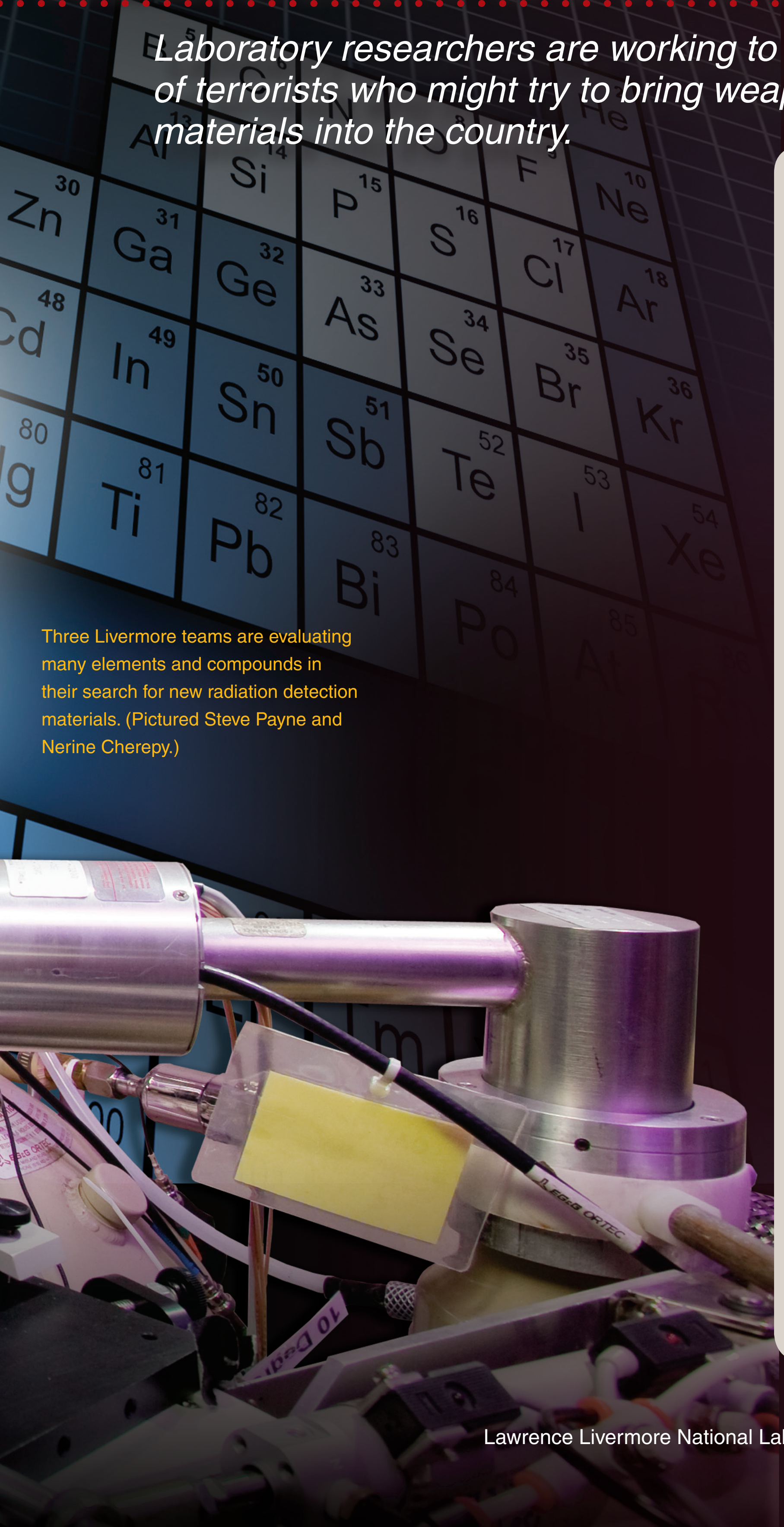
The Hunt for Better

Radiation Detection

Laboratory researchers are working to stay ahead of terrorists who might try to bring weapons-grade materials into the country.



Three Livermore teams are evaluating many elements and compounds in their search for new radiation detection materials. (Pictured Steve Payne and Nerine Cherepy.)

