Los Alamos National Laboratory

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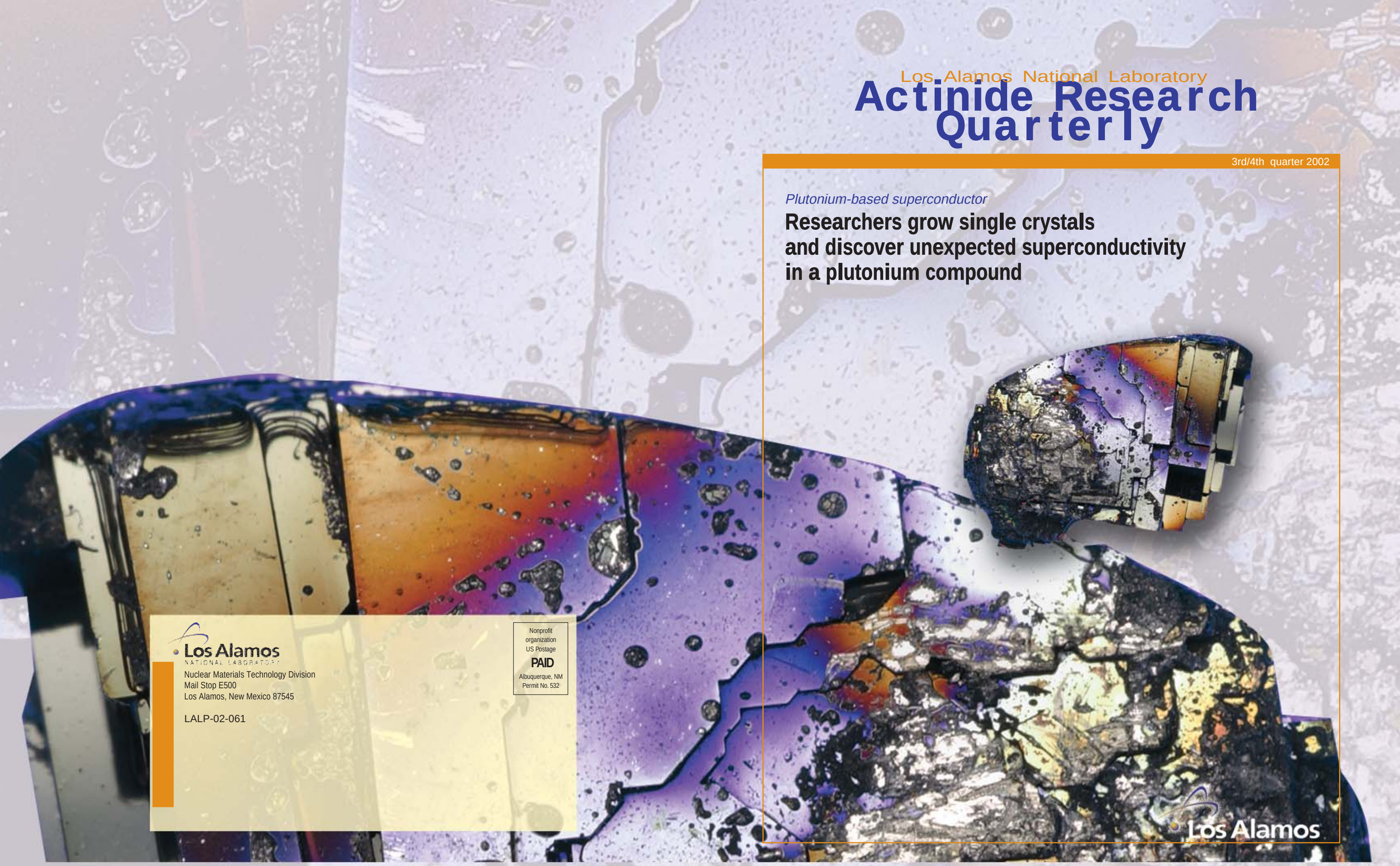

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About the Cover

photo by Mick Greenbank

The cover photograph shows a color-enhanced intermetallic crystal formed by the flux-growth technique. The crystal is a cerium-based analogue of a newly discovered plutonium-based superconductor. The story begins on the next page.

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Actinide Research Quarterly highlights recent achievements and ongoing programs of the Nuclear Materials Technology (NMT) Division. We welcome your suggestions and contributions. ARQ can be read on the World Wide Web at: http://www.lanl.gov/orgs/nmt/nmtdo/AQarchive/AQhome/AQhome.html.

NNSA National Nuclear Security Administration

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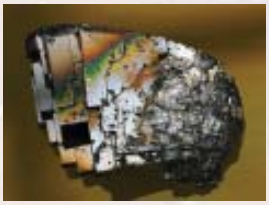
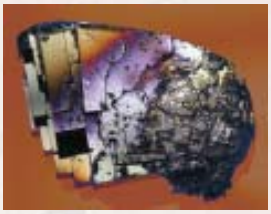
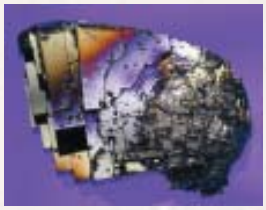






Plutonium-based superconductor

Researchers grow single crystals; discover unexpected superconductivity in a plutonium compound

The seemingly unrelated fields of plutonium metallurgy and unconventional superconductivity have a long and coupled history at Los Alamos. In the early 1980s Jim Smith, Zachary Fisk, and Sig Hecker developed much of the present understanding of the role of f electrons in influencing the metallurgical properties of elemental plutonium. The three were working in what was then Physical Metallurgy (CMB-5), parts of which now exist in both Nuclear Materials Science (NMT-16) and Condensed Matter and Thermal Physics (MST-10).

At the same time, Fisk and Smith discovered "heavy fermion superconductivity"—an unconventional type of superconductivity driven by the magnetic properties of f electrons—in UPt_3 and UBe_{13}, two uranium compounds that superconduct near 1 Kelvin and still attract international attention. This achievement is arguably the most significant in the history of experimental condensed-matter physics at Los Alamos and netted Fisk and Smith a major international prize, the American Physical Society International Prize for New Materials, in 1990.

This article was contributed by Luis A. Morales of Nuclear Materials Science (NMT-16), and John L. Sarrao and Joe D. Thompson of Condensed Matter and Thermal Physics (MST-10)

Los Alamos researchers Luis Morales (at microscope) and John Sarrao have discovered a plutonium-based superconductor.

Superconductor discovery

Recently, a renaissance in the interaction of plutonium metallurgy and condensed-matter physics has culminated in the discovery of the first plutonium-based superconductor—$PuCoGa_5$—a compound containing plutonium, cobalt, and gallium, which has an unexpectedly high superconducting transition temperature of 18.5 Kelvin (or –255 degrees Celsius).

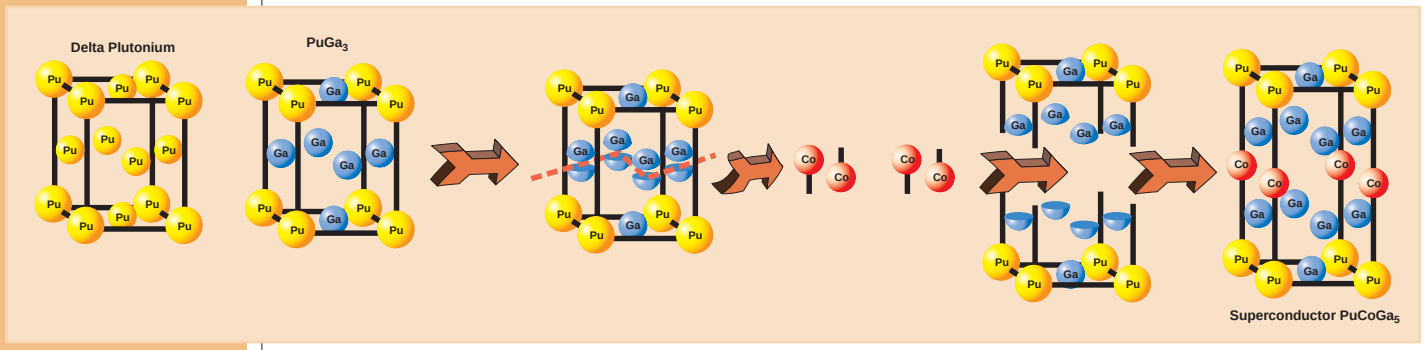
The discovery was reported in the Nov. 21, 2002, issue of the prestigious journal *Nature*.

Superconductivity is an unusual state of matter in which electrical current flows without resistance as a result of the material's electrons acting in pairs. Although the temperatures at which superconductivity is observed are usually quite low, superconductors are of interest both from a fundamental scientific perspective as well as for applications such as superconducting magnets. Only a handful of intermetallic compounds display superconductivity above 18 Kelvin.

A team led by Luis Morales of Nuclear Materials Science (NMT-16) and John Sarrao of Condensed Matter and Thermal Physics (MST-10) has established a capability within Wing 2 of the Chemistry and Metallurgy Research (CMR) Building to grow single crystals of transuranic intermetallic compounds using flux-growth techniques (see sidebar, page 4). The research is part of a Laboratory Directed Research and Development (LDRD) project whose goal is to advance the first-principles understanding of the electronic structure of plutonium.

Single crystals are valuable when trying to understand the intrinsic properties of materials, and this is especially true for compounds that

PLUTONIUM SUPER-CONDUCTOR



The cubic crystal structure of δ-phase plutonium is quite similar to that of plutonium-trigallium ($PuGa_3$). In the tetragonal crystal of plutonium-cobalt-pentagallium ($PuCoGa_5$), layers of plutonium-gallium and cobalt-digallium are alternately stacked along the crystallographic c axis. The relationship of the superconductor's crystal structure to the others can be simplistically appreciated as the insertion of a cobalt layer at the location indicated by the dotted red line in $PuGa_3$.

display radiation-induced self-damage, like those containing plutonium. This project is complementary to one led by Albert Migliori of the Materials Science and Technology (MST) Division's National High Magnetic Field Laboratory to advance the thermodynamic understanding of plutonium. Taken together, these efforts represent a significant investment in elaborating plutonium's fundamental properties.

A few surprises

The synthesis of new compounds is often serendipitous, and the case of the newly discovered plutonium-based superconductor was no exception. The researchers were exploring plutonium-cobalt-gallium ternary solutions as a precursor to growing single crystals of gallium-stabilized δ-phase plutonium when surprisingly large crystals resulted.

Working with Brian Scott of Actinide, Catalysis, and Separations Chemistry (C-SIC), the researchers determined that the crystal structure was that of a single crystal of the previously unreported tetragonal compound $PuCoGa_5$. The compound is a layered relative of δ-phase plutonium and plutonium-trigallium, in which layers of plutonium-trigallium and cobalt-digallium are alternately stacked along the crystallographic c axis.

