. He joined the Laser Program at LLNL in 1985, where he did optical materials research and development. He transferred to Chemistry in 1991. He received a B.S. in engineering mechanics in 1961 at the University of Illinois and a Ph.D. in physics at Cornell University in 1966. Before coming to the Laboratory, he was on the technical staff at Bell Telephone Laboratories and professor of physics at Indiana University. His areas of research have been in solid-state physics and materials science with an emphasis on development and characterization of optical materials. He has more than 130 publications in these fields and holds three patents.

LLNL and DOE Collaborate on Successful Fusion Facility Cleanup

Livermore expertise in handling tritium and low-level radioactive waste, combined with careful planning and multidisciplinary teamwork, led to success.

CAN an industrial building that has been contaminated by large amounts of radioactive and toxic materials be cleaned up well enough to be returned to general use? Thanks to Lawrence Livermore's decontamination expertise, the answer is yes.

Livermore and DOE's Oakland Operations Office teamed up to decontaminate, decommission, and close out—on time and under budget—the Ann Arbor Inertial Confinement Fusion Facility in Michigan. This abandoned facility, which KMS Fusion had used for laser fusion experiments (funded in part by the DOE) from 1978 through 1991, included 60 chemistry laboratories associated with loading tritium into millimeter-size glass laser targets. Thousands of mostly laboratory-size containers of chemicals and solvents, some containing tritium, were scattered around the 9,000-square-meter (100,000-square-foot) building. Initially, there was some question whether it could be decontaminated or would have to be razed.

At DOE–Oakland's request, the Laboratory took over technical oversight of the decontamination and decommissioning effort in April 1994 and, working side-by-side with DOE–Oakland personnel, successfully completed the work a year later—two weeks early—and within the agreed-upon $2.5-million budget. The effort was first estimated to take three years and cost $5 million.

DOE selected Livermore for this effort because of its existing expertise in handling bulk tritium and low-level radioactive waste and for its ability to quickly assemble multidisciplinary teams to meet project objectives under tight time and dollar constraints. Livermore recently demonstrated these capabilities in the decontamination and decommissioning of its own Tritium Facility (see article, *Energy & Technology Review*, March 1995).

"Cleaning up a facility contaminated with tritium involves three activities: decontamination, decommissioning, and closeout," said Mark Mintz, manager of Livermore's Tritium Facility and leader of the Laboratory's overall effort (see Figure 1). "Decontamination involves removal of radioactive or chemically hazardous substances. Decommissioning is the shutdown, if necessary, and removal of all the experimental and laboratory equipment and office furnishings. Closeout essentially means proper disposition of the government property in the facility." And certification of the wastes had to occur before removal and disposition could take place. (See box pp. 16–17.)

