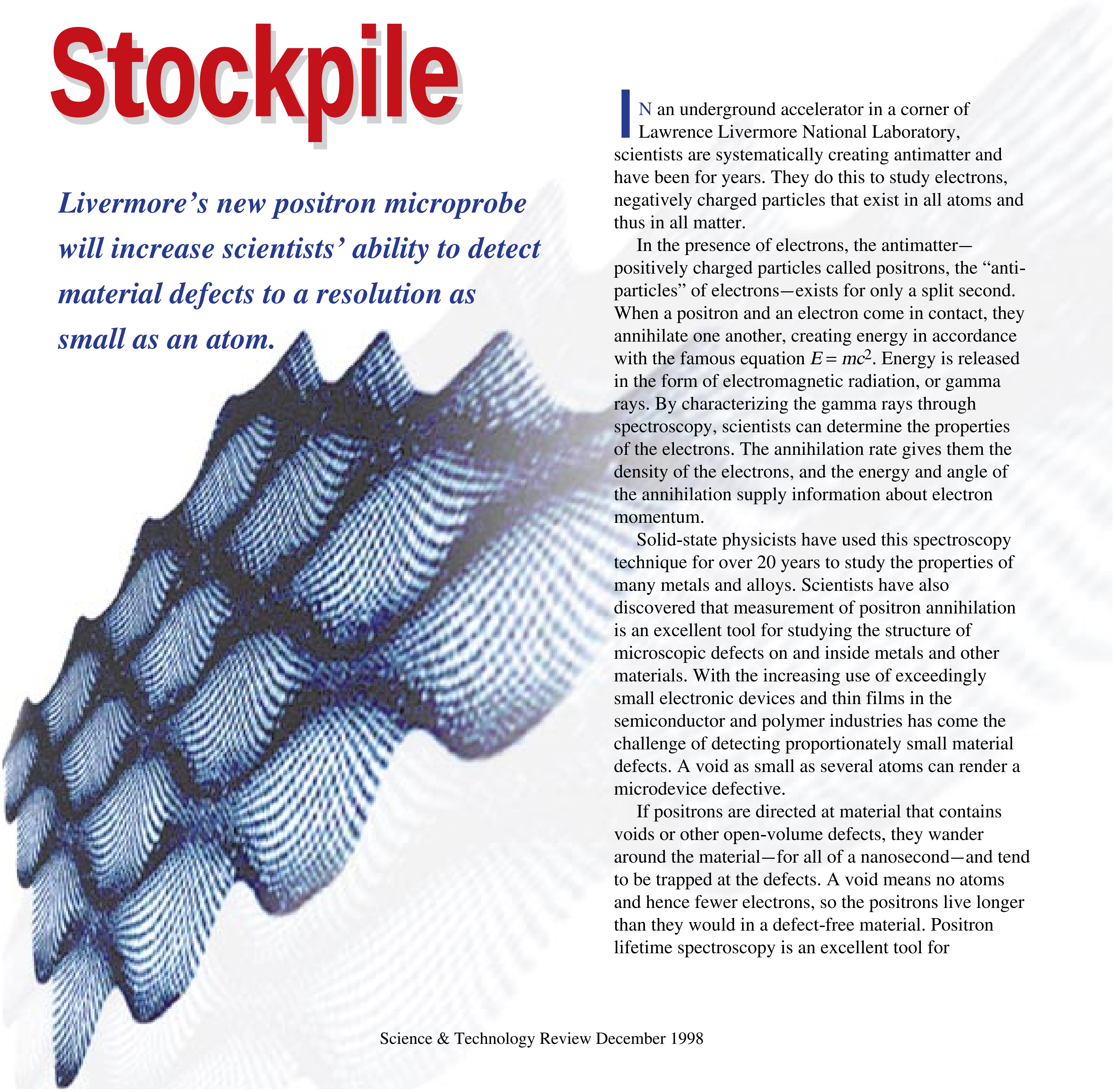
Antimatter Helps to Protect Our Nuclear Stockpile

Livermore's new positron microprobe will increase scientists' ability to detect material defects to a resolution as small as an atom.



IN an underground accelerator in a corner of Lawrence Livermore National Laboratory, scientists are systematically creating antimatter and have been for years. They do this to study electrons, negatively charged particles that exist in all atoms and thus in all matter.