Even more surprising to the researchers was the fact that their new compound displayed superconductivity at 18 Kelvin. Elemental plutonium is poised on the border between localized and itinerant f-electron behavior. This leads both to the complex metallurgy and significant differences that exist between α-phase plutonium (the phase at room temperature) and δ-phase plutonium (the phase stable at high temperatures) and makes it unlikely to display conventional superconductivity.

(For a general introduction to the properties of metallic plutonium, see the article by A. Michael Boring and James L. Smith in *Los Alamos Science* No. 26, 2000.)

The strong electron correlation effects that are present in plutonium tend to favor magnetic order that is generally harmful for superconductivity.

The researchers' current understanding of the plutonium-based superconductor suggests that the superconductivity they observe may be one of a very small handful of superconductors (the copper oxide-based high-temperature superconductors are the most famous representative of this family) whose superconductivity actually derives from magnetic correlations. The heavy fermion superconductors are another example of these materials.

In fact, the researchers think they have come full circle with their new plutonium-based superconductor in revisiting the problems pioneered by Fisk, Smith, and Hecker in understanding the role of f electrons in the metallurgy of plutonium.

A positive benefit

There are several more practical consequences of the fact that the superconductor contains plutonium. The superconducting transition temperature decreases as a function of time at a rate of about 0.25 degrees Celsius per month. However, this aging effect also has a positive benefit.

The critical current that a superconductor can support, which is important for applications, derives from the material's ability to trap and hold magnetic flux. The radiation-induced

damage that causes the decrease in transition temperature also increases its ability to pin magnetic flux and leads to a "critical current" that is quite large and increases with time. Critical current is the amount of electrical current that a superconductor can support and still display zero resistance.

If not for the fact that this property derives from the presence of plutonium, $PuCoGa_5$ would be an outstanding material from which to produce superconducting magnets.

Because researchers have learned so much about damage mechanisms from studying plutonium aging in other contexts, $PuCoGa_5$ promises to be an important test material in the research community's understanding of the so-called mixed state of superconductors, even if its commercial viability is limited.

(For more on damage mechanisms in aging plutonium, see the articles by Wilhelm G. Wolfer in *Los Alamos Science*, No. 26, 2000, and by Thomas Zocco and collaborators in *Actinide Research Quarterly*, 4th Quarter, 2001.)

Superconductivity and plutonium metallurgy

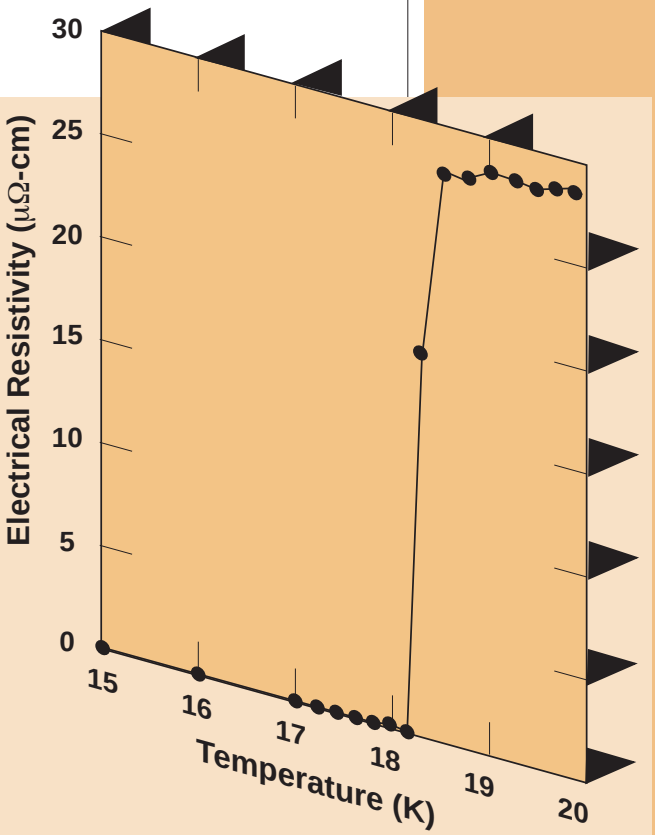
Superconductivity is an unusual phenomenon in which the electrical resistivity of a material drops to zero at some transition temperature—as seen, for example, in the accompanying plot for plutonium-cobalt-gallium$_5$ ($PuCoGa_5$). In essence, this means that the material offers no resistance to the flow of electrical current. Although this behavior is observed at rather low temperatures, it nonetheless holds a great deal of potential for technological applications, in addition to its considerable interest for scientific researchers.

Despite the hurdles that remain for practical implementation, superconductors offer the possibility of widespread delivery of electrical current without frictional losses. This could significantly lower the cost of electricity.

Nor are the applications of this phenomenon entirely futuristic: Magnets made from superconducting materials are routinely used for applications such as MRI (magnetic-resonance-imaging) instruments. For such magnets, the temperature at which a material superconducts is often less important than how much current the material can carry (the "critical current") before it loses its superconductivity.

The current-carrying capacity of a superconductor depends in large part on its microstructure and defect properties. It turns out that the damage mechanisms associated with radioactive decay of plutonium, which are well understood from studies of plutonium aging, are quite well suited for high critical currents in $PuCoGa_5$. If not for the health and safety issues associated with plutonium, $PuCoGa_5$ would be a rather ideal candidate for commercial use in superconducting magnets.

Most superconductors are understandable within the BCS (Bardeen-Cooper-Schrieffer) theory of superconductivity, a Nobel Prize-winning theory of the 1950s) and have transition temperatures below 10 Kelvin. In the last decade or so, a new family of superconductors, the high-T_c cuprates, have been discovered with much higher transition temperatures, about 100 Kelvin.



The higher transition temperature results from the presence of magnetic interactions in the material. Magnetism usually destroys superconductivity, but in this case (and in the case of $PuCoGa_5$), it appears that the higher transition temperatures are a consequence of the magnetism. The physics underlying this phenomenon is directly related to the degree of electron hybridization (for example, of the f electrons in plutonium), which also drives plutonium's complex metallurgy.

Plutonium Superconductor

Future study

While $PuCoGa_5$ may be an interesting compound unto itself, what prospects does it raise for advancing the general understanding of actinide materials?

Although the researchers have only been studying this material for a short while, it is already clear that it, like elemental plutonium, is poised on the boundary between localized and itinerant f-electron behavior, in which the f electrons can't decide whether they want to contribute to structural bonding, yielding complex low-symmetry structures, or remain uncoupled and only influence magnetic properties.

The ability of electronic structure calculations to correctly predict which limit is realized in real materials challenges the state of the art in the field. This uncertainty is a central factor in limiting understanding of the equation of state of plutonium.

$PuCoGa_5$ has already attracted attention within the international condensed-matter physics community, and electronic structure calculations have already been reported in Germany and Japan, as well as at Los Alamos. The accuracy of these calculations, which can be validated experimentally by further measurements of the properties of $PuCoGa_5$, will directly benefit the understanding of elemental plutonium's electronic structure.

The superconductivity in $PuCoGa_5$ has also been confirmed experimentally by a group at the Institute for Transuranic Elements in Karlsruhe, Germany. Stimulated by the Los Alamos researchers' discovery, this growing community promises to not only improve the understanding of $PuCoGa_5$, but also will engage a new generation of materials scientists in the challenges of plutonium.

And, because the properties of plutonium-containing intermetallic compounds are unexplored, only time will tell what additional surprises await scientists in their continuing synthesis of single-crystal compounds.

Growing single crystals

Photographed through a glovebox window, this single crystal of the superconductor plutonium-cobalt-pentagallium was formed using the flux-growth technique.

The method used to grow the single crystals discussed in this article is the flux-growth technique, which was initially championed by Zachary Fisk in the early 1980s. The technique involves dissolving the constituent elements of the desired compound in an excess of a low-melting metal—a flux; a process analogous to growing sugar crystals from supersaturated water solutions in high school chemistry.

The Los Alamos researchers grew their plutonium superconductor from excess gallium, but they have also grown single crystals from excess indium and antimony. To grow the crystals, the researchers place the starting material, including the excess flux, in an alumina crucible that is sealed in an evacuated quartz ampoule.

The sealed ampoule is heated to high temperature (about 1,000 degrees Celsius) and then cooled slowly over one or two days to an intermediate temperature of about 600 degrees Celsius. At this point, a centrifuge is used to separate the solid crystals from the excess liquid flux.

The resulting crystals are well faceted, large (about 1 gram of total mass is not difficult to achieve), and of high quality.

Call for papers

Plutonium Futures—The Science Conference

The second announcement and call for papers is available for the next Plutonium Futures—The Science Conference. The international conference will be held July 6–10, 2003, in Albuquerque, N.M. The conference addresses scientific and technical issues surrounding plutonium and other actinides and attempts to educate the public and students on these topics.

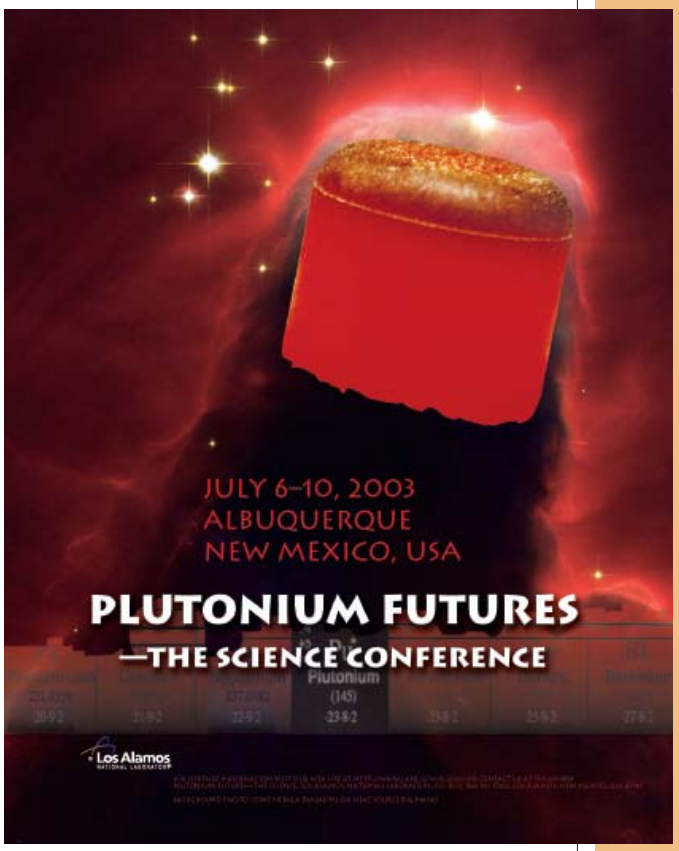
The conference committee expects an international scientific/academic conference similar to the conference held in Santa Fe, N.M., in 2000, which was attended by more than 400 international participants from 60 institutions and 15 countries.