DETECTING illicit sources of plutonium and highly enriched uranium is a tricky business for first responders, airport security personnel, and U.S. port and border inspectors. Both plutonium and highly enriched uranium are typically identified using a combination of devices, working together, to detect their invisible gamma and neutron radiation emissions. For many years, the existing detection technologies were deemed adequate, but the world became a far more dangerous place after September 11, 2001. Since then, concerns of radioactive materials falling into the wrong hands have been rife. Ensuring that the country remains safe from a nuclear or radiological attack is driving the search for more definitive radiation detection and identification technologies.

The Department of Energy (DOE) has for decades been building the science and engineering basis for the detection of radiological materials, and the Laboratory has made a significant contribution to this ongoing effort. Livermore researchers have worked on improving the efficiency and energy resolution of radiation detectors. They have also developed systems that illuminate objects such as cargo containers and use these radiographic techniques to find hidden nuclear materials. (See *S&TR*, May 2004, pp. 12–15, and the highlight beginning on p. 17.) In 2005, the Department of Homeland Security (DHS) made a bold request for developing significantly more effective materials to detect gamma and neutron radiation emissions. New materials for smaller, faster, and more accurate sensors could improve the nation's ability to unambiguously identify radiation from illicit sources.

The development of new detector materials used to be a time-consuming, trial-and-error, often decades-long process. Today, greatly improved diagnostic tools for measuring material properties combined with faster, more accurate computer simulations allow researchers to rapidly evaluate materials for performance and ease of fabrication.

Physicist Steve Payne leads three teams in a radiation detector materials campaign at Livermore, in a collaboration that includes Pacific Northwest, Lawrence Berkeley, Oak Ridge, Brookhaven, and Sandia national laboratories; Fisk, Stanford, and Washington State universities; the University of Nebraska; and numerous private firms including, Radiation Monitoring Devices, Inc. "With so many partners working together, we can avoid duplication of facilities, which is more cost-effective," says Payne.

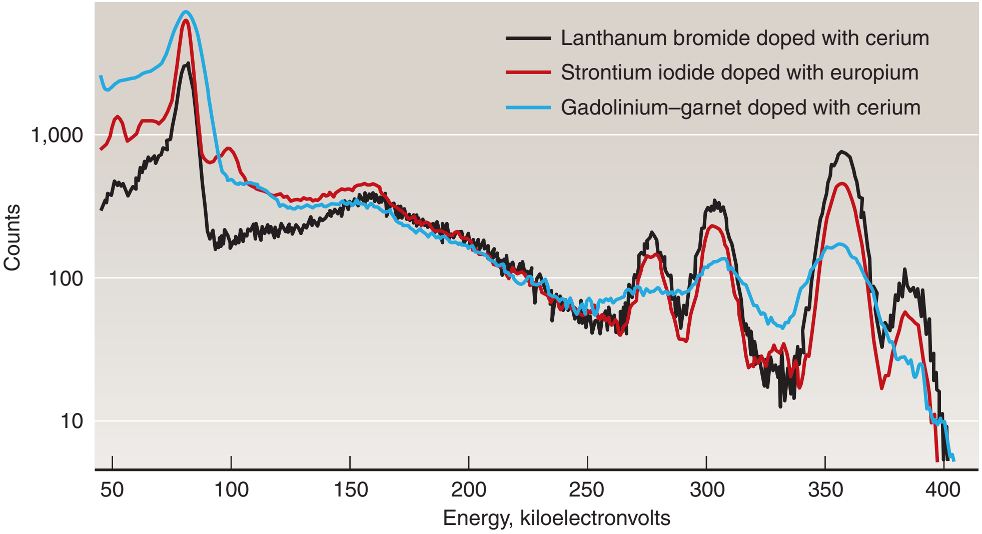
Getting It Right the First Time

For many field applications, radiation detectors must be inexpensive and robust, operate at ambient temperature, provide high efficiency, and be small enough for use in covert operations. They must also provide unambiguous identification of a material. For example, the tiny amount of thorium in cat litter is radioactive, but thorium is not a security threat. Additionally, detectors must be able to pick up very weak signals, such as from plutonium heavily shielded with lead to avoid detection.

