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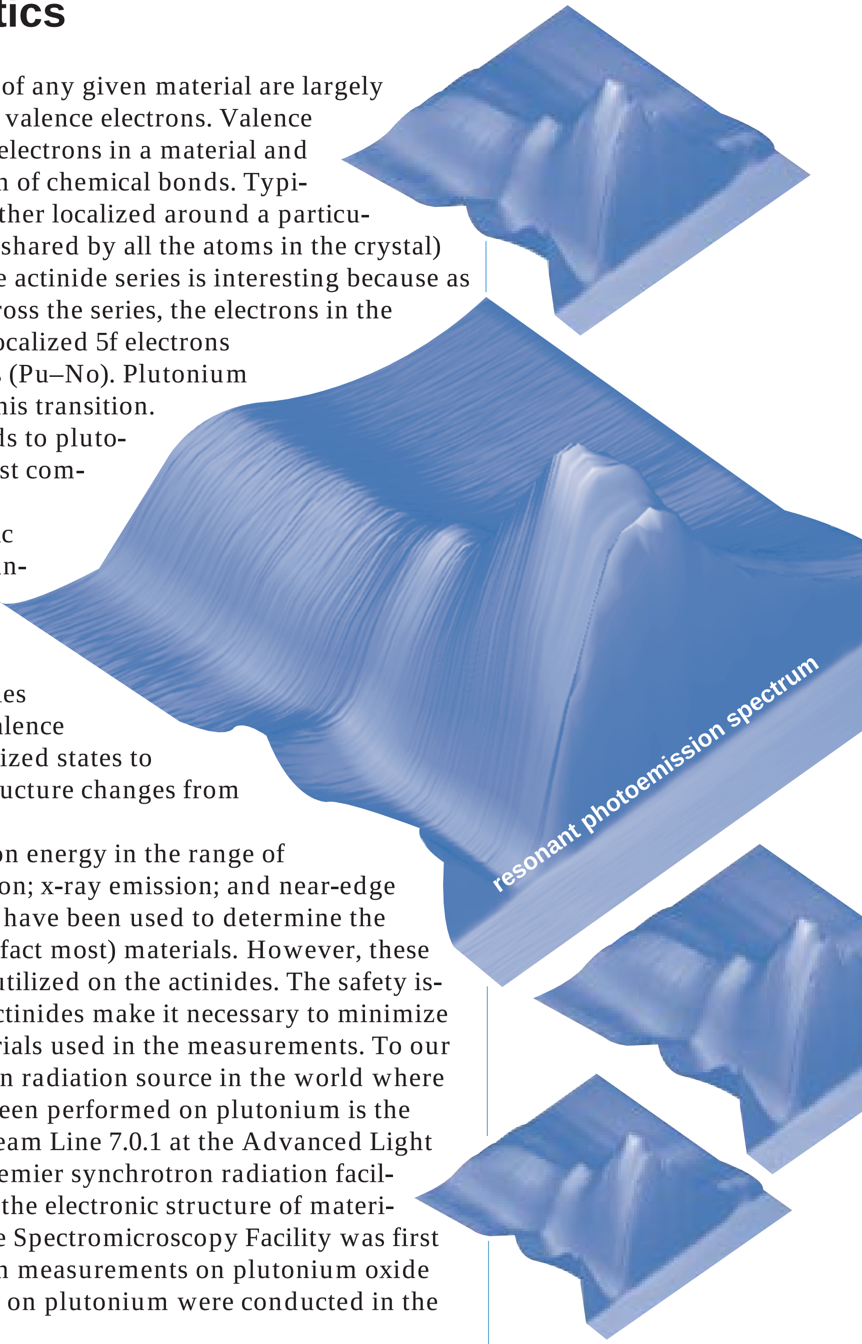
In This Issue

Photoemission Studies at the Advanced Light Source Shed Light on Plutonium Phase Characteristics

The physical characteristics of any given material are largely derived from the behavior of its valence electrons. Valence electrons are the lowest-energy electrons in a material and are responsible for the formation of chemical bonds. Typically, in a metal, electrons are either localized around a particular atom or are delocalized (i.e., shared by all the atoms in the crystal) throughout the entire metal. The actinide series is interesting because as the atomic number increases across the series, the electrons in the actinide metals transit from delocalized 5f electrons (Ac–Pu) to localized 5f electrons (Pu–No). Plutonium (element 94) is located right at this transition. This placement in the series leads to plutonium metal being one of the most complex materials known. Metallic plutonium displays six allotropic phases (α, β, γ, δ, δ′, and ε) at standard pressure. A 20% volume expansion occurs during the change from the α phase to the δ phase. These physical properties have been attributed to the 5f valence electrons changing from delocalized states to localized states as the crystal structure changes from the α to the δ phase.