Featured speakers will include Pierre D'hondt, director of reactor safety at SCK-CEN, who will speak about advances in mixed-oxide (MOX) fuel technology; Roland Schenkel from the Institute for Transuranium Elements, who will speak about the recent highlights of actinide research at ITU; and Teresa Fryberger from the Department of Energy Office of Science, who will speak about environmental aspects of plutonium and other actinides.

Nikolas Ponomorev-Stepnoi, vice president of Russian Research Center Kurchatov Institute, Vic Reis from SAIC, and Los Alamos' Sig Hecker also will be plenary speakers. Helen Caldicott, founder of Physicians for Social Responsibility, will speak at the banquet about medical implications of plutonium research.

The conference will kick off with a special tutorial for students on Sunday, July 6. On Monday, July 7, the plenary speakers will establish the motivational, historical, and scientific reasons for holding the conference.

On July 8–9, students and technical experts will present posters and presentations. The conference includes a banquet on Wednesday evening, July 9, and concludes on Thursday at noon.



The organizing committee encourages graduate students and postdoctoral fellows along with their research advisors to attend the conference. The committee will defray some or all of the costs for the students. The conference will pay for most of the travel and hotel expenses for students who present papers. Registration fees will be waived for students who attend but do not present papers.

The conference brochure and call for papers is available online at the conference site at www.lanl.gov/pu2003. For more information, contact Kathy DeLucas, (505) 665-3618 or duke@lanl.gov.

NDA

This article was contributed by Thomas E. Sampson of Safeguards Science and Technology (NIS-5)

Developments in NIS-5 are vital to Laboratory missions

Nondestructive assay and diagnostic techniques keep track of plutonium and uranium

Nondestructive assay (NDA) is a term applied to nuclear (mostly) measurement techniques for plutonium, uranium, and other actinides. Scientists in the Nuclear Materials Technology (NMT) Division rely on nondestructive assay in research areas as varied as plutonium disposition and heat source development for NASA missions. Many of the NDA technologies used at the Laboratory and around the world were developed by Los Alamos' Safeguards Science and Technology Group (NIS-5).

Collaborations between NIS-5 and NMT Division also have contributed to the success of stockpile stewardship, manufacturing, waste management, and nuclear material accounting programs. NIS-5, with support from the Department of Energy (DOE) Office of Policy Integration and Technical Support (SO-13), also works with other DOE facilities in addressing nuclear material accountability problems throughout the DOE complex.

NDA has two characteristics that make it attractive to researchers working with nuclear materials. First, it does not alter the physical or chemical state of the material. Second, the measurements can be made on bulk quantities of nuclear materials without breeching the container or containment of the material.

These characteristics allow NDA measurements to be made outside of gloveboxes, on entire 55-gallon drums, on filters in air ducts, on solutions inside processing systems, and on bulk materials packaged for disposition.

How NDA works

NDA techniques measure either the naturally occurring radiation emitted from actinide isotopes or the radiation that is stimulated or induced by another radioactive source. Most of the measured radiations are characteristic of and can be used to quantify the mass of a single isotope inside a sealed container.

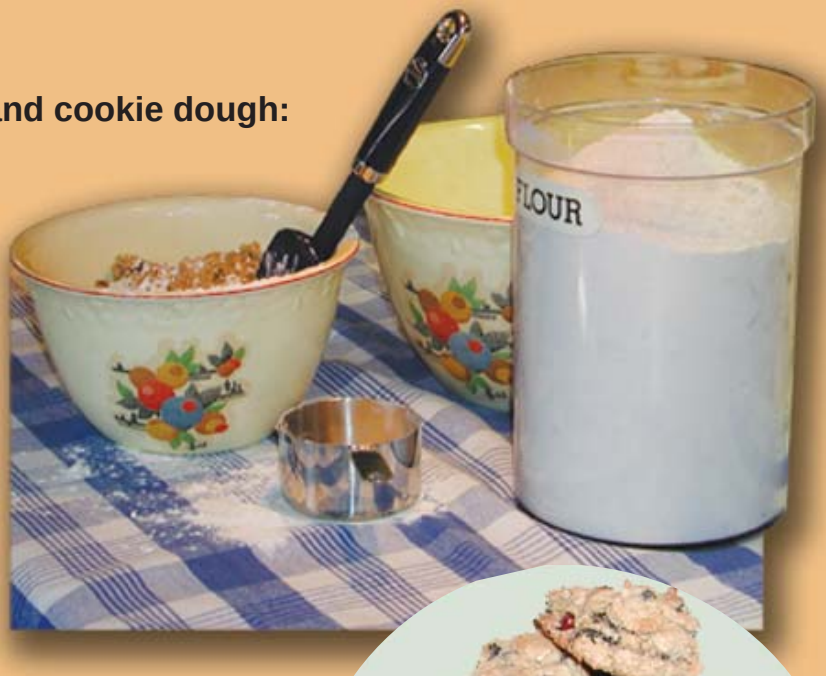
Three types of radiation provide the source for almost all NDA measurements: gamma rays or x-rays, neutrons, and heat.

Naturally occurring gamma-ray emission arises from the decay of most actinide isotopes. The energy and intensity of the gamma rays emitted from any isotope provide a unique signature that can be used to identify and quantify the isotope. X-rays are emitted in many decay processes or can arise from stimulation by other radioactive sources. X-ray emission is characteristic of the element while gamma-ray emission is characteristic of the isotope.

Several processes lead to the emission of neutrons from actinide materials. The most common process is the spontaneous fission of the plutonium-240 isotope present at a level of about 6 percent in weapons plutonium. In this process, several neutrons are emitted simultaneously when the nuclide undergoes spontaneous fission. The simultaneous detection of two or more of these neutrons provides a signature and a means of quantifying the mass of the spontaneously fissioning isotope.

Neutron-based NDA measurements are typically made with a neutron coincidence counter, a device that measures the rate of simultaneous neutron emission. The technology behind thermal neutron multiplicity counting was developed at Los Alamos and won an R&D 100 Award in 1992.

Nuclear material accountability and cookie dough: What's the connection?



Accounting for all of the plutonium passing through a facility process can be likened to accounting for all of the flour used in baking a batch of cookies. Consider how difficult it would be to do this for your baking project.

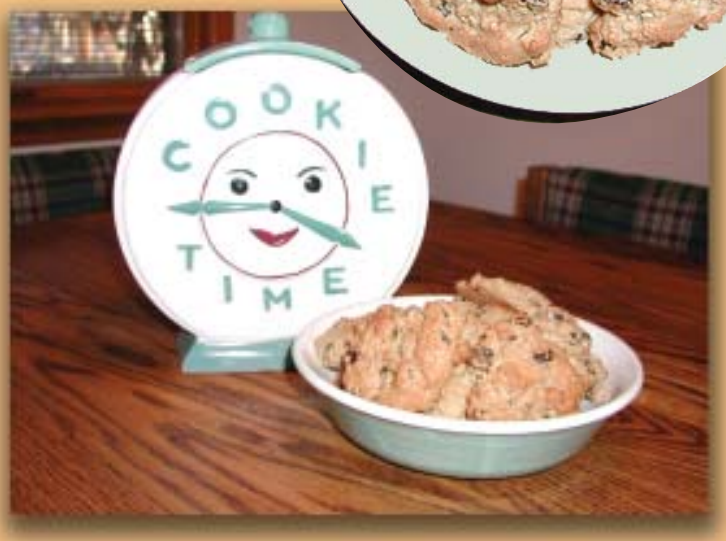
• Determine how much flour was in the bag to start with (correct to the nearest gram).

• Measure the flour into the mixing bowl and account for all that was spilled on the countertop or stuck in the measuring cup.

• Mix all the other ingredients with the flour. Remember, you are not interested in the other ingredients, only in the flour.

• Put the dough onto the cookie sheet. Account for the flour left in the cookie dough that remains in the mixing bowl and on the mixing spoon. (We won't complicate this by licking the spoon because the analogous process does not exist in plutonium processing.)

• Bake the cookies. Remove the cookies from the oven, cool them, and place them in cookie jar. Account for the flour in the crumbs that fall off the cookies. Account for the flour in the burned-on residue on the cookie sheet. (Rats! I set the temperature too high.)



• Measure the flour content of each cookie in the jar and double-check your measurement by measuring the flour content of the entire jar.

• "Balance the books" with a measurement of the amount of flour left in the bag.

NDA

The heat emitted from actinide materials arises primarily from alpha decay of the individual isotopes composing the sample. The amount of heat from a single gram of each isotope is a known physical quantity. The measurement of this heat in a calorimeter coupled with the measurement of the relative abundance of the individual isotopes in the sample (isotopic composition) provides a nondestructive measurement of the elemental mass in the sample.

Detection and quantification of these types of radiation provide the basis for the NDA techniques that are used today for nuclear material control and accounting in all nuclear facilities (see sidebar on page 7).

These same techniques also form the basis for quantitative measurements on waste for disposal, qualitative detection of the presence of nuclear materials for physical security and homeland security applications, and the quantification of pure plutonium materials for disposition under international treaties.

Holdup measurements—portable instrumentation

The nuclear material residues that become trapped or held up in the piping and ductwork of a processing facility must be measured to provide assurances of criticality safety and to ensure complete accounting of all the material processed. These "holdup" materials provide a particularly difficult measurement challenge because of measurement geometry and access problems.

Los Alamos scientists recently tested two new portable gamma-ray technologies online.

The first is a compact, lightweight, battery-powered cadmium telluride (CdTe) detector that measures gamma-ray isotopic composition"*in-situ* without liquid nitrogen. This commercially available, room-temperature semiconductor detector is easily positioned in online measurement locations without the difficulties associated with comparatively bulky, liquid-nitrogen-cooled, high-purity germanium detectors.

In a first-ever application, Los Alamos scientists have measured the complete plutonium isotopic composition for a wide range of materials using the room-temperature CdTe detector. The testing covered a range of three percent to twenty-six percent plutonium-240. Development for uranium analysis is under way. Applications currently include online inventory in process equipment or containers (before welding to verify loading limits), and verification of the inventory in storage vaults.