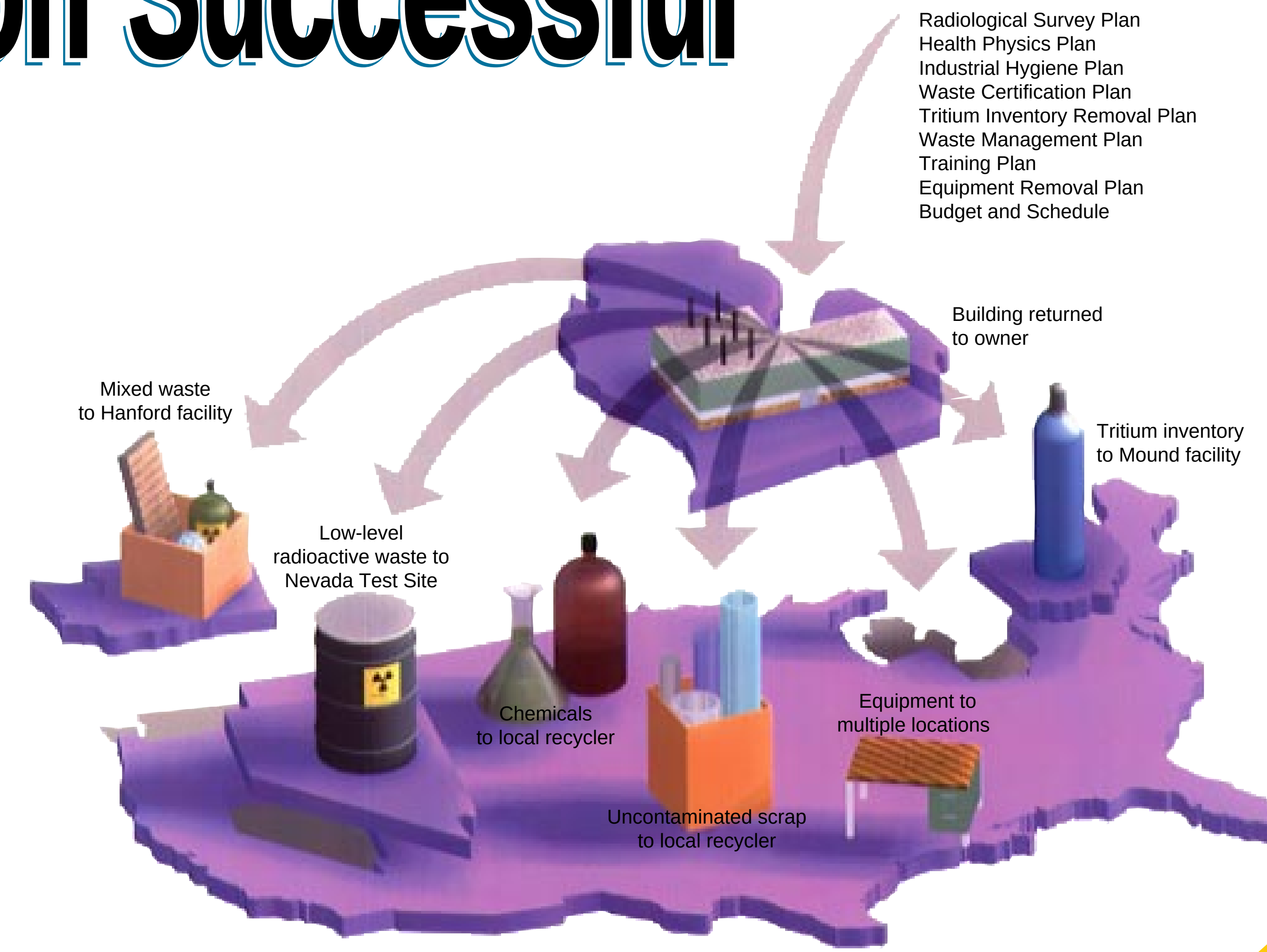


Figure 1. Plans for decontamination, decommissioning, and closeout of the Ann Arbor Inertial Confinement Fusion Facility in Michigan were key to the outcome of Livermore's effort.

A number of factors made the work challenging. The facility had been abandoned for nearly two years, during which time Michigan's cold winters, combined with deferred maintenance of the heating system, had caused some pipes to freeze and burst. Flooding in areas where chemicals were kept spread contamination and dissolved labels on containers. In addition, some tritium spread inside the building as a result of fighting a fire in a copier room. Tritium also contaminated some asbestos, itself a hazardous material.

Teams Go to Work

To execute the project, the Laboratory formed a team consisting of hazardous waste management experts,

Waste Certification

Laboratory tritium experts turned for help to another small cadre of experts from LLNL's Waste Certification program. These individuals quickly established a process at KMS to ensure that low-level radioactive and mixed (radioactive and hazardous) waste from the closed facility met the stringent acceptance criteria for disposal at both DOE's Nevada Test Site (NTS) and Hanford, Washington, complex.

Two Waste Certification program members, manager Bob Fischer and waste certification engineer John Shingleton, spent several weeks coordinating the waste certification activities in Michigan, while others supported the effort from Livermore. One of the most important tasks at Livermore was developing a waste sampling and analysis plan, which was done by waste certification engineer Blanca Haendler.

Actual sampling was conducted by a Livermore Hazardous Waste Management team. Once the sampling results were back from an outside testing laboratory, Haendler reviewed all the data, which showed that the liquids were primarily mixed waste. As a result, LLNL people arranged for storage at the Hanford site because it is the designated storage/disposal site for DOE mixed waste from non-defense-related programs such as that from KMS.

At KMS, Fischer and Shingleton conducted classroom and hands-on training to ensure that workers understood the requirements imposed by the two waste storage/disposal sites. The training was patterned after existing LLNL training courses and modified for the facility in light of State of Michigan regulations.

Figure 2. At the Ann Arbor Initial Confinement Fusion Facility, waste was packaged for transport after certification by the Livermore Waste Certification staff.

Another important task at KMS was to establish a computer database, modeled after those used at LLNL, to characterize the processes involved in the generation of KMS waste streams. To that end, three former KMS employees were retained to help LLNL specialists conduct a comprehensive room-by-room evaluation of former processes and to identify the contents of 10 stored waste drums. The object was to characterize both the legacy waste (in the drums generated by KMS during its operations) and the process waste that would be generated from the decontamination and decommissioning activities.

All 10 legacy waste drums were sorted and repackaged item by item (Figures 2 and 3), with a few materials removed for special treatment. Then Waste Certification people worked with another Hazardous Waste Management team to package the waste and transport it (with accompanying documentation) to Hanford and NTS.

Fischer notes that waste certification work at Ann Arbor required expertise in container procurement, calibration, tritium monitoring systems, document control, training, nonconformance reporting, waste tracking, surveillance, shipping, certification, radiation detectors, health physics, industrial hygiene, and transportation. Indeed, detailed knowledge in all these areas is needed for everyday tasks that the group performs.

Certifying Waste Day to Day

At Livermore, the Waste Certification program ensures that LLNL manages its radioactive waste to meet the requirements of the designated waste disposal facilities, in particular, low-level radioactive waste destined for disposal at NTS. Some of the work focuses on sampling and analysis of waste streams such as liquid decontamination wastes, gravels, contaminated soil, and high-efficiency particulate air filters.

Other waste streams—such as contaminated laboratory trash, contaminated equipment, and empty containers—are characterized using process knowledge. In evaluating these waste streams, the program relies on detailed questionnaires. Cognizant managers must carefully scrutinize all aspects of waste generation before starting an experiment or procedure that will generate radioactive waste. Then they fill out the forms—and proceed with the task—accordingly.

For example, the process knowledge form asks about specific procedures regarding the use of materials and generation of waste containing gases, radionuclides, and hazardous, toxic, or corrosive substances. The form also specifies special treatment for safety hazards such as any free liquids, fine particles, or compressed gas that might be generated as wastes. Liquids, for example, must be solidified to a "peanut butter" type of consistency before shipment to NTS. In addition, radioactive gases and compressed gases (e.g., aerosol cans) must be depressurized or absorbed. The reasoning behind these restrictions is that NTS employees and the environment might be endangered by contaminated liquids or hazardous fine particles freed from a ruptured waste drum. Such safeguards are also important to protect public health during transport.