Current detector technology is limited in its ability to meet the requirements of people on the front lines who are responsible for ensuring the nation's safety. Some of today's best gamma-ray detectors are those that use germanium as the sensor material. The Laboratory has been at the forefront of efforts to make germanium detectors as small and field-portable as possible, and a challenge has been that to achieve the best resolution, the germanium must be cooled to below room temperature. (See *S&TR*, October/November 2009, pp. 8–9.) Detectors for low-energy neutrons, known as thermal neutrons, are typically tubes filled with helium gas, but these instruments are large, require high voltage to operate, and are sensitive to vibration. The best material for detecting high-energy, or fast, neutrons is a crystal called stilbene, but the crystal is difficult to grow, expensive, and available from just one company in Ukraine.

The Livermore teams of chemists, physicists, and engineers, along with their collaborators, set out to find new, better materials for all three types of detectors—materials that meet the needs of as many users as possible. Payne says, "In our search for novel materials with the necessary properties, we began with the entire periodic table." Each team used a different process of elimination because the requirements for detecting the various forms of radiation are different.

Earlier work at Livermore on new detector materials for thermal neutrons was funded by the Laboratory Directed Research and Development Program. As that project and others demonstrated success, outside funding for new detector materials followed, first from DOE's National Nuclear Security Administration (NNSA) and later from DHS and the Defense Threat Reduction Agency.



Gamma-ray spectra from a barium-133 radioactive source were acquired using three different scintillator materials. In the energy ranges most important for identifying plutonium, the Livermore-developed strontium iodide, with the narrowest pulse widths, offers the best resolution.

Locating Gamma Rays

Gamma rays, produced through radioactive decay, have the highest energy in the electromagnetic spectrum and thus can penetrate most materials. Because of this extreme penetrability, gamma rays can be detected even when the radiation source is shielded by concrete, dirt, or a few centimeters of lead. However, gamma rays can only be viewed indirectly by observing their interactions with detector materials.

High-purity germanium, a semiconductor, has been the standard for detecting gamma rays for years. An alternative method is scintillation, in which radiation interacts with a material to produce a brief but measurable flash of light. Livermore chemist Nerine Cherepy and her team are on the hunt for a material that will produce the brightest flash of light when exposed to plutonium or highly enriched uranium. The precision of the scintillator material's response, or energy resolution, defines the material's ability to distinguish between gamma rays that have similar energies. Such discrimination is needed because not all gamma rays are indicative of a source that poses a threat.

Scintillators of lanthanum bromide doped with cerium, $LaBr_3(Ce)$, offer the highest energy resolution among commercial devices. However, $LaBr_3$(Ce) is difficult to grow, is radioactive because of the presence of lanthanum-138, and is expensive. The goal for Cherepy's team is to find a high-resolution material that is not radioactive, operates at room temperature, is inexpensive, and can be manufactured in large volumes. After much analysis and elimination—with an array of crystals grown from various compounds by national laboratory, industrial, and university partners—Cherepy and her team narrowed their search to two materials: transparent ceramic gadolinium-based garnets and strontium iodide crystals.

Transparent ceramics have already been used in Livermore's solid-state heat-capacity laser, a system that is setting the stage for "directed energy" laser weapons. (See *S&TR*, April 2006, pp. 10–17.) "Livermore has established expertise in transparent ceramics," notes Cherepy. For scintillators, transparent ceramic garnet is relatively inexpensive, strong, and can be fabricated into large, uniform, robust devices. The team has developed a novel fabrication technique for transparent ceramics starting from nanoparticles, for which they earned a Nano 50 Award in 2008.