The second technology is a new automated system for making holdup measurements in process equipment. These measurements use

Taking measurements of nuclear materials held up in piping and ductwork is easier and safer, thanks to analysis software developed by researchers in the Safeguards Science and Technology Group (NIS-5) in a cooperative effort with Oak Ridge Y-12 Plant staff. Several different types of small, portable detectors can be attached to a telescoping pole to quantify materials trapped in piping and ductwork, as shown in the photo at the right. The portable detectors make the job of the man in the cartoon on page 8 a lot easier and safer. In the photo below, Duc Vo (left) and James Pecos prepare to make a measurement of the isotopic composition of the plutonium in a container of plutonium oxide using a cadmium telluride (CdTe) detector at the TA-55 Plutonium Facility, The container is visible through the glovebox window. Vo holds the portable CdTe detector in his right hand and the battery-powered electronics in his left. The same portable electronics used with the CdTe detector is also used with other detector types for the holdup measurements pictured at the right.

gamma rays from individual isotopes to quantify plutonium-239 and uranium-235, as well as other isotopes. Software developed in a cooperative effort between Los Alamos and Oak Ridge Y-12 Plant staff automates data acquisition with the newest portable gamma-ray spectrometers.

New Los Alamos-developed algorithms accurately determine holdup quantities using analysis algorithms that correct for departures of real deposits from ideal geometries and for gamma-ray self-attenuation. Automation allows rapid execution of large numbers of measurements in very short count times (five to fifteen seconds) and rapid quantification of measurement results. These new measurement methods reduce radiation exposure as well as minimize overall measurement costs.

Solution assay

Researchers have been processing and purifying uranium and plutonium in solution since the dawn of the atomic age, and in the 1970s they began using nondestructive assay because it is relatively straightforward to apply to solutions because of their uniform composition.

The gamma-ray emission from plutonium-239 is most often used to quantify that isotope, while gamma rays from uranium-235 serve the purpose well for uranium. The measurement techniques incorporate methods for correcting for the self-absorption of the gamma rays produced in the solution; a correction required to measure solutions with varying concentrations of plutonium or uranium.

NDA methods for solutions can be applied

NDA

The instrument operator places a plastic vial containing a plutonium-bearing solution into the measurement head of the Solution Assay Instrument (SAI) inside a glovebox. A high-purity germanium detector, located outside the glovebox underneath the SAI head, measures the gamma rays from the samples after they penetrate the stainless steel floor of the glovebox.

to many different configurations, including ion-exchange columns for plutonium, solvent-extraction systems for uranium, solutions flowing in pipes, and small samples of approximately 50 milliliters in plastic vials.

At Los Alamos, plutonium solution accountability measurements are usually performed with the solution in a plastic vial inside a glovebox and the detector outside "looking" through the glovebox floor.

By carefully applying these techniques, researchers can get results that are equivalent to conventional analytical chemistry measurements without producing the wastes associated with chemical analysis.

These techniques were applied to enriched uranium in a technology called the Nuclear Materials Solution Assay System, which also earned an R&D 100 Award for the Laboratory in 1988.

Calorimetry

NDA measurements of plutonium in bulk samples in sealed containers are primarily done through a technique called calorimetry, which measures the total power (watts) produced by a plutonium sample.

Calorimeters traditionally have been fabricated using a sensor of nickel wire wound around the measurement chamber. The nickel wire provides a temperature-sensitive resistance leading to highly accurate and precise electrical measurements of the power produced by the sample.

This fabrication method, however, is somewhat of an art and the few experienced practitioners are retiring. A team of researchers is developing calorimeters with thermopile-based solid-state sensors. These new sensors reduce fabrication costs and can be configured to make the calorimeters more sensitive than traditional wire-wound calorimeters—measuring samples as small as 1 gram of plutonium.

This new calorimetry technology is being applied to the measurement of kilogram quantities of enriched uranium, an application that will improve controlling and accounting for this difficult-to-measure material.

Researchers at Los Alamos also are building a mobile calorimetry laboratory that will take this new technology to sites around the country to assist in measurements of materials slated for disposal. This mobile user facility will make measurements using four different calorimeters. The calorimeters range in size from two inches in diameter to more than two feet in diameter, which is large enough to do the first-ever calorimetry measurements on fifty-five-gallon waste drums

Gamma-ray isotopic analysis

Because most NDA measurements only quantify a single isotope, these measurements require knowledge of the isotopic composition of the measured material to convert the measurement results for the single isotope to elemental mass. Gamma-ray spectrometry is used to nondestructively determine the isotopic composition of essentially all materials present in the nuclear fuel cycle, both plutonium and uranium.

Gamma-ray spectrometry measurements are performed on samples of arbitrary size, geometry, and physical and chemical composition, and do not require calibration, or even knowledge, of the containment of the sample. This technique, now in worldwide use, was developed at Los Alamos in the mid-1970s.

Los Alamos researchers also developed software called PC-FRAM for this type of analysis. PC-FRAM is the most advanced software of its type in the world and affords an analysis of plutonium isotopic composition of samples contained in as much as 2.5 centimeters of lead.

The commercially available software analyzes all the isotopic measurements at the

Los Alamos isotopic analysis software can determine the isotopic composition of plutonium components inside a weapons storage container (above) as well as plutonium in lead-shielded short-term storage containers (below).

NDA

The nondestructive assay module of the Advanced Recovery and Integrated Extraction System (ARIES) at TA-55 is used to quantify the plutonium oxide converted from the plutonium metal in dismantled weapons and encapsulated in containers like the one shown in the inset. The triple-ply containers are approved by the Department of Energy for long-term storage of up to 50 years.

TA-55 Plutonium Facility as well as all Los Alamos waste sent to the Waste Isolation Pilot Plant (WIPP). It can verify the contents of weapons storage containers and is used in inspections by the International Atomic Energy Agency.

Integrated NDA systems

Los Alamos researchers have combined several NDA instruments into a robot-controlled system under a central host-computer control.

The Advanced Recovery and Integrated Extraction System (ARIES) is a set of six modules that extracts the plutonium metal from surplus weapons components, converts the metal to oxide, and packages the oxide in containers suitable for long-term storage or disposition.

The ARIES NDA system quantifies the plutonium oxide from dismantled weapons components in preparation for mixed-oxide (MOX) fuel fabrication and ultimate disposal.

The ARIES NDA module performs unattended, around-the-clock measurements with calorimetry, neutron coincidence multiplicity counting, and gamma-ray isotopic analysis. This part of the ARIES system is the model for a functionally similar system being built in Russia to assist the Russian plutonium disposition program.

This system was developed as the prototype for the NDA system to be installed in the Pit Disassembly and Conversion Facility (PDCF) to be constructed at the Savannah River Site.

Hanford and Pacific Northwest National Laboratory

Six decades of actinide production and cleanup

The first non-Indian settlers arrived in the Columbia River basin in what is now southcentral Washington state in the mid-1800s. They found a dry, almost treeless desert; the major rivers through the area—the Columbia, Snake, and Yakima—had little effect on the sagebrush-dominated landscape.

Irrigation projects built in the early 1900s allowed a small number of farmers to scratch out a living. Richland, Hanford, and White Bluffs, the farming towns along the river, weathered economic ups and downs as the area was affected by drought, the Depression, and the construction of Grand Coulee Dam. World War II brought an end to two of the towns and radically changed the third.

The Hanford Site is located in a semiarid shrub-steppe environment in southeastern Washington.

World War II and the Manhattan Project

Research with uranium-235 demonstrated the feasibility of a uranium atomic bomb, but that isotope was rare and difficult to separate. In March 1942, Glenn Seaborg's group at the University of California produced the first plutonium-239. The U.S. Army Corps of Engineers was given the responsiblity of developing both uranium and plutonium weapons. In June 1942, the Manhattan Engineering District was formed to accomplish the task.

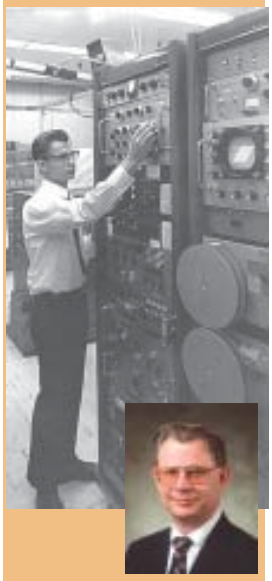
Plutonium production required a nuclear reactor to transmute uranium and chemical separation plants to extract the plutonium and purify it. In addition to the scientific challenges, the Manhattan Project had to deal with the demands of wartime secrecy, chronic shortages, and the poorly understood dangers of this radioactive element. Site selection criteria called for a large, undeveloped, remote, sparsely populated area with a supply of clean water and electricity.

The area along the Columbia River was selected. The towns of White Bluffs and Hanford, along with the surrounding farms, were confiscated. Richland was turned into a government town. Groundbreaking at the site took place in March 1943.

The towns of White Bluff and Hanford were evacuated in 1943 to build plutonium production facilities. Because plutonium had never been produced on a large scale and there was the potential for accidents, the production facilities had to be located away from the populated East Coast and other Manhattan Project sites. Between 1,200 and 1,500 people were evicted in a one-month period. The high school in Hanford is one of the few structures that remains from the old towns. These two photos show the high school as it appeared in the early 1950s (top) and today (bottom).

History: Hanford and PNNL

This editorial was contributed by Dr. Ned Wogman, director of Science and Technology for National Security and director of Homeland Security at Pacific Northwest National Laboratory.



Guest contributor Dr. Ned Wogman then, as a young scientist in the 1960s working with gamma-ray spectroscopy of lunar material, and now.

HISTORY: HANFORD AND PNNL

In less than two years and under a cloud of secrecy, the reactors and facilities necessary to produce the plutonium used in nuclear weapons to end World War II were built at the Hanford Site. By October 1944, the first reprocessing facility, T-Plant (in the background), began operating. U-Plant (in the foreground) was under construction during the mid-1940s.

Thirty months later the site had three nuclear reactors, three processing canyons, sixty-four underground storage tanks for high-level waste, 385 miles of road, 158 miles of railroad, thirty miles of electrical transmission lines, and hundreds of miles of fence. Richland changed from a farming town of 500 to a government town of 17,500—and another 50,000 workers were housed just north of Richland.

The first chemical processing plant was designated T-Plant. This was the world's first large-scale plutonium separations facility. The plant contained twenty-two sections with two cells in each section.

Once in operation, high levels of radioactivity would preclude personal access to the cells, so the separations equipment was designed for remote operation. B-Plant was similarly constructed but was sixty-five feet shorter, as it lacked the head-end testing cell.

photos courtesy of Pacific Northwest National Laboratory

Both plants used the bismuth phosphate process. In this process, the cladding jackets were first dissolved from the fuel rod. Then a series of precipitation, centrifugation, and redissolution steps purified the plutonium. The valence of the plutonium was manipulated to keep it in solution (+4) or to precipitate it (+6).

The solution coming out of T- and B-Plants went through a bulk reduction process, a batch process that reduced 330-gallon batches to eight gallons. The final stage was isolation. Hexavalent plutonium was precipitated as plutonium peroxide, then dissolved in nitric acid and boiled to produce a wet nitrate paste.