In the presence of electrons, the antimatter—positively charged particles called positrons, the "anti-particles" of electrons—exists for only a split second. When a positron and an electron come in contact, they annihilate one another, creating energy in accordance with the famous equation $E = mc^2$. Energy is released in the form of electromagnetic radiation, or gamma rays. By characterizing the gamma rays through spectroscopy, scientists can determine the properties of the electrons. The annihilation rate gives them the density of the electrons, and the energy and angle of the annihilation supply information about electron momentum.

Solid-state physicists have used this spectroscopy technique for over 20 years to study the properties of many metals and alloys. Scientists have also discovered that measurement of positron annihilation is an excellent tool for studying the structure of microscopic defects on and inside metals and other materials. With the increasing use of exceedingly small electronic devices and thin films in the semiconductor and polymer industries has come the challenge of detecting proportionately small material defects. A void as small as several atoms can render a microdevice defective.

If positrons are directed at material that contains voids or other open-volume defects, they wander around the material—for all of a nanosecond—and tend to be trapped at the defects. A void means no atoms and hence fewer electrons, so the positrons live longer than they would in a defect-free material. Positron lifetime spectroscopy is an excellent tool for

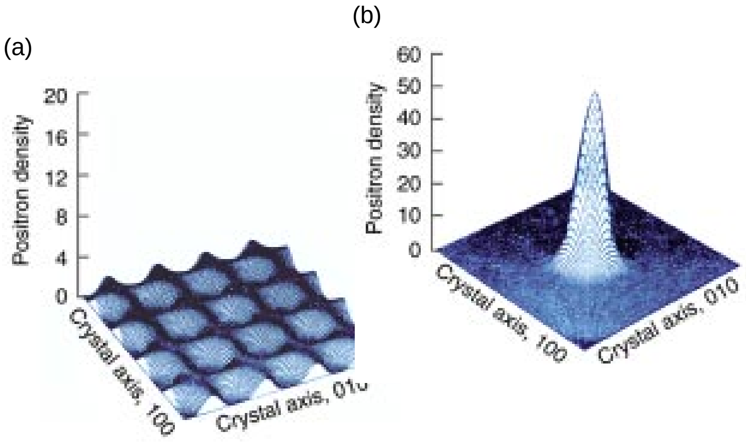


Figure 1. Images based on modeling calculations show how positrons are distributed in (a) defect-free material and (b) material with a defect. Positrons can migrate to defects as small as a single atom. The length of a positron's life is an indicator of the size of the defect.

nondestructive evaluation of defects as small as an atom (Figure 1).

Defects affect much of a material's behavior. They evolve from changes in temperature and pressure or over long periods of time, affecting the ultimate strength of the whole system. Livermore researchers have studied many problems caused by defects, including defect species in high-temperature superconductors, defects active during oxygen transport in uranium corrosion, and radiation damage in metals. Optical microscopy, neutron scattering, transmission electron microscopy, scanning tunneling microscopy, atomic force microscopy, and x-ray scattering are methods that have been developed to find vacancies, voids, and other defects in materials. Each technique is useful at specific depths for certain defect sizes and concentrations, and each has found its niche in appropriate research or manufacturing activities (Figure 2). But positron annihilation lifetime spectroscopy is the best of the lot for finding the smallest defects and a range of defect concentrations at virtually any depth in metals, semiconductors, and molecular or organic compounds.

As part of the Department of Energy's science-based Stockpile Stewardship Program, researchers at Livermore's Positron Facility, under the leadership of physicist Richard Howell, are enhancing current capabilities as well as developing new ones to detect changes in weapons materials. Livermore's new three-dimensional (3D) positron microprobe is scheduled to come online in the fall of 1999. Its pulsed positron beam with a radius of less than a micrometer will have the highest spatial resolution of any positron analysis system in the world.

Positron lifetime analysis can detect the size, location, and concentration of defects that result from stress, strain, radiation damage, fatigue, corrosion, delamination, and embrittlement. Says Howell, "Early detection is important because defects on the atomic level may ultimately evolve into mechanical failure."

He adds, "The new microprobe and its positron beam will aid stockpile stewardship and other projects at the Laboratory. Material scientists from other national laboratories, universities, and industry are interested in using them, too. The microprobe and our other positron facilities are enormously valuable for studying a variety of materials."