LLNL currently has 18 low-level radioactive waste streams certified for disposal at NTS, with an additional 5 waste streams conditionally approved. Low-level waste is the most abundant radioactive waste type generated at LLNL. Because of the multiprogram nature of the Laboratory, a wide variety of radionuclides are contained in the waste matrices. Waste certification engineer Kem Hainebach notes that although LLNL has no high-level waste (e.g., from spent nuclear fuel), there is some transuranic waste (e.g., plutonium-contaminated waste from the LLNL Plutonium Facility), defined as wastes containing long-lived radionuclides heavier than uranium, with half-lives greater than 20 years and in concentrations greater than 100 nanocuries per gram of waste. These radionuclides decay primarily by alpha-particle emission. The program is developing the necessary documents and characterization systems to allow shipment of Livermore transuranic waste to the DOE's Waste Isolation Pilot Plant in New Mexico, which is expected to open in 1998.

Figure 3. Livermore's Bob Fischer and Rod Hollister finish packaging and sealing Ann Arbor waste after it is certified for transport.

Using today's standards and procedures, LLNL ensures that there are no prohibited articles—such as batteries, free liquids, low-level mixed waste, or pressurized aerosol cans—in waste ready for disposal. For example, real-time radiography reveals the contents of drums of "legacy waste," that is, low-level, transuranic, and mixed waste that was not generated under a certification and characterization plan. Most of this legacy waste was generated during the 1980s when documentation throughout the DOE complex was inadequate compared to today's standards. The program also takes representative samples from certain waste streams and sends them to an accredited analytical laboratory. Careful review of the analytical results is required before the waste can be certified. Such efforts are very similar to that required at the former KMS facility in Ann Arbor.

Another important aspect of certification work is verifying generator-supplied waste records against the process knowledge evaluation forms. This important quality-control step provides the necessary assurances to low-level waste disposal sites that the waste meets the waste acceptance criteria. After the material is also verified, it is packaged into authorized waste containers for shipment offsite.

The Future: Ecological and Efficient

Waste Certification people are working with LLNL technical managers to plan the best ways to design experiments to reduce the hazards of waste streams—that is, to test and recycle or release noncontaminated materials and to prevent radiological contamination from spreading to noncontaminated materials. For example, program manager Fischer is helping to design waste management programs for the future National Ignition Facility. The old industry way, he notes, was to "try to figure out what to do with waste after you produce it."

Fischer says that in light of the successes at Ann Arbor, the future may also bring more off-site projects. "I think we've shown that our program can be readily deployed to other sites for substantial time and cost savings," he says.

Indeed, the program is already helping a company in Golden, Colorado, to characterize its wastes, and a much larger project has been proposed to DOE in which Livermore experts would train Bechtel employees at NTS to prepare transuranic waste drums, now in interim storage there, for disposal at the Waste Isolation Pilot Plant in New Mexico.

Figure 4. This "before" photo shows a typical lab in the Ann Arbor 9,000-m^2 facility.

a health physicist, industrial hygienists, hazards control technicians, and former KMS Fusion employees who were familiar with the building's past experimental processes. The team also set up a contract group to do many of the everyday services that are taken for granted at an operating facility—security, phone service, garbage pickup, and janitorial service.

At any one time, 15 to 25 Laboratory and DOE people worked at the facility, some of whom stayed there as long as six months. Livermore's Tom Reitz, the project leader responsible for planning and managing the Ann Arbor field operation, notes that it was not easy duty. During the winter, outside temperatures dropped below zero, and the heating system failure dropped inside temperatures to near freezing even after they purchased a large number of electric heaters to keep pipes from freezing again.

The effort required a close working relationship with the DOE and many other agencies. The State of Michigan had to approve all plans. To handle and dispose of low-level radioactive waste and mixed (radioactive and chemically hazardous) waste, the Laboratory had to obtain approvals from DOE's Nevada Operations Office. To ensure compliance with all applicable requirements, DOE–Nevada and the State of Michigan frequently audited the Ann Arbor work.

The major goals of the cleanup effort were to identify and remove the tritium (present mostly as uranium hydride beds in the processing equipment), analyze and dispose of thousands of containers of chemicals (some of which were also radioactive), decontaminate and dispose of the experimental and process equipment, decontaminate the building itself, and, if necessary, remove any contamination found outside the building. See Figure 4, which shows the "before" condition of one area inside the building.

To remove the tritium-containing uranium hydride beds, the team had to restart the old process equipment—but only after assuring it could be done safely. To do that, Livermore scientists wrote operating instructions, performed dry runs, and made some minor modifications to the equipment. The retrieved uranium hydride was put in approved shipping containers and sent to Mound Laboratories for tritium recovery.

To deal with the chemicals, the team consolidated chemical containers by chemical type and identified and characterized the contents of each. They contracted out the analysis and disposition of the chemicals to a local state-licensed laboratory, which was paid for the work mostly through the value of the uncontaminated chemicals

that were recovered. The team established a waste accumulation area for handling and directing the radioactive chemicals to the proper waste receiver.

Part of the challenge of dealing with hazardous materials involved what project workers called "Easter eggs"—sealed vessels with unknown contents. In one case, the team x-rayed a welded container, which revealed that within the vessel was molecular sieve material, a special type of absorbant that had been used to trap tritium. It was disposed of as low-level waste.

The team assumed that the equipment in the building was contaminated until test results showed otherwise. They performed radiological surveys on all the facility's equipment, mostly by wiping the surface (called swiping) and reading the swipes with a scintillation counter. The team bar-coded each item with a unique identifier and set up a database to track all the samples, swipes, data, and equipment. Clean equipment and contaminated equipment that was able to be cleaned were returned to the DOE, to the General Services Administration, or was sent to government surplus. Low-level waste was sent to the Nevada Test Site for disposal.

Once the team stripped the building of all equipment, they checked the entire facility—i.e., walls, floors, ceilings, ductwork, drains—for residual contamination. Again, they did this mostly by swiping, but they also analyzed bulk building materials such as concrete and sheet rock for radioactivity. Fortunately, most contamination was limited to a surface layer; but for areas too deeply contaminated to clean, the team had to completely gut the main tritium area by removing stud walls and ducting. Here, too, Reitz recalled difficulty in removing contaminated blowers from the slippery, snow-covered roof without compromising the equipment's plastic-bag wrappings to prevent any contamination from spreading.