Originally the bismuth phosphate process took twenty-six hours to extract 250 grams of plutonium from one ton of irradiated fuel. By 1955, process improvements reduced the cycle time to four-and-a-half hours.

The plutonium nitrate paste was shipped to Los Alamos, N.M., for conversion to metallic plutonium. The first shipment of plutonium left Hanford on Feb. 2, 1945, and, after traveling by way of Portland and Los Angeles, arrived in Los Alamos on Feb. 3, starting a long association between Los Alamos and Hanford.

The first shipments culminated in the construction of the first nuclear bomb, which was detonated on July 16, 1945, at the Trinity Site near Alamogordo, N.M. On Aug. 9, 1945, a bomb containing Hanford plutonium was detonated over Nagasaki, Japan. Five days later, Japan surrendered.

The future of the Hanford Site was thrown into uncertainty. By December 1946, site employment had dropped from 10,000 to 5,000. The Manhattan Project assumed a caretaker role, and power was reduced on the operating reactors.

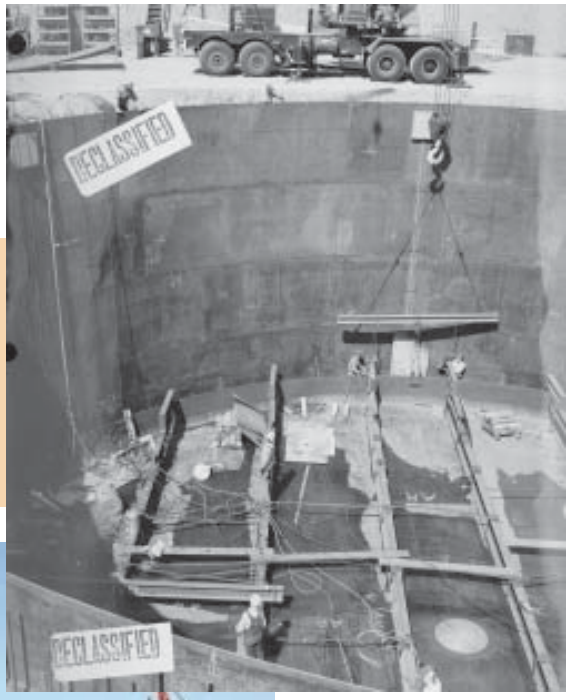
The Cold War

The postwar offer by President Truman to transition control of nuclear weapons and energy to the United Nations was vetoed by the USSR, which was pursuing its own nuclear weapons program. In 1947, nuclear weapons production became a priority. In March 1947, President Truman "declared" the Cold War.

Two new reactors were brought online at Hanford in 1949 and 1950, along with a new separations plant, Z-Plant, or the Plutonium Finishing Plant. Up to that time, the separations process at Hanford produced a wet plutonium nitrate paste that was shipped to Los Alamos for final extraction of plutonium metal. At Z-Plant, oxalate, oxide, and fluoride processing steps produced "buttons"—metallic plutonium in disks resembling hockey pucks.

The Cold War produced military and political pressures: the communist takeover of Czechoslovakia, the Berlin Airlift, the Russian A-bomb, Mao's takeover of China, the Rosenberg-Fuchs-Greenglass-Hiss spy cases, NATO, McCarthyism, and the Warsaw Pact.

Hanford responded with new facilities. The REDOX plant was designed



A steel girder is lowered by a crane to the floor as construction begins on the inner wall of a 1.1-million-gallon double-shell tank.

The Hanford Site contains 177 cylindrical underground storage tanks with holding capacities ranging from 55,000 to 1.1 million gallons. The tanks, built between 1943 and 1985, contain approximately 54 million gallons of hazardous and radioactive wastes; enough to fill nearly 2,700 railroad tanker cars. The first tanks built had a single carbon-steel wall and floor covered by a dome and outer shell made of concrete. The newer double-shell tanks contain two carbon-steel liners along the walls and floor and a single steel dome liner. All of these were enclosed within an outer shell of reinforced concrete. Double-shell tanks were built starting in 1968.

History: Hanford and PNNL

The N-Reactor, a plutonium production reactor located on the Hanford Site, operated from 1963 to 1987. The reactor's main mission was to produce weapons-grade plutonium; however, the reactor could also produce steam to generate electricity. The long building on the left is the power-generating plant.

beginning in 1947, constructed beginning in 1949, and went operational in 1952. The process used methyl isobutyl ketone and aluminum nitrate in the first continuous processing plutonium extraction operation.

The plant included a 132-foot-tall silo to house packed columns of ion-exchange material to purify the plutonium and remove fission products. The REDOX plant started processing 3.125 metric tons of irradiated fuel per day; by 1958 it was processing eleven to twelve tons per day.

U-Plant, which had been a training facility for T- and B-Plants, was retrofitted to use tributyl phosphate and saturated kerosene to extract uranium from waste solutions; most of the uranium supply in the United States was in Hanford's waste tanks. Ferrocyanide was added to waste streams to precipitate cesium-137. Two more reactors were built, twenty-one new single-shell underground storage tanks were built, and the PUREX plant came online.

PUREX, the plutonium-uranium extraction plant, came about from the realization that the REDOX process used dangerous explosive chemicals, lacked the capacity to meet the perceived need as the Korean War escalated, and was expensive because the aluminum nitrate could not be recycled.

A study group was formed in 1951 to develop a process to address those issues and handle 200 metric tons of irradiated fuel per month, increasing to 400 metric tons when the large KE and KW reactors came on line. The 1,000-foot-long building was completed in 1955; hot start-up was in January 1956.

The unique feature of PUREX was pulse columns to put organic and water solvents into contact for chemical separation. These small columns replaced the packed columns of the REDOX process and reduced construction costs. In 1972, PUREX started a scheduled eighteen-month shutdown for planned upgrades to accommodate N-Reactor fuel. The shutdown lasted eleven years.

Concern about leaking single-shell underground storage tanks resulted in the construction of double-shelled tanks. Major safety upgrades were completed, and two new cells were constructed to convert plutonium nitrate to the safer plutonium oxide powder.

At restart in 1983, the design capacity was 3,000 metric tons per year—about eight metric tons per day. The average processing, however, was three metric tons per day.

The Plutonium Finishing Plant completes the chemical processing story. It was built to convert plutonium nitrate paste to metallic plutonium because of safety concerns about shipping the paste to Los Alamos.

Construction began in 1948. The plant used Los Alamos chemistry in a series of interconnected gloveboxes. The plutonium nitrate from the separations plant was purified through an oxalate precipitation step. Then hydrogen fluoride gas was forced through the precipitate at high temperature to produce plutonium tetrafluoride powder.

The powder was reduced with calcium, gallium, and small amounts of other

substances in a vacuum at high temperature to produce buttons of plutonium metal, similar in size to hockey pucks. The buttons were shaped in the plant for use in weapons until 1965, when shaping was taken over by Rocky Flats outside of Denver, Colo. In 1962, the plant started producing plutonium for use in commercial power reactors as part of an Atomic Energy Commission program.

By 1968, 30 percent of the output was destined for EURATOM reactors and nuclear research. In 1973, operations slowed with the PUREX closure and the plant was upgraded to accept powdered plutonium oxide. The plant restarted in 1984, after the 1983 PUREX restart. Final closure came in June 1989.

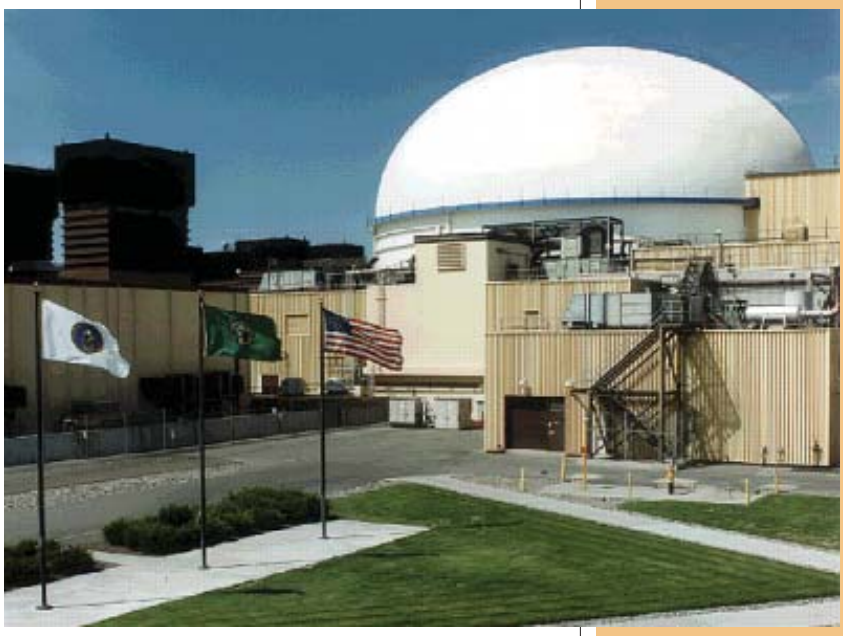
President Eisenhower's Atoms for Peace program led Hanford's contractor, General Electric, to form the Hanford Laboratories in 1955 to develop plutonium technology for power reactors. In 1963, GE decided to pull out of Hanford to avoid a possible conflict of interest with GE's commercial reactor business.

Site operations were segmented into reactor operations, chemical separation, fuel production, engineering, and research, and put out for bid. The research segment was renamed Pacific Northwest Laboratory (PNL); Battelle successfully bid on the contract and began operating the Laboratory in 1965.

In the first ten years, Battelle built six laboratory and support buildings in Richland, established a research site in Seattle and a marine laboratory on the Olympic Peninsula, and purchased a research aircraft. Staffing grew from 2,200 to 2,800.

The Lab provided the engineering basis for N-Reactor, a plutonium production reactor that generated commercial power with the steam produced by the reactor coolant.

The Lab also started the Fast Flux Test Facility (FFTF), a liquid metal fast breeder reactor. This program was lost in 1975 in a management disagreement with the Atomic Energy Commission. The program staff that transferred with the program dropped PNL employment to 1,300.



The Fast Flux Test Facility (FFTF), located north of Richland, is a 400-megawatt thermal, liquid metal (sodium) cooled reactor. The white dome in the background is the containment building that holds the reactor vessel. The building was designed to prevent the release of radioactive material into the atmosphere in case of a severe reactor accident.

The loss of the FFTF, coupled with uncertainty in the subsequent years as the Atomic Energy Commission was replaced by the Energy Research and Development Administration (ERDA), which was in turn replaced by the Department of Energy (DOE), led to a reevaluation by the Laboratory of its future. Emphasis was shifted from engineering to research and development and a diversification of both mission and customer bases. By 1985, staffing had increased to 2,800.