Soft x-ray techniques (photon energy in the range of 10–1000 eV) such as photoelectron; x-ray emission; and near-edge x-ray absorption spectroscopies have been used to determine the electronic structure of many (in fact most) materials. However, these techniques have not been fully utilized on the actinides. The safety issues involved in handling the actinides make it necessary to minimize the amount of radioactive materials used in the measurements. To our knowledge, the only synchrotron radiation source in the world where soft x-ray measurements have been performed on plutonium is the Spectromicroscopy Facility at Beam Line 7.0.1 at the Advanced Light Source (ALS). The ALS is the premier synchrotron radiation facility in the world for the study of the electronic structure of materials using soft x-ray photons. The Spectromicroscopy Facility was first used for resonant photoemission measurements on plutonium oxide in 1993. Our first measurements on plutonium were conducted in the fall of 1998.



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Photoemission Studies at the Advanced Light Source Shed Light on Plutonium Phase Characteristics *(continued)*

This article was contributed by **Jeff Terry** and **Roland Schulze** (both NMT-11). Others contributing to the research were **Jason Lashley** (MST Division), **Tom Zocco** (NMT-11), **J. Doug Farr** (NMT-11), **Eli Rotenberg, Keith Heinzelman, and David Shuh** (Lawrence Berkeley Laboratory), and **Mike Blau** and **Jim Tobin** (Lawrence Livermore National Laboratory).

The Spectromicroscopy Facility is designed so that measurements can be made on small quantities of hazardous material. This facility has a photon flux of 10^{13} photon/sec at a photon energy of 100 eV with 0.01 eV resolution. The high photon flux allows one to focus the beam down to a size of 50 microns and still have enough light intensity at the sample for measurements to be conducted in a reasonable time frame, 1–10 minutes per spectrum. Therefore, the sample size can be on the order of 100 microns in diameter. This greatly minimizes the amount of plutonium on site during the experiment. The microfocussing capability also allowed us to focus onto a single δ-plutonium microcrystallite grown by Jason Lashley (MST Division).

We performed core-level photoemission, valence band photoemission, and near-edge x-ray absorption spectroscopy on both polycrystalline α-plutonium and δ-plutonium microcrystals. Only the photoemission experiments will be described here. Photoelectron spectroscopy is predicated upon the photoelectric effect first described by Einstein in 1905. The experimental photoemission setup at the Spectromicroscopy Facility is shown in Figure 1. An incident photon is absorbed by an atom in the solid, and an electron is ejected. An electron energy analyzer is used to measure the direction and kinetic energy of the emitted electron. The kinetic energy of the photoelectron is directly related to its binding energy in the solid.

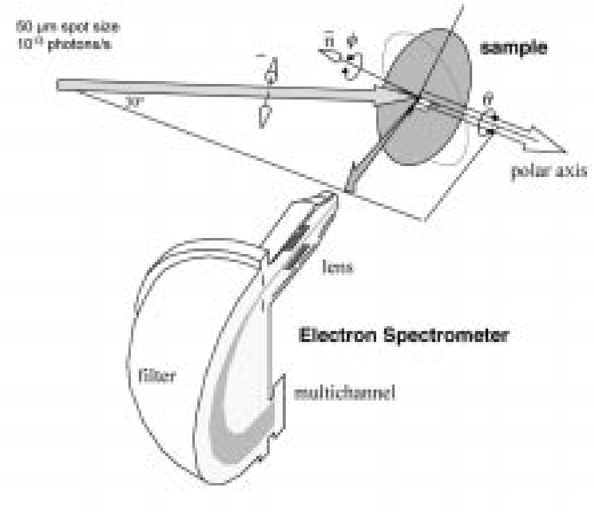


Figure 1. The photoemission setup at the Spectromicroscopy Facility at ALS. The position of the analyzer is fixed with respect to the incoming photons. Angle-resolved photoemission is performed by rotating the sample.