The development of the microprobe builds on Livermore's 20 years of experience with positron experiments related to magnetic fusion, superconductors, plasmas, high explosives, encapsulated plutonium, and aircraft composite materials.

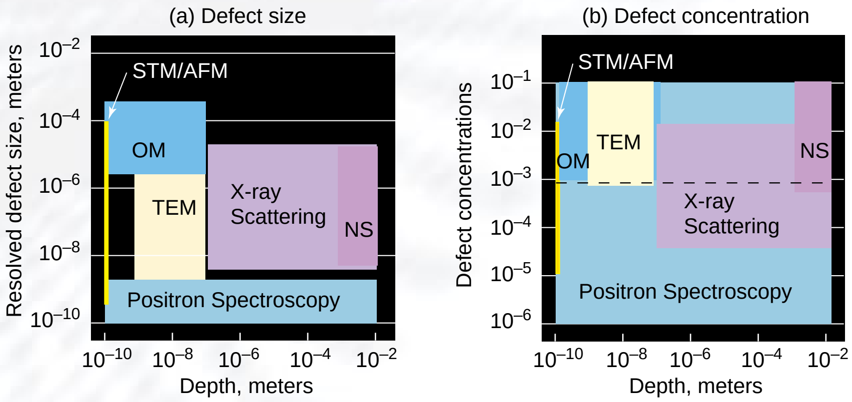


Figure 2. Of several methods for detecting defects in materials, positron spectroscopy can locate (a) the smallest defects and (b) the smallest defect concentrations at various depths. (STM = scanning tunneling microscopy; AFM = atomic force microscopy; NS = neutron scattering; OM = optical microscopy; TEM = transmission electron microscopy.)

Creating Positrons

Positrons are formed through naturally occurring radioactivity called beta (+) decay. When a radioactive nucleus decays via beta (+) decay, a positron is produced. Positron annihilation spectroscopy traditionally has been performed by placing a sample material in proximity to a radioactive source such as sodium-22. With the radioactive source inside an accelerator and the positron beam passing through a lens, the energy and direction of the positrons can be controlled. Livermore's high-energy Pelletron Accelerator uses this method of creating a beam of megaelectronvolt positrons.

Another process, known as pair production, can be used to create a high-current beam, or one with a high number of positrons, in the range of tens of billions of positrons per second. An electron beam from a linear accelerator (linac) hits a thick tungsten target and generates a high-energy photon beam. As the photons traverse the target, each is converted into an electron and a positron. All high-current positron beams, including Livermore's new 3D microprobe, use this method as their source of kiloelectronvolt positrons.

High Energies Go Deeper

Livermore's high-energy positron beam comes from a 3-megaelectronvolt Pelletron Accelerator, one of just two such sources in the world (Figure 3). The beam is derived from a sodium-22 source moderated by a tungsten foil positioned at the high-energy end of the accelerator. Both the moderated positrons and positrons directly emitted from the source are captured and accelerated. They produce a beam with a current of 850,000 positrons per second.

To measure positron lifetimes, the time at which the positrons are implanted in the sample is determined as each positron passes through a scintillator. Annihilation gamma rays from the implanted positrons are then detected by a barium fluoride detector. The annihilation lifetime is calculated from the time difference between the two detectors to a system resolution of about 250 picoseconds. Positrons leave the implantation detector with an average energy of 2.6 megaelectronvolts and a beam diameter of 1 centimeter directed at a sample placed 4 centimeters downstream. At this high energy level, positrons are implanted from millimeters to centimeters deep, depending on the density of the sample.

Says Howell, "We use the high-energy beam to analyze thick samples. The beam can even pass through thin windows before implanting positrons into a sample, allowing nondestructive *in situ* measurements in controlled environments." Up to 50 samples a day can be analyzed with this system.

This high-energy beam has been used to determine aging effects in aircraft components made of carbon fiber resin composites. Howell and his team measured the changes in hole volume brought on by accelerated aging at elevated temperatures and in hostile atmospheres. These data were correlated with infrared spectroscopy and mechanical tests to provide a complete description of the changes during aging.

For the DOE's Enhanced Surveillance Program, which is part of its Stockpile Stewardship Program, the high-energy beam is helping to determine the effects of self-irradiation on plutonium. Data from these experiments are used to validate Livermore models that predict the time scale of void swelling, embrittlement, and related radiation effects.