In April 1986, a nuclear power plant exploded in Chernobyl in the former USSR, now Ukraine. Like the N-Reactor at Hanford, the Chernobyl reactor was a graphite core reactor. Public opinion led to the closure of N-Reactor based on the fear that a similar accident was possible at Hanford. The fall of the Soviet Union in the early 1990s effectively ended nuclear weapons programs at Hanford.

HISTORY: HANFORD AND PNNL

Tank waste at Hanford varies from crystallized material called saltcake, shown in the top photo inside a single-shell tank, to clear liquids. Saltcake in waste tanks was produced after waste was processed through concentrators that boiled off water, reducing the volume of waste. Slightly concentrated waste was then returned to the tanks where solids crystallized and settled as the solution cooled. In the lower photo, high-pressure water is used to blast simulated saltcake into smaller fragments that can be more easily removed from the single-shell tanks.

Cleanup after the Cold War

Forty years of plutonium production, accomplished under a veil of secrecy, left a legacy of waste at Hanford. Some of the waste was contained in known locations—underground storage tanks and buried drums. Other waste had been discharged into the ground. Most of the waste contained radioactive isotopes and transuranic elements.

Natural groundwater was in contact with waste plumes and contamination was seeping into the Columbia River. The Hanford Site held two-thirds of all nuclear waste, by volume, in the DOE complex, including 177 underground tanks (sixty-eight suspected or known leakers) holding fifty-three million gallons of waste containing 200 million curies of radioactive materials.

Various cleanup scenarios were presented, often based on scant scientific evidence, that would take almost a century to complete and cost hundreds of billions of dollars. Fortunately, the public hysteria died down as new crises *du jour* came along, and scientists began the analysis necessary to remediate the site.

Characterization of the waste is an ongoing process. Over the years, waste streams from different processes were mixed. Tank wastes sat for decades in environments of caustic chemicals and ionizing radiation. Researchers are determining the current chemical composition of the wastes and the physical stratification of waste components in the tanks.

Underground plumes present other challenges. Their boundaries are diffuse, the wastes exist in low concentrations, and physical and chemical complexes may have been formed with soil particles. Characterization of both tanks and plumes involves physical sampling, records analysis, and computer modeling.

Researchers are also determining strategies to protect future generations from the effects of Hanford wastes.

Vitrification of high-level waste has been selected as the best technology; construction of

a vitrification plant is about to start. At the same time, research is continuing to determine the effect of waste types and concentrations on the durability of the glass.

A variety of schemes have been developed for underground plumes. Volatile chemicals can be driven off by heating; organics may be converted by microorganisms; and radioisotopes can be vitrified in place, bonded to soil particles, or physically removed.

This research requires resources and expertise beyond that available at Hanford. A variety of collaborative arrangements with industry, academia, and the national laboratory system bring needed skills and knowledge to bear on Hanford waste remediation.

For example, between August 1988 and February 2002, Pacific Northwest National Laboratory placed 118 contracts for project work with Los Alamos National Laboratory; key words in those contracts include technetium, ferrocyanide explosive, plutonium glass and ceramics, tank waste samples, isotope measurements, plutonium sample analysis, and tank waste remediation. The total value of these contracts is more than $71 million.

In the early 1990s, Pacific Northwest Laboratory became one of the multiprogram laboratories in the DOE's national laboratory system. However, as with all name changes, it took a while for the paperwork to be completed. In 1995, the Laboratory officially added "National" to its name, becoming Pacific Northwest National Laboratory.

Vitrification is one method being used to clean up the legacy of waste brought about by forty years of plutonium production at Hanford. In vitrification, heavy metals and radioactive elements are chemically processed into a durable, leach-resistant glass. Vitrification technology has been under development at Pacific Northwest National Laboratory for more than twenty-five years. It has been applied to high-level radioactive waste and municipal solid waste. Waste glass can be formed into useful products such as shingles, rock wool insulation, aggregate, and clean fill.

The future

Plutonium production at Hanford contributed to victory in World War II and deterred World War III while the Soviet system bankrupted itself. The combined efforts of researchers at Hanford and those in the current DOE laboratory complex will ensure that the radioactive legacy at Hanford will be dealt with safely and permanently.

SAVANNAH RIVER SITE

This article was contributed by Ann Gibbs, Senior Fellow, SavannahRiver Site.

From production to cleanup

Fifty years of transuranic waste at Savannah River Site

Three years into the Cold War, in 1950, President Truman asked the E. I. du Pont de Nemours Company to build and operate a plant to produce materials for nuclear weapons. The design and construction several years earlier of the Hanford Works in Washington state (see article on page 13) had given the planners of this new mission a good idea of what would be required. The site had to have a large land area with a ready supply of water for the production of heavy water and cooling for the reactors, good transportation, a population sufficient to supply workers, and distance from major population centers.

After considering sites across the United States, the Atomic Energy Commission chose a location on the Savannah River near the towns of Augusta, Ga., and Aiken, S.C. The site had the necessary water supply, was in a semiarid region with a heavy clay soil that would be acceptable for waste storage for a relatively long period of time, was sparsely populated, and had a good train transportation network.

No task such as this can be accomplished without heartache—and the establishment of the Savannah River Plant (SRP) was no exception. Entire towns and farms had to be relocated to acquire the needed space of about 300 square miles.

A house is moved from its original site (circa 1951) to make way for construction of the Savannah River Plant. Empty fields are now filled with pine trees planted by the U.S. Forestry Service.

What a difference 50 years make

In 1951, houses that had been people's homes for generations had to be moved to make room for the Savannah River Plant (SRP), which was needed at the time to produce nuclear material for weapons. By 2000, that capability had been shut down for ten years. The cleanup of what is now called the Savannah River Site (SRS) involves shipping waste in special containers known as TRUPACT-IIs to the Waste Isolation Pilot Plant (WIPP) in New Mexico. Each shipment—including certification and licensing of the TRUPACT-IIs, certification costs for the waste, training for waste certification personnel and drivers, and security—costs more than did moving all the homes on SRP property in 1951.

The first shipment of transuranic waste from the Savannah River Site to the Waste Isolation Pilot Plant leaves the site in 2001.

While the approximately 1,500 families were compensated for their land and even family graves were moved, the dislocation ruptured the ties of generations. Production activities at Savannah River provided employment for many of the displaced residents. The same production activities generated waste, which will keep the displaced residents' descendants employed for years in decommissioning, demolition, surveillance, containment, and shipping activities.

The original production mission at SRP was plutonium. The mission was later enlarged to include production of tritium for hydrogen bombs, plutonium-238 for space applications, and other isotopes, such as californium-252, which were of interest for medical and other research.

These production activities required five nuclear reactors that used heavy water—water enriched with deuterium—as their coolant and moderator. The production activities also required two aqueous separations facilities to separate the plutonium from the uranium targets, a solid/gas separation facility to separate the tritium from the lithium targets, a fabrication plant for the production of the targets, a production area for heavy water, and miscellaneous support facilities such as laboratories, power stations, shops, and a waste disposal area.

Waste handling becomes a major mission

At the end of the Cold War in 1989, the Savannah River Plant became the Savannah River Site (SRS) under a new contractor: Westinghouse Electric Company. Waste handling became a larger part of the mission with the start-up of the Defense Waste Processing Facility (DWPF) in 1992.

The DWPF facility takes the high-activity liquid wastes containing actinides generated over the years at SRS and turns them into glass encased in stainless steel cylinders. The immobilized waste is currently stored at Savannah River in underground silos and is awaiting a final repository site.

The solid wastes have been disposed of or stored at a central point in the site that is more than five miles from any plant boundary. This "burial ground" is on a high point that has a clay soil which retards the migration of radionuclides (with the exception of tritium) to the

Transuranic waste is stored undercover among buried waste in the center of the site.

SAVANNAH RIVER SITE

One way researchers determine the transuranic content of a waste drum is with a passive-active neutron (PAN) instrument, developed at Los Alamos in the 1980s. In active mode, the instrument uses a generator like the one in this photo to produce 14 mega-electron-volt neutrons that bombard the waste to determine its transuranic content.

groundwater and off-site areas. The original seventy-six acres used from 1953 to 1972 will be permanently sealed in 2003 under a Resource Conservation and Recovery Act (RCRA)-approved closure cap. Additional acreage currently is used for disposal of low-level waste and concrete pads have been built in the acreage for storage of transuranic (TRU) waste drums.

TRU waste is defined as waste containing any of the radioactive nuclides above uranium in the periodic table that have a half-life of more than twenty years in a concentration greater than 100 nanocuries per gram of waste. Because of the potential hazards to the environment, SRS has always separated waste containing or suspected of containing these radionuclides.

While some of this waste has been buried and will be under the RCRA closure cap, the vast majority has been stored in drums on pads for future retrieval. The pads are covered with giant tent-like enclosures called RUBs (after the manufacturer) to protect the drums from rain.

Until recently, the quantity of waste containing transuranic nuclides was relatively low because the plutonium in the waste was recycled to meet production schedules. Nuclides above plutonium were even more valuable and were recycled, consequently producing little waste. Most of the waste at SRS contains either plutonium-238, plutonium-239, neptunium-237, or americium-241.

Because there were no field-measurement instruments capable of determining low concentrations of transuranic radionuclides in 1972, all waste originating in operations areas containing transuranic nuclides was stored. SRS now finds that many of the containers do not meet the definition of TRU waste because they have concentrations of less than 100 nanocuries per gram. These containers should be disposed of as low-level waste, but they suffer from another consequence of too much conservatism—they are noted as containing "solvent" rags.

The "solvent rag" designation was applied indiscriminately to all containers in storage when there was an interpretation by Region 4 EPA in 1990 that any waste containing materials that had been exposed to solvents was hazardous. The time needed to make the case for not applying the designation exceeded the budget available and the probable consequences were far in the future, so the designation was made.

Waste-drum inspection

In the mid-1990s, SRS began a vent and purge program to insert filter vents on all of its unvented stored drums. At the same time, programs for sorting the post-1990 drums into low-level and TRU waste, and retrieval of drums stored in earthen berms, began.

These drums of waste are now being analyzed and characterized for shipment to the Waste Isolation Pilot Plant (WIPP) near Carlsbad, N.M. Shipment to WIPP requires an Acceptable Knowledge (AK) document detailing how the waste was generated and then confirmation of the AK.

Several technologies are used to confirm the makeup of waste. Real-time radiography or digital radiography is performed on the containers to ensure that prohibited items such as aerosol cans or liquids are not present. After weighing, a nondestructive assay (NDA) is done to quantify the TRU waste present and its radionuclide distribution.