Figure 2 shows the Pu 4f core-level photoemission spectra from the α- and δ-plutonium samples. The spectrum arising from core-level photoemission is sensitive to energy differences in the initial state (no core hole) and the final state (core hole and free photoelectron). The two core-level spectra in Figure 2 are similar in that they both show two features, a sharp feature at low binding energy and a broad feature at higher binding energy. The low-binding-energy feature can be ascribed to a metallic initial state with a delocalized final state. As the 5f electrons are more delocalized in the α- than in the δ-plutonium, the greater number of delocalized electrons in the α-plutonium leads to greater intensity in this peak than the δ-plutonium.

Presently, the higher-binding-energy feature is not completely understood. It is likely that this feature has two states contributing to it. We believe that, in simplistic forms, one possible component contributing to the feature is made up of emission to an electronic final state with localized 5f electrons. If this is the case, it suggests that both localized and delocalized 5f electrons are present in the α- and δ-plutonium phases. The second possible component of this feature is from an initial state that has been oxidized.

Valence band spectra show that a small amount of oxygen remains on the surface after the sample preparation. The binding energy of PuO_x is higher than that of the metal and would be expected to be seen around the position of the unknown high-binding-energy feature. A final determination of the exact nature of this high-binding-energy feature will shed light on the poorly understood valence electronic structure of plutonium, as well as on the presence of localized and delocalized 5f electrons in the different plutonium phases.

Our initial goal of the valence electronic structure measurements was to determine the density-of-state of the 5f valence electrons in α- and δ-plutonium. This experiment can be directly compared with electronic structure calculations performed by theoreticians. Figures 3(a and b) show the resonant photoemission spectra of the valence band from δ- and

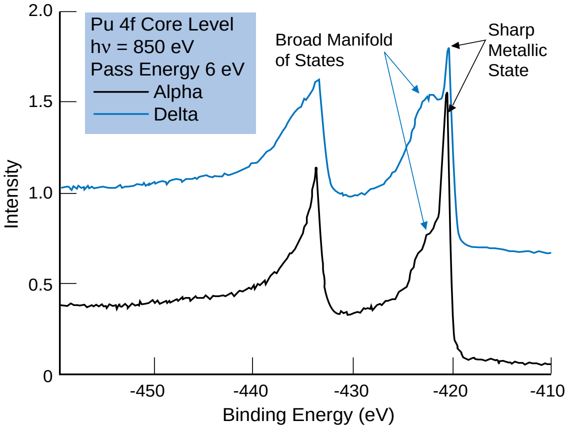


Figure 2. Core-level photoemission spectra from a large crystallite δ-plutonium sample and a polycrystalline α-plutonium sample are shown. These spectra were collected with a photon energy of 850 eV and an analyzer pass energy of 6 eV. Note that two large components are visible in each spectrum, a sharp feature at low binding energy and a broad feature at higher binding energy.

α-plutonium, respectively. In a resonant photoemission experiment, valence band photoemission spectra are collected as the photon energy is scanned through a core-level absorption edge. This can result in an enhancement of the emission from specific valence levels. In the case of plutonium, scanning the photon energy through the 5d absorption edge results in a resonant enhancement of the 5f valence emission. The 5d–5f resonant photoemission measurements in plutonium are a measurement of the 5f contribution to the valence density of states.