The exterior of the facility was surveyed in a similar way. This work was done under a separate DOE–Oakland contract by Energy Technology and Engineering Center (ETEC), which also contributed supporting staff for the facility's decontamination and decommissioning activities. ETEC tested walls, doors, and roofs and collected hundreds of samples of the 5 hectares (12 acres) of surrounding grounds. Fortunately, they found no radiological or chemical contamination above background levels.

Challenges Met

At the end of the project, the DOE returned the cleaned building to its commercial owner for unrestricted use. In a fitting conclusion to the effort, the last project people to leave the facility were two former KMS Fusion workers who had spent years working there.

Phillip E. Hill, then-leader of DOE–Oakland's Closeout, Decontamination, and Decommissioning project, summarized, "the project was accomplished efficiently and effectively as a result of DOE and LLNL working together to return the facility to the owner for unrestricted use. With an experienced team headed by Mark Mintz and Tom Reitz, LLNL successfully achieved the Department's goal of returning the facility to the owner while minimizing the cost to DOE."

Key Words: cleanup, decontamination, decommission, disposal, low-level waste, tritium, waste certification.

For further information about the tritium removal project, contact Mark Mintz (510) 422-8394 (mintz1@llnl.gov).
For further information about waste certification, contact Robert P. Fischer (510) 422-3004 (fischer7@llnl.gov).

About the Scientists

ROBERT P. FISCHER joined the Laboratory's Environmental Operations Group in 1988 after working in the hazardous waste industry since graduation from college. He attended San Jose State University, where he received a B.A. in chemistry and a B.S. in environmental studies in 1986. Currently manager of the Waste Certification program in the Environmental Protection Department, Fischer is also chairperson of the department's Nevada Test Site Working Group.

MARK MINTZ came to the Laboratory in 1992 and joined the Tritium Operations Group in Defense and Nuclear Technologies Directorate. Currently the Tritium Facility Manager, Mintz has written many articles on tritium handling and systems design and articles about materials science. He received a B.S. in physics from the University of North Carolina, Chapel Hill, in 1972, an M.S. in nuclear engineering from North Carolina State University, Raleigh, in 1975, and a Ph.D. in materials science from the University of California, Davis, in 1986. Prior to working at LLNL, Mintz worked for Sandia National Laboratories, Livermore, and General Atomics.

Solving the Mammoth Mountain CO_2 Mystery

IN 1990 a forest ranger was almost asphyxiated when he entered a floorless, snow-covered cabin near Mammoth Mountain on the eastern side of the Sierra Nevada. At about the same time, trees began to die in four patches, which over the next several years expanded to cover 30 to 35 hectares (76 to 86 acres). (See figure next page.) At first no one thought to connect the cabin incident with the dead trees. But as scientists riddled out the case of the dying trees, the near-asphyxiation of the forest ranger provided a critical clue.

Researchers from the U.S. Geological Survey (USGS) were the first to study the problem in 1994.[1] U.S. Forest Service biologists helped rule out drought and insect infestation as possible causes for the dead trees. The ranger's asphyxia symptoms prompted the USGS to look at carbon dioxide (CO_2) levels in the soil because high CO_2 concentrations are harmful to plants and animals. The USGS also brought experts from Lawrence Livermore and Lawrence Berkeley National Laboratories to analyze soil gas components.

The USGS took about 100 soil gas samples from various areas around Mammoth Mountain—in the patches of dead and dying trees, at the cabin where the ranger had been so short of breath, near the fumaroles (volcanic gas vent), and in areas of healthy trees. Carbon dioxide concentrations analyzed by a portable gas chromatograph ranged from less than 1% in healthy forest, a typical figure for forest soils, to more than 90% at several locations within tree-kill areas. Where CO_2 concentrations exceeded 30%, most trees were dead. Other lethal agents were not apparent, and the soils showed no sign of elevated temperatures. The USGS also estimated that the soils in the tree-kill areas were releasing as much as 40 metric tons of CO_2 per hectare per day, which compares with typical CO_2 releases of 10 to 20 kilograms per hectare per day from normal forest soil.

Carbon dioxide was clearly a problem, but where was it coming from? A possible source was Mammoth Mountain itself, which last erupted about 500 years ago. More recently, a series of magnitude 6 earthquakes in 1980 was followed by swarms of temblors in 1983, 1989, and this year. Many volcanoes release large quantities of CO_2, but they do so at the summit and during periods of low-level eruptive activity. Mammoth, on the other hand, shows no signs of erupting.

A less likely source for the CO_2 releases was the soil. The soil of a healthy forest is enriched in CO_2 because of the

The tree-kill area near Horseshoe Lake (currently closed to campers). Other volcanoes in the world, such as Mt. Etna, release CO_2 when they are not erupting, but Mammoth is unique in the tree-kill associated with the releases.