One experiment compared two samples of plutonium: a 21-year-old sample and one that had recently been cast. Positron lifetime spectroscopy revealed that most vacancies in both samples were filled with helium, which is produced naturally when plutonium decays. Despite high concentrations of these vacancies, which are a necessary precondition for void growth and swelling, no voids were found. The results of this experiment were good news for our stockpile.

3D with the Microprobe

The team uses the Positron Facility's 100-megaelectronvolt electron linac for its 3D scanning positron microprobe. The linac's electron beam produces

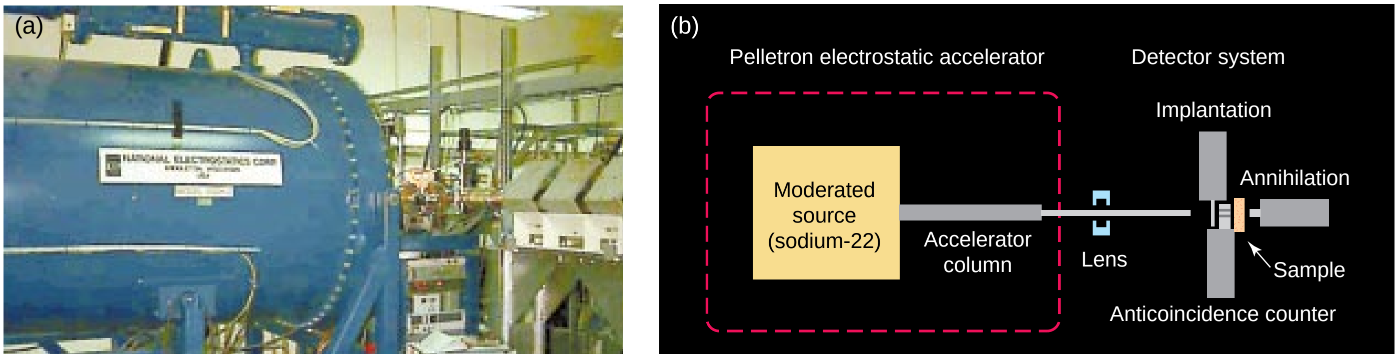


Figure 3. (a) Photo and (b) schematic of Livermore's high-energy positron annihilation lifetime system. Decay of sodium-22 produces positrons in the accelerator. Instrumentation near the sample measures when positrons are implanted in the sample and detects gamma rays produced when positrons annihilate with electrons in the sample. This system is used to study samples from millimeters to centimeters thick.

positrons through pair production for the highest current positron beam in the world. The electron beam has been upgraded to produce an even higher current beam of up to 10 billion (10^{10}) positrons per second.

Enormous numbers of positrons are necessary to produce the tightly focused beam that the microprobe needs for 3D mapping of defects with high spatial resolution. A pulsed beam and variable positron output energies in the range of 1 to 50 kiloelectronvolts are also required. Many individual features of this new system are found in other systems, but Livermore's positron microprobe system is the first to integrate all of them.

The microprobe is most similar to a scanning electron microscope, in which a tightly focused continuous electron beam scans small regions of a sample and detects defects. However, the scanning electron microscope cannot detect defects on the atomic scale, information that Livermore needs for stockpile stewardship and other applications. With the fast-pulsed, tightly focused beam of Livermore's new microprobe, positron lifetime annihilation spectroscopy will have spatial resolution at the atomic level.