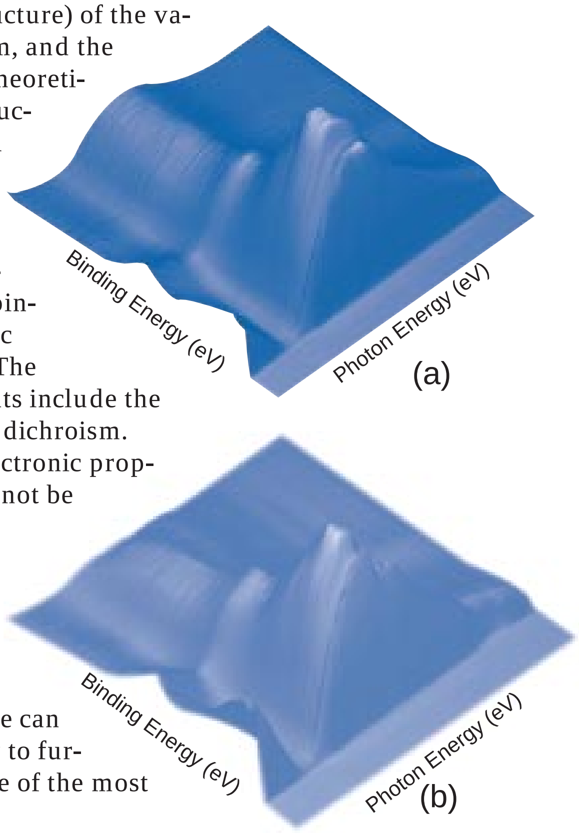
Two types of information are obtained in the resonant photoemission measurements that can greatly enhance the understanding of plutonium metal. Two-dimensional data slices can be taken in either the constant-photon-energy direction or the constant-binding-energy direction. Slices taken with a constant photon energy are the equivalent to standard photoemission spectra with the exception that specific valence states are emphasized. Different states turn on or become enhanced at different photon energies.

The data in Figures 3(a and b) show clear differences in the resonant photoemission spectra from the α- and δ-plutonium. This is most evident in the state at binding energy 0 eV. If this peak is followed as the photon energy is varied, one notices that in the δ-plutonium (Figure 3a), after 100 eV, the peak smoothly increased to a maximum and then smoothly decreased after the maximum was reached. In direct contrast, the α-plutonium (Figure 3b) showed oscillatory behavior after the initial maximum was reached. Theoretical calculations of the resonant photoemission are currently ongoing to try to understand these differences.

A large theoretical effort has been undertaken to understand the plutonium data that we have collected. The next phase of the valence-band, electronic structure measurements will involve measuring the electron dispersion relation (band structure) of the valence electrons in δ-plutonium, and the data will be compared with theoretical calculations. The band structure can only be measured on a single crystal of a material, and it is imperative to have excellent crystals for the measurement. Future investigations will be based upon spin-resolving and photon-dichroic photoelectron spectroscopy. The photon-dichroic measurements include the variant magnetic x-ray linear dichroism.

The unique 5f valence electronic properties of plutonium metal cannot be explained by the typical one-electron calculations used to describe prototypical metals. Calculations on plutonium metal incorporate these properties into calculations in an ad hoc manner. We can measure these effects directly to further our understanding of one of the most complex materials known.

Figure 3. The 5d–5f resonant photoemission spectrum with analyzer pass energy of 1 eV showing the 5f density of states in the valence band from (a) a large crystallite δ-plutonium sample, and (b) a polycrystalline α-plutonium sample.



O_2F in FOOF Holds Promise for Pu Decontamination

This article was contributed by **Kyu C. Kim,** NMT Chief Scientist.

The traditional method of recovering uranium and plutonium from process streams is to dissolve actinide-containing residues and wastes in strong acidic media and put them through a series of separation processes such as precipitation, filtration, and ion exchange. Over the half century of nuclear materials processing activities, this method has produced voluminous nuclear wastes stored at various nuclear sites. The search for safe disposal methods continues to this day. From a processing efficiency standpoint, a better way of separating actinides from their major constituents is to act upon only the minor components without dissolving the entire matrices. It would be ideal if a gaseous reactant could be flowed through the residue matrices, and only the volatile actinides recovered as gaseous products, leaving behind the actinide-depleted residues. This article describes identification and analysis of an exotic chemical that holds a great promise in actinide recovery, decontamination of process equipment, and waste reduction in actinide processing.

In the early 1980s the Los Alamos laser isotope separation program for plutonium had completed the scientific concept demonstration and moved toward a small-scale process demonstration. This program was called molecular laser isotope separation (MLIS) and code-named Lippizan. Before this we had an uranium isotope separation program code-named Jumper. Our sister laboratory at Livermore had two similar programs; their process was called atomic vapor laser isotope separation.