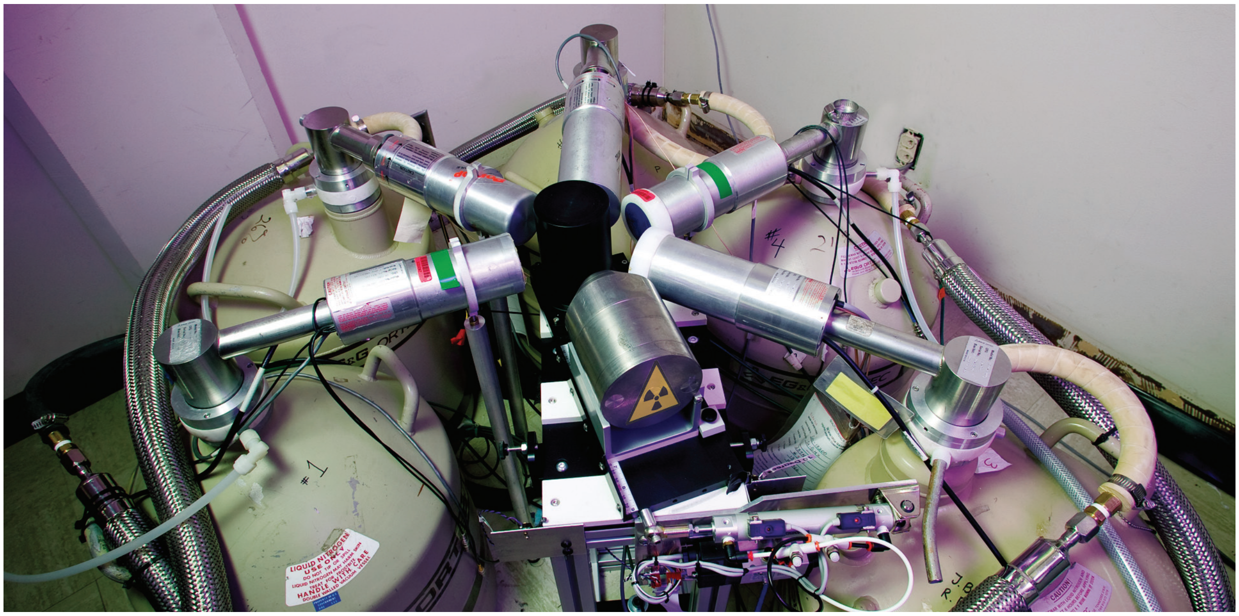
The best energy resolution for any of the various garnet compounds is about 4 percent, when exposed to gamma energy of 662 kiloelectronvolts. (This energy, from a cesium-137 source, is the standard used in experiments for measuring the resolution of a gamma-ray scintillator.) The energy resolution of $LaBr_3$(Ce) is 2.6 percent, and for gamma detection, smaller is better. The team's goal is to find a material with a resolution of 2 percent.

Strontium iodide doped with europium, SrI_2(Eu), has proved to be the best scintillator material yet. "It is an easily grown crystal with excellent energy resolution," says Cherepy. It produces more photons—more light—than $LaBr_3$(Ce) and is not radioactive. Small crystals have demonstrated 2.5-percent energy resolution, slightly better than $LaBr_3$(Ce). "The resolution degrades a bit with larger crystals," says Cherepy. "However, we expect to achieve 2-percent resolution by improving the purity of crystals as they grow in size and by optimizing the detector's optics and digital electronics."

An array of tools and computer codes is used to characterize and analyze the materials being studied. A particularly interesting new tool is the scintillator light-yield nonproportionality instrument, or SLYNCI, which was developed by Payne and collaborators at Lawrence Berkeley. Nonproportionality—an inconsistency between the energy deposited in a scintillator and the number of visible photons produced—has been a problem with scintillators for decades. If too much exists, scintillator pulses are not precise, degrading energy resolution. Doping is one way to improve proportionality. SLYNCI has been crucial in evaluating the physics of nonproportionality in various scintillators.