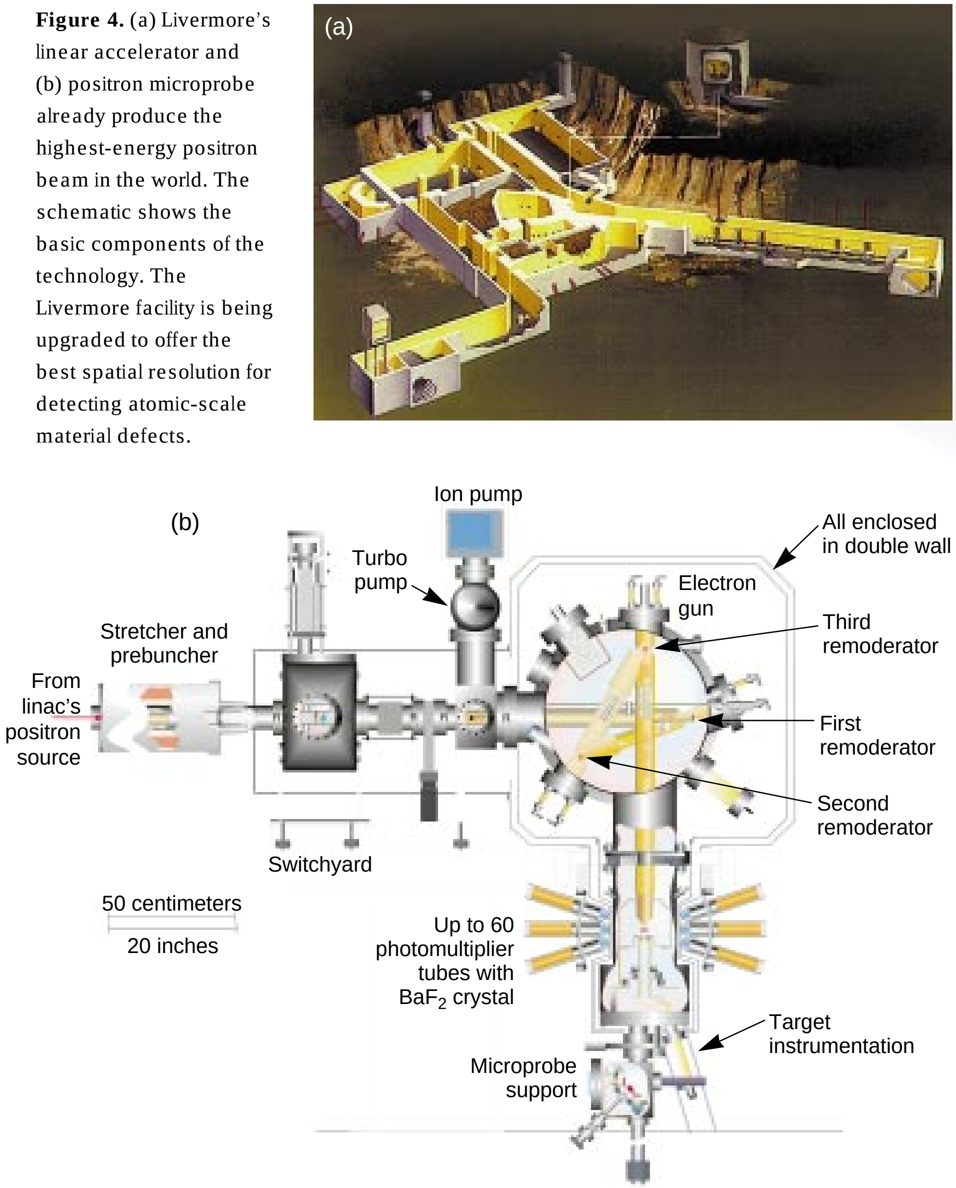


Figure 4. (a) Livermore's linear accelerator and (b) positron microprobe already produce the highest-energy positron beam in the world. The schematic shows the basic components of the technology. The Livermore facility is being upgraded to offer the best spatial resolution for detecting atomic-scale material defects.

Figure 4 shows a rendering of key elements of the 3D microprobe. As the initial high-current positron beam races through the system, its pulse is shortened and its diameter is focused to less than 1 micrometer. With the small spot size comes some loss of current, although the team has succeeded in keeping those losses to a minimum.[1]

By controlling the implantation location and energy of the microprobe's beam, the team will be able to detect and identify depth-dependent concentrations of vacancies, voids, gas-filled voids, and other negatively charged defects to a depth of a few micrometers. Typical depth and lateral resolution will be less than 0.2 micrometer. Maximum depth for implanting positrons with this beam will be 10 micrometers, which makes this mapping method ideal for very thin materials. The location of buried features can be identified with high precision by sweeping the energy and location of the positron beam in small steps.

The microprobe will give an entirely new view of defects in materials, and it will give scientists their first detailed look at defects around microscopic cracks. The microprobe will also supply valuable new information about defects at grain boundaries, which is important for stockpile stewardship studies of aging and stressed plutonium. It will give the closest look yet at electromigration, a defect caused by electrical current passing through a material. Electromigration is a major concern for the semiconductor industry because it can damage microcircuitry.