An MLIS process uses stable molecules that bear the isotope of interest in their molecular structures. Both uranium and plutonium form stable gaseous molecules called hexafluorides. Six fluorine atoms occupy the six apexes in an octahedral geometry with a uranium or plutonium atom in the center. This particular soccer-ball-like geometry accounts for the chemical stability as a gaseous molecule. Most of the world's supply of separated uranium isotope ^{235}U comes largely from a gaseous diffusion process employing uranium hexafluoride as the working medium. Plutonium hexafluoride is the only known stable gaseous molecule at room temperature, so it is not surprising that several isotope separation schemes were devised in the early years using this molecule.

Once produced, plutonium hexafluoride is fairly stable under process conditions such as circulation, compression, and expansion, inside the process pipes. However, it is not so easy to make plutonium hexafluoride under mild chemical conditions. This problem is further complicated when the MLIS end product is solid particles, and plutonium hexafluoride also decomposes slowly to a solid product that coats the inside wall of the process pipes as the process continues. So we needed a way or ways to recover the solid product from the process line. The only way to accomplish this recovery was then to turn the solid product back to gaseous product in situ. We were searching for chemicals to accomplish the product recovery and cleanup of the process line.

By the early to mid 1980s we had identified a few chemicals that could be used for the purpose. Krypton difluoride (KrF_2), dioxygen difluoride (FOOF), and the fluorine atom were the most promising among all we investigated. Krypton and oxygen in these molecules are merely carriers of F atoms. In the case of FOOF we discovered subsequently that the active species involved in the actual fluorination reaction was a radical species—O_2F—derivable from FOOF. This story is about the discovery of O_2F and characterization of some of its properties. A radical has unpaired electrons unlike a stable molecular electronic configuration so that it is usually very reactive with neighboring species, and it does not hang around long (less than several seconds at most) for us to detect readily in its isolated state.

The structure of FOOF had been studied previously by microwave spectroscopy by other researchers. This compound is also very unstable. It can be stored indefinitely only at very low temperature, such as that of liquid nitrogen. The infrared spectrum of solid (frozen) FOOF at 77 K had been recorded, and all fundamental vibrations assigned also. But the spectral quality was very poor because these measurements had been made on a minute quantity of impure samples. A serious obstacle to the study of FOOF in the gas phase had been the difficulty in obtaining sufficient amounts of the pure compound and preserving it for a sufficient length of time for spectroscopic investigation.

In late 1984 we recorded for the first time a complete infrared spectrum of gaseous FOOF obtained under well-defined conditions using a special spectrometer that we had developed at Los Alamos. For the first time we were able to make accurate vibrational frequency assignments and determine their intensities.

Immediately after recording the first complete spectrum of FOOF, we noticed an unexplainable spectral feature centered at 1490 cm^{-1} (See Figure 1.) This spectral feature did not vary in its relative intensity with the rest of the peaks. Furthermore, it is the only feature in the entire fundamental infrared region apart from those of FOOF. The molecular vibrational theory tells us that whatever species is responsible for this feature must be a compound simpler than FOOF or a heteronuclear, diatomic molecule. We did not consider the idea of a radical because of the attributes of radicals discussed above. The half-life of a radical species is typically so short that one cannot record its spectrum over many minutes as required for a spectral scan. And yet this unexplained feature persisted in all samples of FOOF. We also eliminated early on the possibility that it might be the diatomic radical OF because molecules such as FOOF do not break apart at their stronger O-O bonds spontaneously under these experimental conditions.

Within the first week of successfully recording the FOOF spectrum, we had to speculate that we might have an unusual radical species—O_2F—coexisting with FOOF in our sample. How then could we prove our hypothesis that a reactive radical species like O_2F coexists with another unstable molecule—FOOF? To answer this question unambiguously we devised an experiment to produce O_2F directly and record its spectrum in the absence of FOOF. Now, this experimental design is based on the assumption that the half-life of O_2F is sufficiently long (for example, seconds) so that we could probe continually while we produced this short-lived species in an experimental design, described below.