Identifying High-Energy Neutrons

While working on a Ph.D. at Moscow State University in Russia, Livermore physicist Natalia Zaitseva developed a method for growing extremely large crystals faster than ever before. Zaitseva perfected the process after coming to the Laboratory and won a 1994 R&D 100 Award with her team. At the time, the researchers were focused on producing

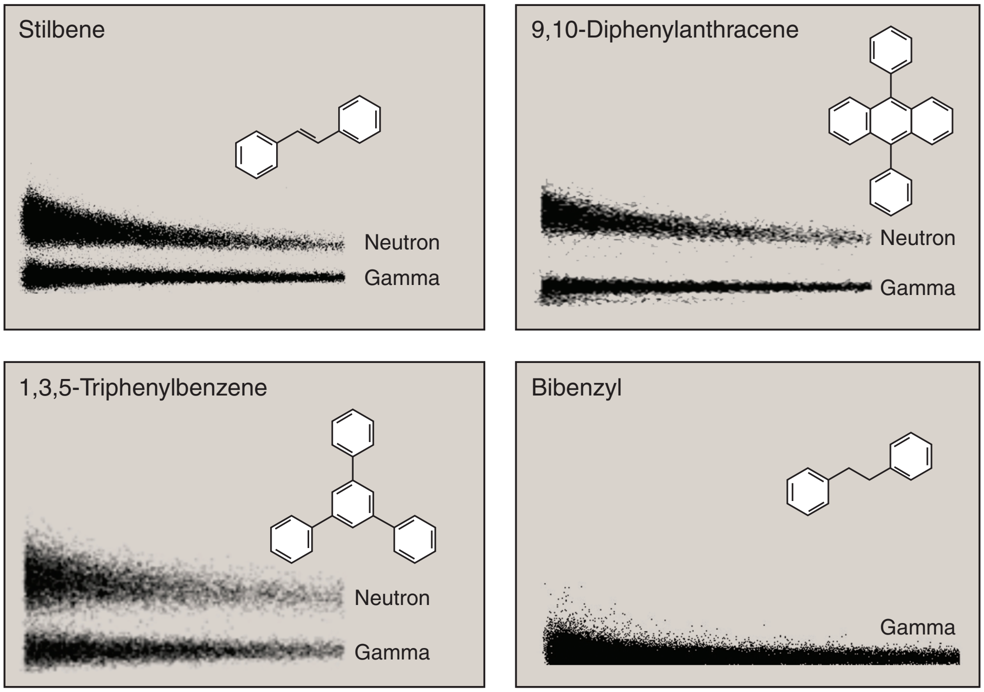


SLYNCI, the scintillator light-yield nonproportionality instrument, measures the difference between the energy deposited in a scintillator and the number of protons produced.

large crystals of high-quality potassium dihydrogen phosphate (KDP) for Livermore lasers such as the National Ignition Facility (NIF). In 1996, the team produced in just 27 days a KDP crystal measuring 44 centimeters across. Growing a crystal that size under standard growing conditions would have taken 15 months. The NIF laser design required a huge amount of KDP—about 600 large slices—for conversion of the laser's infrared light to shorter wavelengths as the beams travel to the target chamber. In 1997, the Livermore team produced the world's largest single-crystal optical element—a pyramid-shaped KDP crystal measuring more than a half-meter tall and weighing about 250 kilograms—in just six weeks. Today, Zaitseva's team is applying everything learned with KDP—an inorganic substance—to organic substances for neutron detection.

In examining possible compounds, Zaitseva's aim was to find a material that would most effectively separate out a signature for neutrons from the strong background of gamma radiation, a process known as pulse-shape discrimination (PSD). Decades ago, researchers found that the organic crystal stilbene could quickly discriminate neutrons from gamma radiation. Today, liquid organic scintillators are more commonly used for PSD because of stilbene's limited availability and high

In the 1990s, Natalia Zaitseva developed a rapid-growth technique for producing very large crystals in record-shattering time. She now leads a team that grows organic crystals for use in fast-neutron detectors.



Gamma rays and fast neutrons are distinguished from one another by pulse-shape discrimination. The signatures differ depending on the compound being used as the scintillator. Although stilbene is the best commercial scintillator available, some compounds, such as 9,10-diphenylanthracene and 1,3,5-triphenylbenzene, show as good as or better discrimination and can be grown more easily. Other compounds, such as bibenzyl, reveal no discrimination.

cost. However, because of flammability, toxicity, and environmental concerns, NNSA and others are now searching for alternative materials.