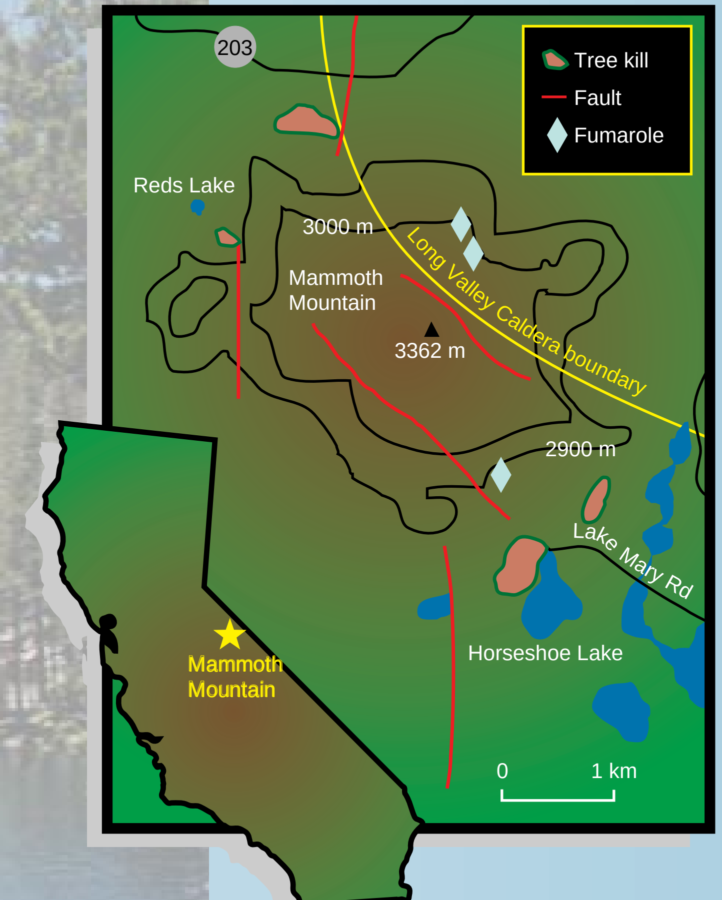
biological process of decomposition, which uses up oxygen and converts it to CO_2. But normal CO_2 enrichment is minor compared to the quantities found at Mammoth. Analysis of soil gas samples from areas of healthy forest indicated normal levels of biogenic CO_2. But in tree-kill areas and near the fumaroles, biogenic CO_2 made up only a tiny fraction of the total. So the most likely source for these anomalous CO_2 levels was indeed the mountain and volcanic activity deep inside.

At this point, the Livermore and Berkeley laboratories provided their expertise in gas analyses. Berkeley's analysis of carbon-13 and other gases in samples from tree-kill areas indicated "signatures" that were typical of magmatic CO_2, signatures that were remarkably similar to those found at the fumarole where CO_2 would be expected to be of magmatic origin.

Carbon-14 Clincher

Livermore's analyses of carbon-14 (^{14}C), at its Center for Accelerator Mass Spectrometry, provided the clincher in determining the source of the CO_2. Mass spectrometry (MS) is a technique used to determine the mass of an atomic species or a molecular compound. Accelerator mass spectrometry (AMS), as it is applied at Livermore, adds three steps to MS. After the initial acceleration to kilovolt energies and the separation of the ion beam by mass, a second acceleration of millions of volts is applied. Then the ion beam is stripped to a charge state where at least three electrons are removed from the atoms of interest, which destroys all molecular species. Finally, the isotope has its mass, energy, velocity, and charge redundantly determined, which removes background interference. The resulting sensitivity is typically six orders of magnitude greater than that of conventional MS. AMS can find one atom of ^{14}C in a trillion other carbon atoms.

In soil gas samples taken from healthy forest 1,500 meters from the nearest tree-kill area, Livermore scientists found that ^{14}C levels were in keeping with those typically associated with



Just east of Yosemite National Park, California, Mammoth Mountain is at the southwestern edge of the 750,000-year-old Long Valley Caldera and at the southern end of the Inyo Craters volcanic chain. All four tree-kill areas are near faults on the flanks of Mammoth Mountain.

biogenic CO_2. Healthy forest soils have high levels of ^{14}C because they are young. Volcanic magma, on the other hand, has been underground for millions of years and has no ^{14}C.

The constant, predictable decay of ^{14}C is what makes it an effective dating tool. Carbon-14 is a natural, radioactive carbon that is continuously produced in the upper atmosphere by cosmic-ray interactions. It is present in all green plants, which absorb it from the atmosphere. Through the food chain, all organisms ingest ^{14}C over the course of their lives. Once an organism dies, it ceases to take in ^{14}C, so the amount of ^{14}C in its tissues steadily decreases. By measuring residual ^{14}C with AMS, materials from 500 to 50,000 years old can be dated with remarkable precision.

An analysis of soil gas samples from the tree-kill areas showed extremely low ^{14}C levels. In areas of apparently healthy forest, over 100 meters from the nearest dead or dying trees, ^{14}C levels were only slightly higher. Carbon-14-free, magmatic CO_2 was apparently diluting the ^{14}C in the soil.

Scientists now knew the source of the CO_2. But they needed to verify that the CO_2 had made its way from the soil into the trees and that it was in fact CO_2 killing the trees. While some increase in CO_2 in the atmosphere is beneficial for trees, too much CO_2 in the soil is not. Livermore researchers analyzed pine needles for CO_2 content, and their data showed that the percent of magmatic carbon in needles from healthy forest was zero, in stressed trees it ranged from 2 to 6%, and in dead trees from two different areas it ranged from 2 to 65%. Generally, the more magmatic CO_2 a tree had absorbed, the less healthy the tree appeared. Scientists believe that the CO_2 inhibits the growth of tiny rootlets that normally absorb water and other nutrients from the soil; in other words, the CO_2 is asphyxiating the trees.

This dilution of ^{14}C has produced some startling apparent "ages." Analysis of ^{14}C in a needle from a tree dead only a year showed an age of 7,200 years. The outer, most recent growth ring of a tree dead just a few years showed an apparent age of over 4,000 years, in contrast to its other recent rings, which showed modern ages.

Analysis of four tree cores indicates that in 1990 their ^{14}C levels began to drop relative to modern ^{14}C in the atmosphere, which is when dead trees were first noticed. By absorbing elements that can be "read" in their growth rings, trees are a unique recorder of historic activity.