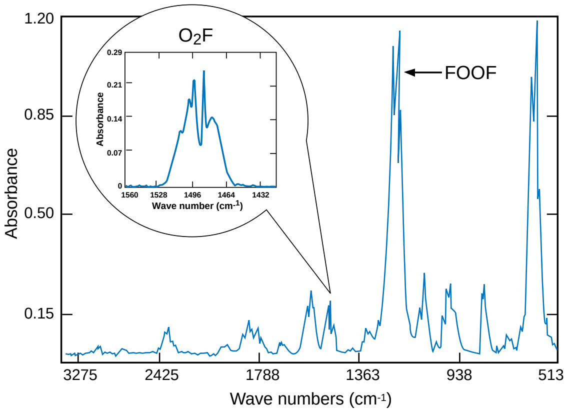


Figure 1. The first complete infrared spectrum of gaseous FOOF. The laser produced O2F spectrum is shown in the inset.

The experimental design thus conceived is laser flash photolysis combined with coaxial probing. A strong laser beam produces F atoms along its beam path, the F atoms react with an oxygen molecule to form the transient radical O_2F, and its presence is detected simultaneously by probing the sample volume within the laser beam path. Although easily described, this arrangement requires that the laser beam and the probe beam should be made co-linear, and the probe beam should be inside the laser beam volume for maximum sensitivity. We would make a mixture of two basic ingredients, fluorine and oxygen, which we considered as the starting materials. An intense ultraviolet laser beam at 248 nm was injected into the sample chamber that contained the mixture. The laser beam was pulsed at less than 10 Hz. The probe infrared beam from the spectrometer was combined using a dichroic mirror and made co-linear along the entire beam path. Our hope was that as the laser beam produced the expected radical species at some low-frequency interval, a steady-state concentration of O_2F would be established between the production and disappearance, and the probe infrared beam would be monitoring this steady-state concentration continually. (A dichroic optical material reflects and transmits different wavelength lights so that two beams of different wavelengths can be combined coaxially and propagated together along the same beam path.)

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Editorial

Nuclear Materials Including "Orphans" Require a Comprehensive Management Strategy

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"Nuclear materials" is a generic term meaning materials of interest to the nuclear industry, sometimes synonymous with "source" and "special materials," as listed in the *Atomic Energy Commission Manual.* A vast knowledge base has been generated over the past six decades to safely manage all nuclear materials to maximize their benefits and minimize their hazards. However, it should be recognized that the knowledge base was not always adequate nor effectively used in the preservation and use of these materials. As a result, many environments have been impacted by nuclear materials, and the potential for impacting more still exists. Therefore, it is important to develop a *comprehensive* strategy to better manage these valuable resources for the future.

A recent estimate by the Brookings Institution points out that the United States spent over $5.5 trillion during the past five decades to build and maintain nuclear defense capabilities, almost $1 trillion of this in nuclear materials production and related activities. Under a series of Congressional initiatives, the DOE has the major responsibilities for long-term management of all nuclear materials from the defense sector and a major portion of those originating from the civilian sector. Currently, numerous programs address the long-term management of materials for national security (including those resulting from weapons dismantlement, nuclear wastes, spent nuclear fuels, and contaminated sites and facilities). These programs are at various stages of development, demonstration, and implementation. Our ability to achieve the goals of our excess fissile material disposition programs is dependent on complex international agreements and is likely to take many decades.

A large inventory of nuclear materials originating within and outside of national security programs exists in addition to those necessary for national security or declared excess. These may be properly described as "orphan nuclear materials." The disposition of orphan nuclear materials is still awaiting recognition and resource allocations. A preliminary but incomplete estimate made last year identified over 30,000 records of 72 million curies of miscellaneous radioactive materials. The actual numbers may be many times those identified last year.

A unique example of orphan nuclear materials is the man-made neutron sources that are used extensively in defense projects, industries, universities, and research organizations. Most of these neutron sources consist of homogeneous mixtures of an alpha- or gamma-emitting radionuclide and a low-atomic-number element or its compound. It is estimated that there are several hundred thousand such sources now in use within the U.S., and about 3,000 are being produced and distributed annually (Figure 1) . A 1985 Congressional mandate makes the DOE responsible for receiving abandoned, damaged, and re-

Figure 1. Scott Allen and George Powell (BUS-4) may be called upon to recover and transport neutron sources widely used in oil and gas well logging. Nuclear material "orphans" such as these sources pose a problem for the managed disposal of excess nuclear materials.

turned neutron sources and disposing of them in an environmentally benign fashion. Inappropriate disposition of these sources has the potential to cause extreme hazard to human beings and irreparable harm to the environment (Figure 2). Los Alamos National Laboratory spent many years to develop a simple, elegant method (chemical separation flow sheet) to separate the two components of the neutron sources. This technology was successfully demonstrated in dismantling over a thousand neutron sources and making them innocuous and environmentally benign. Unfortunately, the implementation of this elegant technology to address large-scale disposition of neutron sources is now in doubt.