Data from positron beam experiments (Figure 5) are already serving to validate Livermore's modeling calculations for material defects and their effects. Spatially resolved data from the microprobe will provide a new level of detail for input to materials models as well as for further validation of Livermore calculations.

Defect Analysis Is Key

The Livermore system's upgraded high-current beam can be used without the microprobe to perform other forms of spectroscopy. Notes Howell, "The new beam will be very attractive to scientists outside Livermore who have no other access to such a high-current positron beam."

As so many products in our world shrink yet become more powerful, the ability to perform nondestructive analysis of these small devices at the atomic level is critical. Positron lifetime analysis is a particularly effective tool for defect identification and quantification, leading to smaller and better electronic devices, more effective polymeric coatings, and stronger and more corrosion-resistant metals and alloys. With newly upgraded positron experimental facilities, Livermore researchers will be positioned to break new ground in the analysis of material defects.

— Katie Walter

Key Words: electrons, linear accelerator, materials science, nondestructive analysis, Pelletron electrostatic accelerator, positron annihilation lifetime spectroscopy, positron microprobe, DOE Stockpile Stewardship Program.

Reference

1. For more information about the positron microprobe, see Richard H. Howell et al., "Positron Beam Lifetime Spectroscopy at Lawrence Livermore National Laboratory," *Application of Accelerators in Research and Industry*, J. L. Duggan and I. L. Morgan, Eds., New York: AIP Press, 1997.

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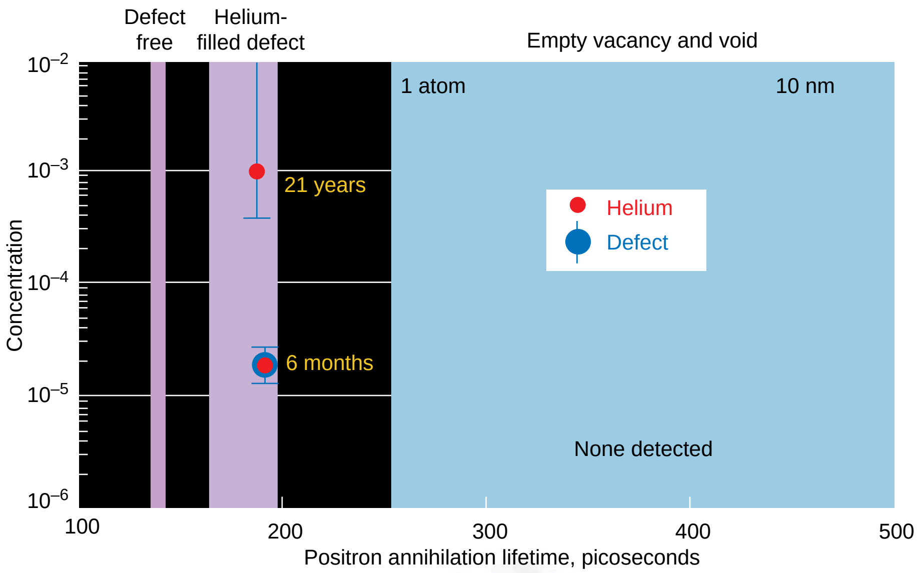
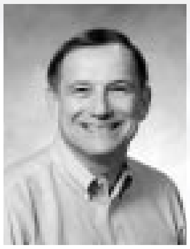


Figure 5. Shaded areas show modeling calculations for positron lifetimes in plutonium that is free of defects, has helium-filled defects, and has empty defects. (As plutonium decays, it produces helium atoms, so helium-filled defects are expected. Empty defects, on the other hand, are more problematic.) Data from experiments on 21-year-old plutonium and a recently cast sample gathered by Livermore's high-energy positron system fall within predictions for helium-filled vacancies and reveal no voids among the defects.

About the Scientist



RICHARD HOWELL received a B.A. in physics from Miami University in 1965 and a Ph.D. in physics from Michigan State University in 1972. When he joined Lawrence Livermore in 1972, he was a member of the Special Projects Division. Since 1974, Howell has been a member of the H Division in the Physics and Space Technology Directorate. Howell has performed basic physics and materials analysis with positrons for over 20 years, publishing over 130 articles on his work.