"We surveyed 140 organic compounds," says Zaitseva, "all of which were prepared with characteristics known to be important for fast-neutron detection." A few such characteristics include the presence of benzene rings for efficient scintillation; high hydrogen content for interactions with neutrons; only low-atomic-number (low-*Z)* constituents, such as hydrogen or carbon, to avoid excessive interaction with gamma radiation; and a delayed emission to better show PSD.

"We bought the compounds as powders, purified them, and prepared the solutions," says Zaitseva. "Some had never before been used to grow crystals." Most of the materials have been well known for their scintillation properties since the 1950s. However, because of the limitations of electrical circuits available then for evaluating PSD, many of the compounds had never been evaluated for their neutron–gamma discrimination properties.

Experiments revealed that some molecules were close to or even better than stilbene at separating the neutron and gamma signals. Other materials produced plenty of light but did not exhibit PSD. When Zaitseva's team could not find an obvious correlation between PSD and molecular structure, they turned to Livermore experts in quantum molecular simulations. Computational modeling of organic molecules and their properties revealed how PSD is tied to the migration of certain excitations in the crystal. "Impurities in the crystals may also decrease PSD," notes Zaitseva.

For this initial survey, Zaitseva's team grew crystals to 1 centimeter using a simple evaporation technique, with growth rates of 1 to 2 millimeters per day. They also grew larger crystals (5 to 10 centimeters) in a custom-built, temperature-reduction device similar to the one used for KDP. Whereas many of the small crystals showed defects, the larger crystals were of high optical quality.

The first organic material grown in the custom device was 1,3,5-triphenylbenzene, which grew to 8 centimeters high and 5 centimeters across in just a week. This experiment proved that the rapid-growth technique is as effective for growing large single-crystal organic scintillators as it is for KDP. Since then, stilbene crystals have also been grown in the new crystallizer.

Rebecca Nikolić and her team are developing a detector made of silicon pillars and boron for capturing neutrons.

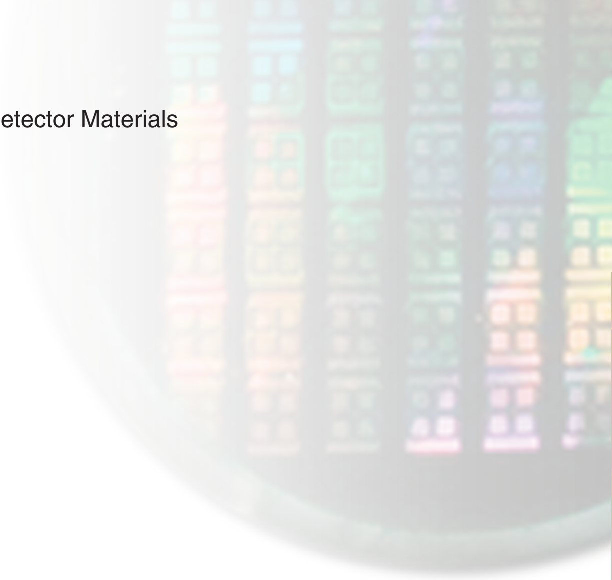
A Substitute for Helium

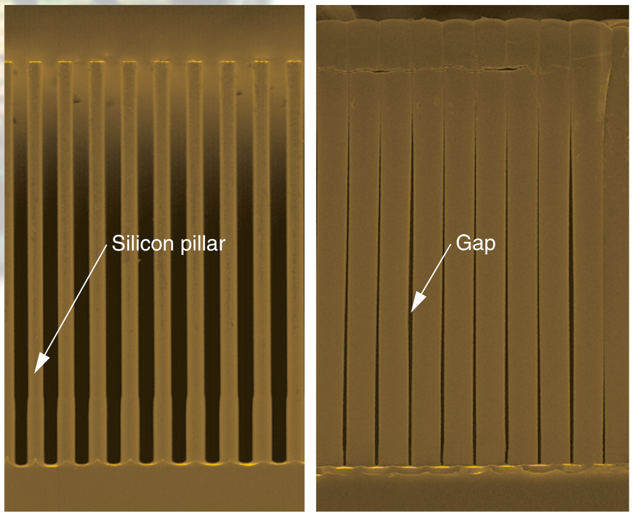
An attempt to hide a plutonium source might include not only shielding the material's gamma rays with heavy metal but also shielding its fast neutrons with plastic. Even if those two shields are effective, thermal neutrons may still be detectable. Fortunately, thermal neutron radiation is fairly rare compared to gamma radiation, and detecting its presence makes identifying the material in question that much easier.