Present challenges to nuclear materials management are too numerous for extensive discussion here. There is, however, a tacit recognition that a common strategic framework is necessary to avoid the environmental blunders of the past. During the past decade, a series of studies and assessments have been performed to identify and quantify the needs of nuclear materials management.

On January 20, 1998, the DOE Office of the Deputy Assistant Secretary for Nuclear Material and Facility Stabilization initiated the Nuclear Material Integration (NMI) Project. The goals of the NMI were to inventory and analyze the nuclear materials in the DOE Complex. The scope of this project included not only materials owned by the Office of Environmental Management (EM) but also those owned by other programs and stored in EM facilities. The ultimate goal of this effort was to develop a comprehensive nuclear material management plan for the nation.

Three material management teams were responsible for the different groups of materials in the DOE Complex: the Transuranic Team, responsible for most transuranic elements; the Uranium / Thorium Team, responsible for most uranium and thorium materials; and the Nonactinide Isotope and Sealed Sources Team, responsible for all radioactive isotopes with an atomic number less than 90, and all sealed sources, irrespective of atomic number. About 250 people from across the country participated in developing data and strategies for the management of all the "excess" nuclear materials within the DOE complex and those that are likely to become DOE's responsibility in the future. The three teams prepared a total of twelve independent reports (nuclear material management plans) and submitted them to DOE / EM in 1998. Separate materials management plans were prepared for depleted uranium, high-enriched uranium, low-enriched uranium, natural uranium, nonactinide isotopes, ^{241}Am, ^{237}NP and ^{238}Pu, ^{239}Pu, ^{242}Pu, thorium, transuranic heavy elements, and ^{233}U. These reports are presently considered "predecisional draft documents" and are not yet available for general distribution.

While many such studies have developed valuable data, heretofore, follow-up actions have been disappointing. Missing in previous follow-up actions is the desire to address the problem as a national or societal issue. Instead it is always addressed as the issue relevant to a particular agency of the government or a subordinate bureaucracy interested only in simplistic solutions. It is this approach that ought to change to achieve the goals of a *comprehensive* strategy to manage all nuclear materials for the benefit of mankind and to instill public confidence in associated technologies and institutions.

In the best traditions of U.S. science, an objective evaluation of the recommendations made by the NMI teams has been requested by an independent organization, the National Academy of Sciences. DOE / EM is championing the cause for an Academy study to review the 12 reports. However, this study is still waiting because of limited DOE resources. It is to be hoped that the Academy will evaluate the reports, and that their recommendations will include *comprehensive* management and preservation of nuclear materials, including neutron sources and other "orphans," that are generated by both the defense and civilian sectors. It is also to be hoped that such recommendations will be implemented in a timely manner.

Figure 2. A damaged americium-beryllium neutron source recovered from a private company by Los Alamos.

The opinions in this editorial are mine; they do not necessarily represent the opinions of Los Alamos National Laboratory, the University of California, the Department of Energy, or the U.S. Government.

Wing 9 Hot Cells Support Work Involving Highly Radioactive Materials

The Wing 9 hot cells in the CMR Facility were designed and constructed in 1960 to provide shielding and remote handling capabilities for work on highly radioactive materials. Commissioned in 1961 by President John F. Kennedy, the hot cells supported the Rover Project and the postmortem of irradiated fuels for breeder reactors. Even as the hot cells began decommissioning for closure in the late 1980s, they were used successfully to demonstrate the characterization and packaging of resident, remote-handled transuranic waste for eventual shipment to the Waste Isolation Pilot Plant (WIPP). This expertise continues to be a Wing 9 capability available to the industry and the DOE complex. Since 1989, a variety of projects have been identified that have extended the life of the facility and are discussed later in this article.