Ongoing CO_2 and ^{14}C Work

Scientists are using this experience at Mammoth to study historic activity at other volcanoes. Work is just beginning on research at Mt. Lassen, which last erupted from 1914 to 1916. Growth rings will be studied for anomalous CO_2 and ^{14}C levels to determine whether a correlation exists between CO_2 levels and the eruption. Livermore researchers also hope to perform comparable studies at Mt. St. Helens and other modern volcanoes. Growth ring analysis of historic activity could prove to have enormous benefits for modern vulcanologists and others attempting to forecast volcanic eruptions.

Key Words: accelerator mass spectrometry, carbon-14, magmatic CO_2, volcanic activity.

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A Closer Look at Osteoporosis

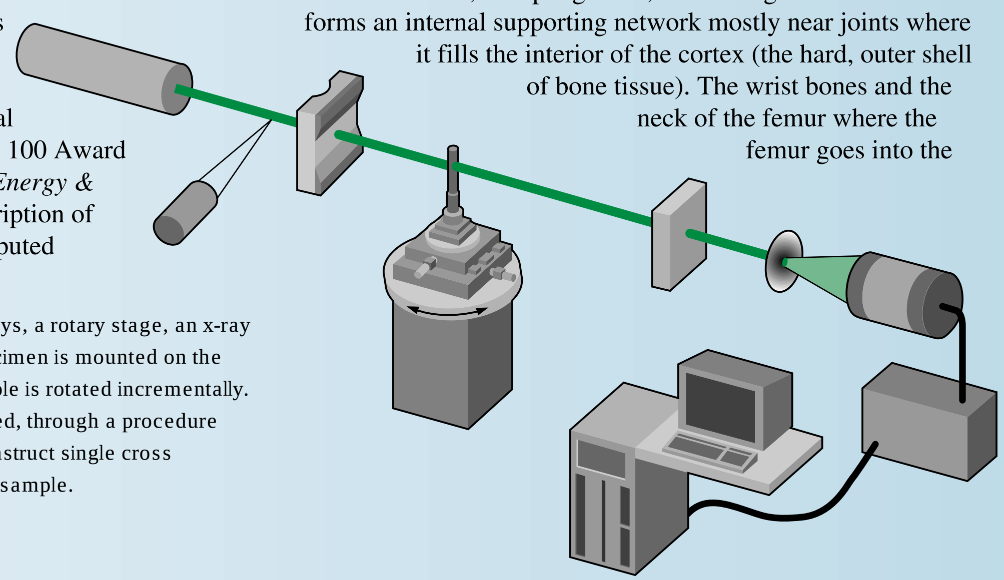
HALF of all women born in this country will suffer a bone fracture because of osteoporosis. In osteoporosis, the bones become so fragile that they can break almost spontaneously. It is also true that more women die each year as a consequence of osteoporotic fracture than die of breast cancer. With numbers like these, the need to find a cure for osteoporosis is an urgent one.

Scientists at Lawrence Livermore National Laboratory are actively involved in this cause using the x-ray tomographic microscope (XTM) to produce three-dimensional images of bone. We are using these images to detect microscopic changes in bone structure of small laboratory animals and to study bone loss as well as increases in bone volume after treatment.

The only other method for producing accurate images of the microstructure of bone is sectioning, a time-consuming process that requires slicing the bone very thinly. This method destroys the sample and often introduces tiny pieces of debris, called artifacts, that can obscure important information. Furthermore, sectioning only produces two-dimensional images, which can be used to depict three-dimensional bone structure but not always with complete accuracy. XTM is the only method currently available for studying bone three-dimensionally without destroying it. This means that studies can even be made *in vivo*.

The XTM at Work

The XTM was developed in 1991 as a spin-off of work on x-ray lasers for the Strategic Defense Initiative, and its inventors at LLNL and Sandia National Laboratories, Livermore, won an R&D 100 Award for the efforts. (See the October 1991 *Energy & Technology Review* for a detailed description of the XTM.) The XTM is a form of computed tomography, or CT, which was developed in the 1970s as a medical diagnostic tool. (The commonly used term "CAT scan" is a vestige of the earlier name "computerized axial tomography.") The LLNL configuration of the XTM is shown below.



The XTM consists of a source of parallel x rays, a rotary stage, an x-ray detector, and an analyzing computer. A specimen is mounted on the stage, and images are collected as the sample is rotated incrementally. These images are computationally assembled, through a procedure called Fourier-filtered back-projection, to construct single cross sections or three-dimensional images of the sample.

The XTM's spatial resolution is about 2 micrometers, shown at right. Using monochromatic (single-energy) synchrotron radiation at Stanford University's Synchrotron Radiation Laboratory (a part of the Stanford Linear Accelerator), the XTM can obtain spatial resolutions better than that of the best medical CT scanners. Monochromatic synchrotron radiation is used rather than conventional x rays; the former produces less distortion and, hence, better resolution because of its high brightness and the nearly parallel quality of its beam, known as collimation. The XTM is also superior to magnetic resonance imaging (MRI) because MRI cannot be used on metallic materials and because the resolution of the XTM is many times greater.