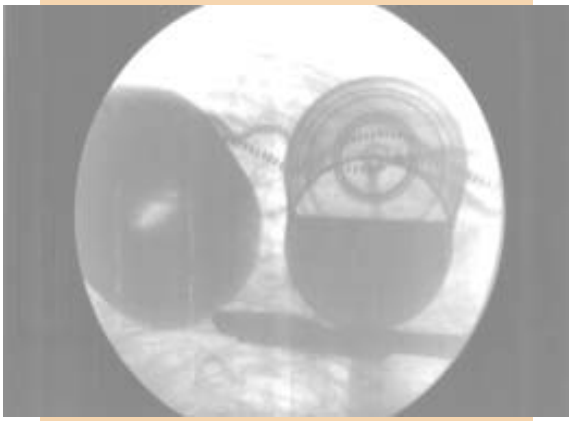
A gas analysis is also done on a sample of the headspace gas in each container to quantify and identify the gases to ensure that shipment in the sealed transportation container (TRUPACT-II) will not incur any risks. Researchers at SRS use a passive-active neutron (PAN) instrument developed at Los Alamos

in the mid-1980s for NDA analyses of low quantities of TRU waste. The PAN at SRS has a sensitivity of approximately 20 nanocuries per gram for 20 kilograms of waste.

In passive mode, the instrument detects spontaneous neutrons and neutrons produced by alpha interaction with fluoride or other light elements. In active mode, the instrument uses a neutron generator that emits 14-million-electron-volt neutrons to produce coincident neutrons from fissionable material such as plutonium-239. The active mode is the most sensitive and is subject to the most interferences. The radioactive nuclide listed in the Acceptable Knowledge document is confirmed by gamma analysis.

Savannah River's PAN response is reliable only up to approximately 1 gram of plutonium-239. Above this quantity, clumping, self-shielding, and other factors make the results questionable.

For amounts above 1 gram, SRS uses a segmented gamma scanner (SGS) to quantify plutonium-239 and obtain a distribution of the other TRU radionuclides. Reference materials used for checking the performance of the NDA instruments were made by Los Alamos and distributed through the WIPP program.



A radiograph of a transuranic waste container showing a roll of tape, protective clothing (you can see the zipper), an aerosol can containing liquid, and a pair of scissors. This container was rejected for shipment to the Waste Isolation Pilot Plant (WIPP) because of the aerosol can, which is pressurized and also contains liquid—both forbidden by WIPP.

The gas analyses are done by puncturing the carbon filter vents drawing off the headspace gases into evacuated containers. The container contents are then analyzed using gas chromatography-mass spectrometry. Reference materials are also distributed by WIPP for these analyses.

Subsequent testing of the drums may include visual examination of the contents to check the x-ray analyses. This is done in a glovebox where all the waste is dumped and sorted by hand while video cameras film the operation.

Savannah River's waste-certification analyses recently were taken over by the Carlsbad Central Certification Project (CCP) and the work contracted out to accelerate the TRU waste shipments from SRS to WIPP. Speeding up the shipments will allow waste from the Mound Laboratory in Ohio to be brought to SRS for interim storage, which in turn will accelerate the closure date of the Mound site. The contractors are using the same techniques as SRS did, but with newer models of equipment.

Information for this article was taken from "History of Du Pont at the Savannah River Plant," W. P. Bebbington, E. I. Du Pont De Nemours and Company, Wilmington, Del., 1990.

CMRR PROJECT

The main focus of the new facility's design will be to ensure that Los Alamos can meet and grow with the requirements of its major client, National Nuclear Security Administration (NNSA) Defense Programs—specifically pit production and enhanced surveillance activities.

A new facility to support chemistry and metallurgy research

DOE gives OK for conceptual design to begin on the CMR Replacement Project

After a year of developing plans and defining the requirements needed in a new chemistry and metallurgy research facility, the Laboratory has been given the go-ahead to begin conceptual design of the facility.

Energy Secretary Spencer Abraham in late July signed a memorandum for Critical Decision–Zero for the replacement of the 50-year-old Chemistry and Metallurgy Research (CMR) Building, which has a planned end-of life on or around 2010. Abraham's signing of the initial Critical Decision–Zero also authorized the Department of Energy (DOE) to begin preparing an Environmental Impact Statement and to hold public meetings on the CMR Replacement (CMRR) Project.

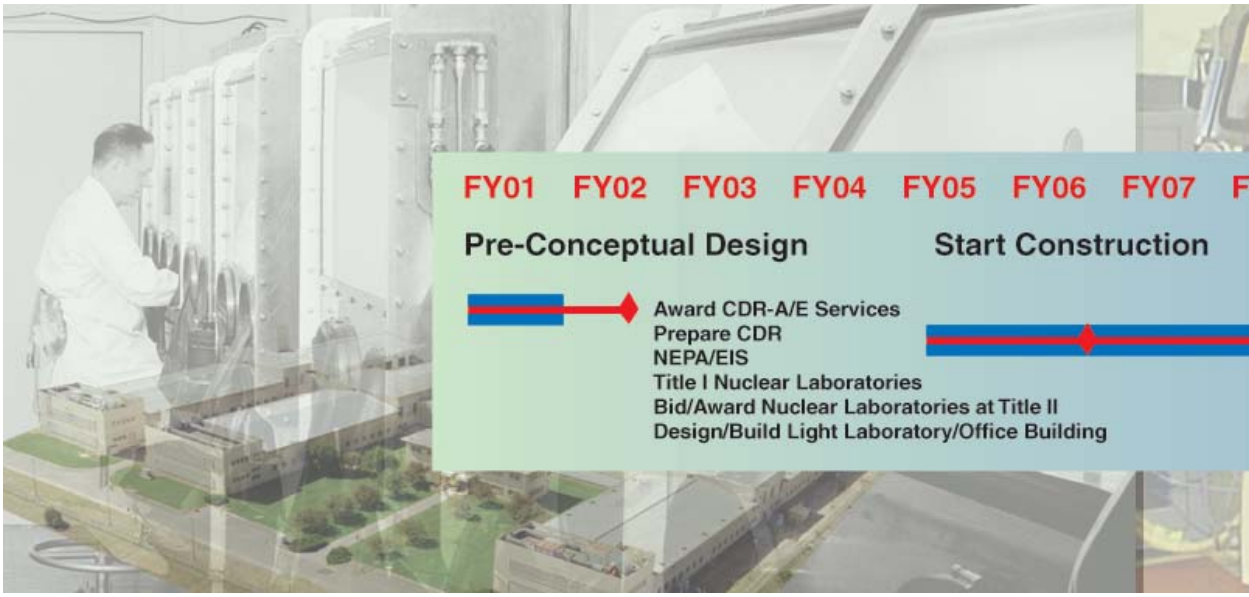
The main focus of the new facility's design will be to ensure that Los Alamos can meet and grow with the requirements of its major client, National Nuclear Security Administration (NNSA) Defense Programs. The CMRR project will provide the space needed for the analytical chemistry, materials characterization, and actinide research and development capabilities currently housed in the existing CMR that support the nuclear programs defined for Los Alamos in the Stockpile Stewardship and Management–Programmatic Environmental Impact Statement.

Research capabilities necessary for stockpile stewardship

The applied chemstry and metallurgical research performed at Los Alamos is crucial to the pit surveillance program, enhanced surveillance program, primary physics, and manufacturing projects. These projects require high-quality chemical and metallurgical analyses of a variety of plutonium metal and alloys to ascertain the effects of age, microstructure, and other potential variables on the stockpile.

Metallurgical research is performed using a wide variety of instrumentation, including electron microscopes, x-ray diffractometers, calorimeters, and a multitude of surface science equipment. The equipment used in the research and development activities in the CMR Building also acts as an enhancement to equipment and activities based at the TA-55 plutonium facility.

Moreover, the CMR facility houses key actinide science capabilities in analytical chemistry, processing and separations, solution chemistry, and spectroscopy. These capabilities support DOE programs in defense, nonproliferation and nuclear safeguards, counterproliferation, nuclear materials technologies, basic science, environmental stewardship, medical radioisotope, and technology development for waste treatment and minimization. The CMR



Nuclear Materials Technology/Los Alamos National Laboratory

facility also provides analytical reference standards for nationwide distribution.

The scale of studies could grow with a new facility. Because of the CMR operational limitations associated with the existing CMR aging, it has been downgraded to a Security Category 3 facility, which limits material compatibility studies. Certain areas of the new CMRR could be rated from "radiological facility"—the ability to work with up to 8.4 grams of plutonium-239 equivalents—to Security Category 1 and 2, where researchers can initiate more and larger-scale studies.

One of the current difficulties with supporting the nation's defense programs is a lack of trained personnel. DOE and NNSA officials and Los Alamos researchers hope that the new facility can be used to recruit and train actinide and nuclear workers to provide a pool of qualified candidates for defense activities throughout the DOE.

Integrated nuclear planning

The CMRR Project is integrated into the science-based stockpile stewardship program and site planning activities that are seeking relocation and consolidation of nuclear facilities at Los Alamos. This integrated nuclear planning activity is aimed at reducing costs and increasing efficiency.

Because the existing CMR and PF-4 aren't adjacent to each other and are not even located at the same site, operations are not as cost efficient as possible.

Relocation of the CMRR special nuclear material facilities to the preferred site at TA-55 could potentially save up to tens of millions of dollars each year by sharing safeguards and security efforts, eliminating equipment redundancies for operations performed now at both CMR and TA-55, and even more simply, by becoming more efficient in moving samples between facilities.

The old CMR Building is the largest at Los Alamos, covering more than a half million square feet. The proposed CMRR will be much smaller—less than 250,000 square feet. The design currently preferred is a three-building option that includes a light laboratory/office building outside the security fence at TA-55, which has radiological laboratories, and two nuclear facility buildings inside the security area at TA-55.

Several proposals are being considered for the old CMR Building. One of the alternatives being proposed in the CMRR Environmental Impact Statement is to decontaminate and decommission the entire CMR facility.

—Denise Sessions and Meredith S. Coonley

One of the problems with the nation's defense programs is a lack of trained personnel. DOE and NNSA officials and Los Alamos researchers hope that the new facility can be used to recruit and train actinide and nuclear workers to provide a pool of qualified candidates for defense activities throughout the DOE.

PLUTONIUM WORKSHOP

How well do we understand plutonium?

Researchers debate the issue at a collaborative workshop

photos by Mick Greenbank

"Plutonium is an element at odds with itself..."

As alluded to in Sig Hecker's opening presentation, the keynote of a recent collaborative workshop between Nuclear Materials Technology (NMT) and Theoretical (T) divisions was the need to understand the processing-structure-properties relationships for plutonium, its alloys, and its compounds.

Because the ability to perform underground testing gave researchers a "shortcut from processing to performance," they never really developed a good picture of material microstructure, "flying half-blind because we could test," according to the former Laboratory director.

With the current mandate for stockpile stewardship without underground testing, the microstructure of the different allotropes and alloys of plutonium has become an issue of major importance, and the workshop provided a diversity of perspectives on that broad topic.