In instruments for neutron detection, helium-3 has for years served as the neutron-absorbing material. However, finding a substitute for this material has become essential for the development of improved detectors. One disadvantage of helium-3 detectors is their size—the tube containing the helium can be up to 1 meter long. In addition, the device requires about 1,000 volts for operation. "Perhaps most importantly," says Payne, "helium-3 is much less available now than it was when the country was building nuclear weapons. Current supplies of helium-3 come, in part, from the dismantling of nuclear weapons where it accumulates as tritium decays." Supplies of this material are fast declining.

A solid-state material substitute for helium-3 has some advantages. Moving from a gas medium to a solid increases the density of the neutron-absorbing material, reducing the size of the detector. Previous researchers developed a two-dimensional detector, but its efficiency was low, only about 5 percent. (Note that with scintillation, as described above for gamma radiation and fast neutrons, the goal is high resolution, which is a low percentage. In contrast, the measure for effectiveness of thermal neutron detectors is efficiency, or the highest possible percentage.)

At Livermore's Center for Micro- and Nanotechnology, engineer Rebecca Nikolić and her team trekked through the periodic table and landed on a combination of silicon and boron, from which they have





The sensor developed by Nikolić's team uses a wafer with 50-micrometer-tall silicon pillars interspersed with boron to detect thermal neutrons. The left image shows the pillars before boron is added, and the right image shows boron-coated pillars with tiny gaps between each.

created a three-dimensional, pillar-shaped sensor. Incoming neutrons interact with boron to produce particles that, in turn, interact with the silicon semiconductor and create a current for an electronic signal. The silicon–boron pillar detector is expected to offer efficiency of more than 50 percent and require less power than the helium-3 tube. Because the silicon wafers can be cut to any size, detectors can be designed to meet the needs of many end users. For example, the wafer can be cut in smaller pieces for covert applications or tiled to cover large areas for portal monitors.

Initially, Nikolić's team etched a silicon wafer with pillars 20 micrometers high, and university collaborators used chemical vapor deposition to fill in the spaces between the pillars with boron. The prototype device had an efficiency of 20 percent, the highest efficiency reported for such a detector. And, it turns out that size matters. "We have found that taller pillars, which provide a thicker boron layer, are more efficient at capturing neutrons," says Nikolić. The team has recently completed a design with 50-micrometer-tall pillars to increase efficiency.

In almost every way, the three-dimensional silicon–boron wafer is superior to the helium-3 tube. The wafer device requires less than 3 volts for operation, and newer designs may require even less. In addition, this highly effective detector will have less than 5 percent the physical volume of the standard helium-3 detector for the same efficiency.

Going Forward

All three research teams are looking at several more years of research. Cherepy's team is continuing efforts to improve the performance of both garnets and strontium iodide. They are also exploring the possibility of engineering high-*Z* nanocomposite polymer scintillators, which could be highly efficient, inexpensive, and provide high resolution. While Zaitseva has shown that large organic crystals can be produced very quickly, her team has yet to determine the final optimal alternative to stilbene. An unexpected discovery was a compound with triple PSD that can discriminate among the three types of radiation—fast neutron versus gamma and thermal neutron versus gamma. This compound may even prove to be an effective material for detecting antineutrinos.

For materials being considered by all three teams, quantum simulations are used to identify and rank detrimental defects. Simulations also help guide optimal characterization experiments, whose results are fed back into future simulations.

"We have just begun to examine yet another possible technique for detecting neutrons," says Payne. "This method uses acoustic detection. Boron captures a neutron, which interacts with other materials, ultimately generating an acoustic wave. Livermore has the right mix of experts for meeting this research challenge."

—Katie Walter

Key Words: crystal growth, gamma radiation, garnet, helium-3, lanthanum bromide (cerium), neutron, radiation detection, scintillation detector, stilbene, strontium iodide, transparent ceramics.

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