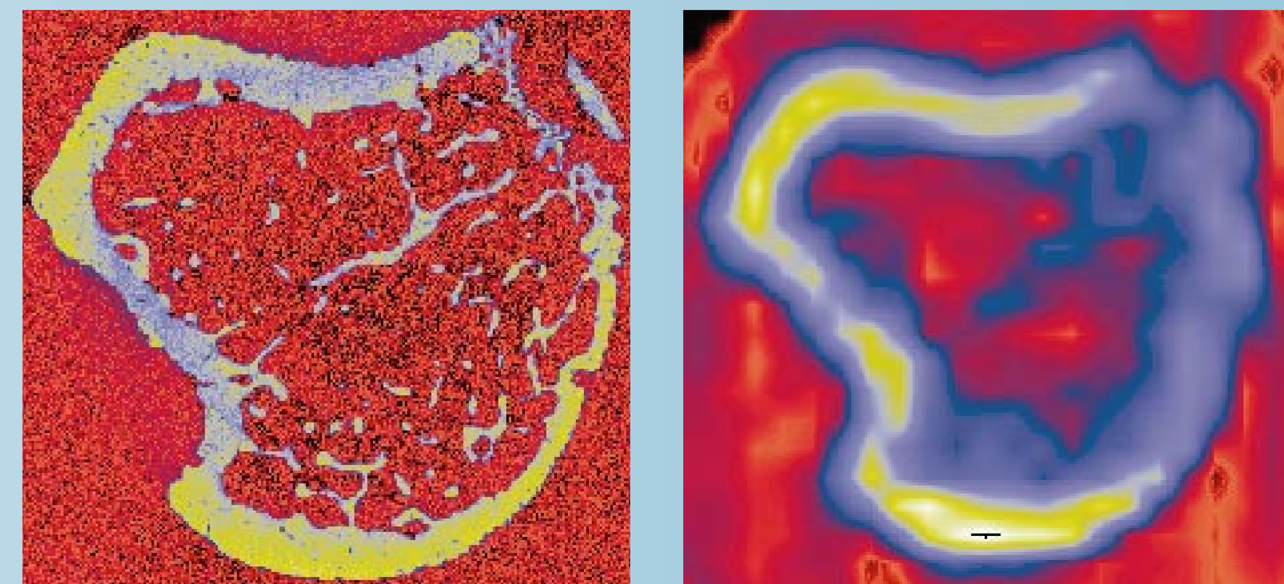
The XTM is excellent for nondestructive evaluation of a wide variety of industrial and military materials, but the radiation dose required to produce the XTM's high-resolution images currently limits its use in medical studies to laboratory animals or cadavers. Work continues to reduce the radiation exposure levels.

Searching for a Cure

Researchers from the Laboratory and the University of California, San Francisco, are studying osteoporosis, looking at bone loss due to estrogen depletion and at potential treatments. The hope is to understand critical clinical time points in the development of osteoporosis to establish more effective interventions.

As with many studies of osteoporosis, our studies focus on trabecular bone, the sponge-like, connecting bone tissue that forms an internal supporting network mostly near joints where it fills the interior of the cortex (the hard, outer shell of bone tissue). The wrist bones and the neck of the femur where the femur goes into the hip joint have considerable trabecular bone; the vertebrae are almost entirely trabecular bone with very little cortex. Most osteoporotic fractures occur at these three sites.

(a) (b)



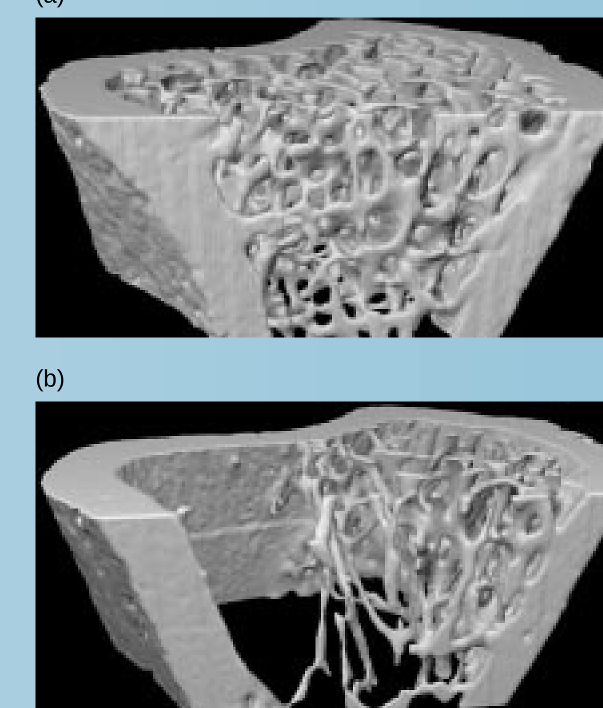
These figures compare (a) a two-dimensional XTM image of a rat's bone structure with (b) an image from a pQCT (Peripheral Quantitative CT) scanner, which is the highest resolution CT scanner commercially available for imaging biological structures.

Female laboratory rats are being used as subjects, half of which have had their ovaries removed to induce estrogen depletion. The non-ovariectomized rats serve as controls. Rats are excellent subjects for osteoporosis studies because estrogen depletion affects the bones of rats and humans in similar ways but much more quickly in rats than in humans.

In the first study, we took XTM images of the rats' proximal tibias before their ovaries were removed, and again five weeks later to determine bone loss. (See images at right.) Trabecular bone volume decreased by approximately 60% in the estrogen-depleted animals compared to the control group. In addition, there was a significant change from an interconnected plate- and strut-like structure to one that was mostly disconnected struts. Dangling trabecular elements, supported only by marrow, were also seen in the ovariectomized animals. While these dangling elements contribute to total bone mass, they do not contribute to the stiffness or strength of the bone. We found that the number of trabecular interconnections decreased by 90% in the rats without ovaries compared to the control group. Combinations of broken trabecular struts and dangling elements most likely contribute to fracture risk.