The complexity of the issue underscored the irony that an element whose nuclear properties make it so attractive for both domestic and defense-related energy production has—as the "swing element" in the actinide series—physico-chemical properties that render it an enigma to empiricists and theoreticians alike.

In turn, this lack of clarity doubly underscores the crucial need for the exchange of information, for collaboration—and therefore, for such a workshop.

The workshop was organized by Shao-Ping Chen of the Theoretical Division Office (T-DO) and Jeremy Mitchell of Nuclear Materials Science (NMT-16), who also served as morning and afternoon session chairs.

Presenters came from T, Materials Science and Technology (MST), NMT, and Applied Physics (X) divisions, and from the Los Alamos Neutron Science Center (LANSCE), and there was enough interest in the morning unclassified session to fill Materials Science Laboratory (MSL) Auditorium beyond capacity, necessitating the setup of a closed-circuit feed to a room across the hall.

The afternoon classified session attracted at least as much interest and was relocated to the Administration Building auditorium to accommodate what one presenter referred to as a group containing a significant number of "fresh faces." Those new faces were one indicator of the breadth of Laboratory interest in this intriguing area, another reason why the sharing of research information is so vital to genuine progress.

The workshop's organizers were pleased with the variety of approaches in the talks, and likewise, with the diversity of presenter backgrounds. They felt that this combination helped strike a good balance in the range of topics, although they also admitted to being unable to accommodate all potential presentations because of the high response rate and limited time availability—the aim being to keep the workshop only a day in duration.

And despite the fact that all presenters ad-

hered to the stringent time limits effectively and diplomatically enforced by the session chairs, discussion time was at a premium. Both the organizers and the presenters concurred that more time for informal discussion and questions would have been desirable.

The electronic structure of plutonium seemed to crop up in just about every talk, frequently as a central issue. The key question, whether the metal's 5f electrons were best described as "localized" or "itinerant" (delocalized), was broached in presentations as diverse as Roland Schulze's plasmon resonance spectroscopy and John Wills's electronic structure calculations in the context of Density Functional Theory.

While there was a modicum of agreement that s, p, and d electrons might best be seen as delocalized with f electrons viewed as localized, one sensed more skepticism than complacency. John Joyce introduced a related issue in raising the possibility of f-electron–conduction-band electron hybridization.

Another important topic was what one presenter termed "phase diagramania." With the largest number of allotropes known for any element, phase diagrams for plutonium—and particularly those for plutonium-gallium alloys—bear some resemblance to a white rabbit's maze.

Marius Stan reexamined the controversial question of a eutectoid point in the Russian version of that latter phase diagram, and several other presenters, including Michael Baskes, Andrew Lawson, and Frank Cherne, discussed relevant aspects of plutonium-gallium interactions.

Crystal-structure quandaries were also in evidence, as presenters considered possible space-group cells for plutonium, the surface chemistry of its hydrides and oxides, and the crucial stockpile-aging issue of structural disordering resulting from helium-cluster formation, a consequence of plutonium's self-irradiation during radioactive decay. Nor did this workshop lack an out-of-the-ordinary component. While simultaneously characterizing the finding as "rather odd," John Sarrao presented what he described as "as good as it gets" evidence for a plutonium-based superconductor (see story on page 1).

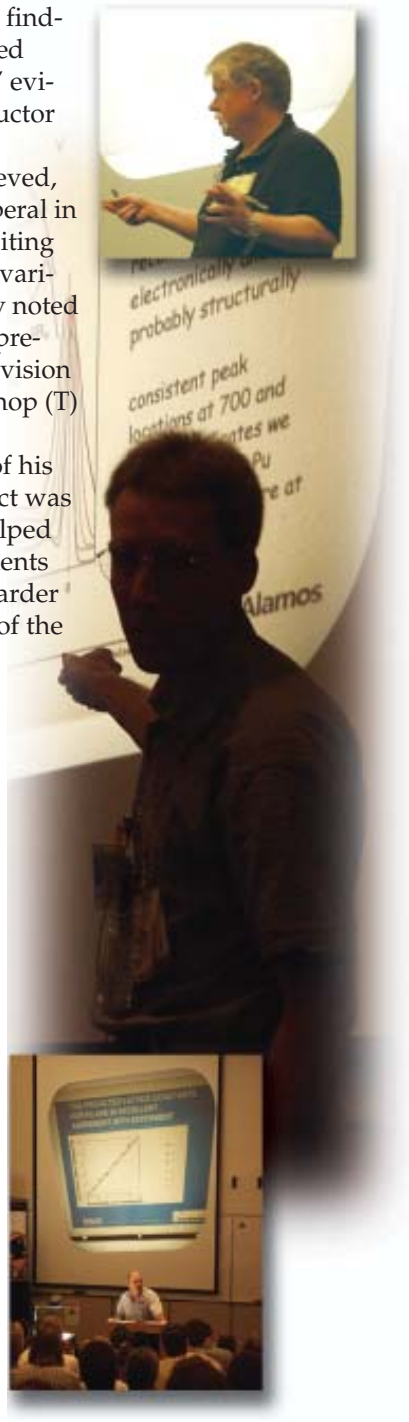
Reflecting on what the workshop achieved, co-organizers Chen and Mitchell were liberal in spreading praise for its success, first crediting the many volunteers who assisted in the various aspects of arranging the details. They noted an openness to collaboration among the presenters and were especially grateful to division leaders Tim George (NMT) and Alan Bishop (T) for fostering that collaborative mindset.

According to theoretician Chen, one of his incentives for proceeding with the project was his six-month stay in NMT-16, which helped him appreciate that conducting experiments on the idiosyncratic metal was "much harder than I thought," particularly in the face of the many regulations that make "good data precious."

But good data becomes even more useful when subjected to constructive scrutiny. And although other forums for plutonium-related discussions certainly exist (NMT seminars, the Plutonium Futures conference, and Seaborg Institute seminars, for example), Chen and Mitchell envision the possibility of smaller, more-focused mini-workshops on topics that emerged from this more diverse meeting, perhaps even as frequently as on a monthly basis.

For those at Los Alamos working in the field as well as for those who are merely intrigued by the possibilities implicit in plutonium's physics, chemistry, and metallurgy, such a possibility is welcome news. Meanwhile, for anyone who was unable to attend this workshop, the proceedings will soon be available on videotape.

—Vin LoPresti



Newsmakers and Notes

Lab hosts second plutonium metal standards exchange workshop

Researchers from six national laboratories and the Atomic Weapons Establishment, Aldermaston, U.K., met in Los Alamos Sept. 10 and 11 for the 2nd Annual Plutonium Metal Standards Exchange Workshop. Participants met with members of the design and production agencies for pit manufacturing to discuss current analytical chemistry results, comparison of current results with Rocky Flats standards, and other issues related to the Plutonium Metal Standards Exchange program.

Los Alamos reestablished the Plutonium Metal Standards Exchange program two years ago. It is a continuation of the Rocky Flats Plutonium Metal Sample Exchange program, which was conducted to ensure the quality and comparability of measurements such as plutonium assay, plutonium isotopics, and impurity analyses. The Rocky Flats program was discontinued in 1989 after more than 30 years.

The purpose of reestablishing the program is to provide participating facilities a way to independently verify their analytical measurement capabilities and to identify problems. At Los Alamos, the focus is pit production and certification measurements.

Under the program, Los Alamos' Actinide Analytical Chemistry (C-AAC) research and development team, in collaboration with Nuclear Materials Management (NMT-4) and Pit Disposition Science and Technology (NMT-15), prepare and distribute plutonium metal samples to various sites.

The samples are then used primarily for destructive measurements to determine elemental concentration, isotopic abundance, and metallic and nonmetallic impurity levels. This data is then statistically evaluated by the C-AAC quality assurance team and Statistical Sciences (D-1) and a report is released semiannually.

Argonne National Laboratories East and West, Lawrence Livermore National Laboratory, New Brunswick Laboratory, Savannah River Site, and AWE are participating in the exchange program along with Los Alamos.

Reestablishing an overall interlaboratory measurement evaluation program has involved the efforts of many Los Alamos organizations. Key support has come from Distributed Finance (BUS-2); C-AAC; the Chemistry Division Office (C-DO); D-1; Weapons System Engineering (ESA-WSE); the Nuclear Materials Technology Division Office (NMT-DO); NMT-4; NMT-15; and Nuclear Materials Science (NMT-16).

For more information on the plutonium metal exchange program or the workshop, contact Lav Tandon (C-AAC) at (505) 665-5458 or tandon@lanl.gov.

Lab team wins White House award

A team from Actinide Process Chemistry (NMT-2) has won a 2002 White House Closing the Circle Award for a unique way to eliminate acid waste at the Plutonium Facility at TA-55. Aquilino Valdez, Ronald Chavez, Benjie T. Martinez, and Don Mullins accepted the award in the recycling category for their Nitric Acid Recovery System. The technology also won a Department of Energy Pollution Prevention Award.

The Nitric Acid Recovery System reconcentrates nitric acid used to purify plutonium at TA-55 and separates a stream of 99.98 percent pure water, with no measurable plutonium. The system also reduces the nitric acid used in processing operations to about 20 percent of the historic usage.

Editor's note

The lead article in our last issue, "Researchers cast first 'spiked' plutonium alloy," reported a major success in replicating how the stockpile ages. We failed to note a historical perspective of radiation-induced aging studies of materials using plutonium 238. The idea of doping materials with plutonium-238 for accelerated aging effects was proposed by Rodney Ewing at Pacific Northwest National Laboratory in the late 1970s and the plutonium 238-doped zircon was prepared during the 1980s at Battelle. The resulting materials were extensively studied by Ewing and other researchers.

Chief scientist says goodbye

In our first-year anniversary issue of *Actinide Research Quarterly*, winter 1996, I said "A newsletter is in some sense like a living plant. A plant is constantly absorbing essential elements to synthesize nutrients for its growth, constantly trying to adapt to its changing environment, and constantly in need of nurturing ... "

In our sixth-year anniversary issue, winter 2001, I said "In the end, every human endeavor is a learning process; publishing this newsletter is no exception. We draw immense satisfaction knowing that our readers and the publication team members have journeyed together the past six years on this learning path."

Indeed I have learned a lot and I have enjoyed tremendously seeing the growth of this publication for the past eight years. Now, it is time for me to say farewell to all our readers and move onto another phase of my career. The publication will continue with the remaining dedicated team members under the aegis of the Nuclear Materials Technology Division and Los Alamos National Laboratory. I hope all our present and future readers will support this newsletter with your continued input and encouragement.

—K.C. Kim

Editor's note: The *Actinide Research Quarterly* publications team welcomes its new scientific advisors: David Clark, Gordon Jarvinen, and Web Keogh. All three are members of the Laboratory's Seaborg Institute for Transactinium Science.