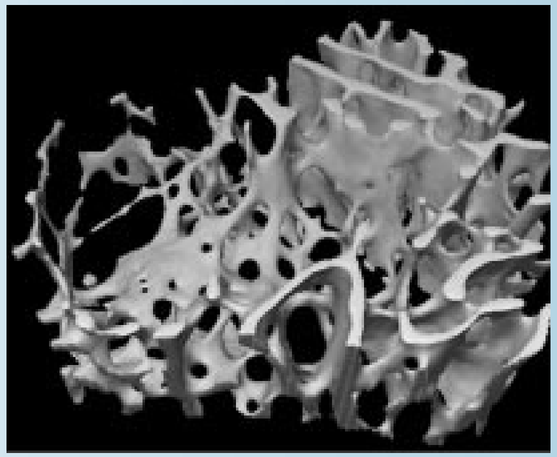
(a)

(b)

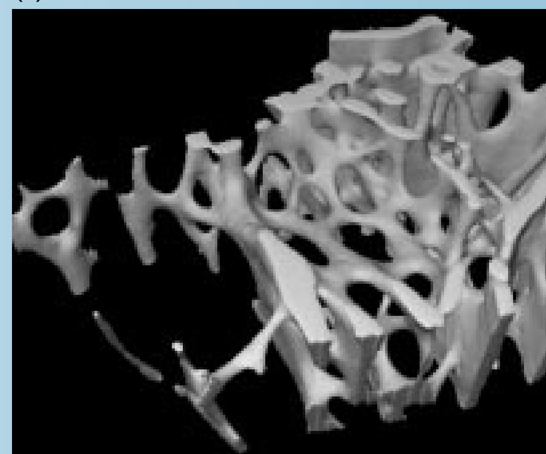


Three-dimensional composites of a rat's proximal tibia (a) just prior to ovariectomy and (b) 5 weeks after the ovariectomy, by which time estrogen depletion has caused osteoporosis.

(a)



(b)



Representative XTM images (a) before ovariectomy and (b) 12 weeks after treatment with human parathyroid hormone (hPTH). The daily dose of hPTH was 400 micrograms per kilogram of body weight. Bone volume has been re-established, but trabecular interconnections have not.

In our most recent study of a potential treatment for osteoporosis, ovariectomized rats were given various intermittent doses of human parathyroid hormone (hPTH) because it appears to be involved in the differentiation and regulation of bone morphogenic proteins. Scientists do not fully understand how these proteins work, but somehow they control the cells that make and resorb bone. Treatment with hPTH began 56 days after the rats' ovaries were removed and continued for four weeks. We found that hPTH did increase trabecular bone volume and trabecular thickness to baseline levels or higher, although it did not re-establish the bone's original structure by recreating lost trabecular interconnections. (See images above.) This and other studies suggest that hPTH's beneficial effects on bone mass do not depend upon the presence of functioning ovaries, which is very good news for post-menopausal women. The failure of hPTH to re-establish trabecular interconnections after 50% of them had been lost may mean either that earlier intervention or prolonged treatment, or both, are required.

Other Work with the XTM

Laboratory scientists also are working with Roche Biosciences of Switzerland to study bone loss caused by continuous use of steroidal anti-inflammatories such as prednisone. Preliminary work has demonstrated that the bone loss caused by medications such as prednisone is very different from estrogen-induced bone loss. Roche has developed a new compound that they believe prevents this bone loss.

We have also used the XTM to study periodontal disease and coronary artery disease. In the future, the XTM may be used to study fracture healing, kidney stone disease, autoimmune diseases such as arthritis, or any other calcified tissues. The key to all of this work is our ability to noninvasively examine body anatomy three dimensionally. With the XTM, we can evaluate therapies and conditions that affect many common but difficult-to-solve health problems. X-ray tomographic microscopy is significantly advancing our understanding of several very important public health issues.

Key Words: computed tomography, human parathyroid hormone, osteoporosis, steroidal anti-inflammatories, x-ray tomographic microscopy (XTM).

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Abstracts

Theory and Modeling in Materials Science

A survey of four research projects shows how theory and modeling efforts by scientists in the Chemistry and Materials Science Directorate at LLNL are advancing our understanding of the property of materials with consideration of underlying structures. To account for radiation effects in some materials, we have created a hierarchy of simulation tools. Focusing on damage processes that occur when semiconductor devices are manufactured, we can now predict the distribution and growth of defects in silicon when dopant ions are implanted by a high-energy ion beam. Tantalum, a ductile metal with important defense applications, is the subject of another modeling project. Our recent model of deformation in tantalum uniquely accounts for its work-hardening behavior, and the same approach can potentially be applied to other types of commercially useful metals. In the area of energetic materials, we are simulating how a shock wave propagates through high explosives as a function of degradation, and we can predict how new explosives will perform under a variety of conditions. Finally, we have developed models that accurately mimic the complicated network and void structure of ultralow-density aerogels. Such models help us understand how molecules flow through aerogels and can facilitate the future use of these unconventional solids in applications that take advantage of their enormous surface area.

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LLNL and DOE Collaborate on Successful Fusion Facility Cleanup

Livermore and DOE's Oakland Operations Office teamed up to decontaminate, decommission, and close out—on time and under budget—the Ann Arbor Inertial Confinement Fusion Facility in Michigan. To execute the project, the Laboratory formed a team of hazardous waste management experts, a health physicist, industrial hygienists, hazards control technicians, and former KMS Fusion employees who were familiar with the building's past experimental processes.The major goals of the cleanup effort were to identify and remove the tritium; analyze and dispose of thousands of containers of chemicals (some radioactive); decontaminate and dispose of equipment; decontaminate the building; remove any other contaminated items; and return the cleaned building to its commercial owner for unrestricted use. They developed a waste sampling and analysis plan; characterized legacy waste (in drums generated during the facility's operation) and process waste generated from this project's activities; and after certification, packaged the waste for storage at the Nevada Test Site and DOE's Hanford, Washington, complex.

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