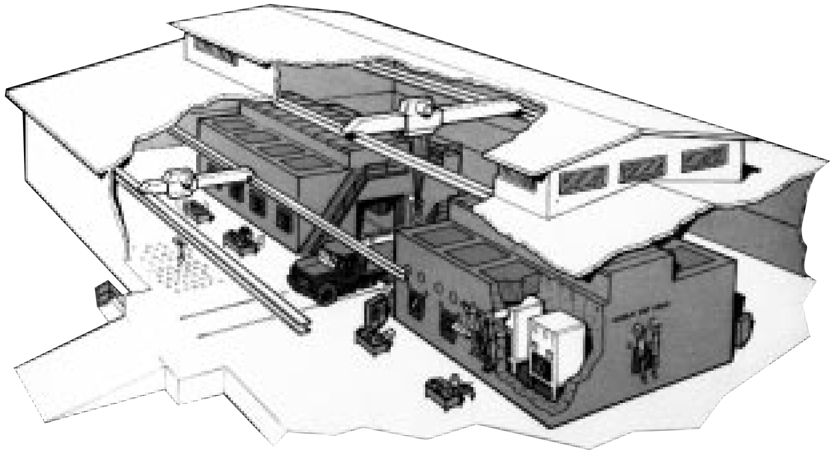

Figure 1. The hot cells in Wing 9 of the CMR Building can remotely handle and process all types of radioactive materials that are alpha, beta, or neutron emitters. Capabilities include characterization and packaging of WIPP waste as well as tasks that may require a room-sized, heavily shielded remote-handling facility for highly radioactive materials.

Each of the 16 6’x6’x11’ hot cells and the corridor that connects them are designed to provide adequate shielding to the worker from a 100,000 Ci source of 1-MeV gamma radiation (Figure 1). Also contained in the hot cells are in-cell remote machining and handling capabilities that allow operations on highly irradiated components. The inner-cell door design also provides a means of connecting two hot cells for installation of large pieces of equipment. The facility can handle all types of radioactive materials that are alpha, beta, gamma, and neutron emitters. An array of 364 heavily shielded storage wells and a crane capacity up to 25 tons support the hot cell capability. The current authorization basis allows a total inventory in the16 hot cells of up to 77 kg of ^{239}Pu equivalent.

Technical and programmatic objectives require the Hot Cell Team in Group NMT-11 to work on programs that encompass state-of-the-art remote handling of highly irradiated materials, components, and systems. A key component of this program includes effective facility management practices to maintain remote handling operations that are compliant with DOE regulations. The Wing 9 staff expertise is the key to success with capabilities such as design engineering, process analysis, project management, welding, fabrication, robotic operations, and handing and shipping of high-activity components. Additionally, the staff has experience with the fabrication of parts made from special nuclear materials and disassembly and analysis of highly irradiated components and materials. Currently, the Wing 9 team is providing expertise to other laboratories and universities for a variety of special remote handling problems. Following are descriptions of some current Wing 9 projects.

The Molybdenum-99 Project

In response to the U.S. medical community’s concern that the nation must rely on a single, limited, foreign source for the radioisotope molybdenum-99 (^{99}Mo), Congress has tasked the DOE to develop a domestic source of ^{99}Mo, which is essential for conducting thousands of life-saving medical diagnostic procedures each day. The ^{99}Mo program involves five primary activities: (1) target fabrication, (2) target irradiation, (3) extraction and purification, (4) transportation, and (5) waste management. The Isotope Production and Distribution branch of the DOE conducted assessments that identified the best ^{99}Mo production option currently available: the fabrication of uranium targets at Los Alamos National Laboratory and target irradiation, and extraction and purification processing at Sandia National Laboratories.

A target fabrication demonstration program has successfully been completed in Wing 9. A target is a 304 stainless steel tube approximately 18 inches long and 1.25 inches in outer diameter. Inside the tubing, ^{235}U is electroplated onto the steel surface to provide a very thin layer of uranium to interact with neutrons. The process takes 36 to 48 hours to deposit approximately 20 grams of uranium coating per target. The target tubes have caps welded at each end and are qualified as "reactor-ready" before transfer to Sandia for irradiation. The fabrication and electroplating process is shown in the glovebox enclosures in Figure 2.

Neptunium Project

The NMT-11 staff of Wing 9 has been asked to fabricate a clad, solid ~7 kg ^{237}Np metal sphere for criticality measurements at TA-18. The ^{237}Np first-daughter product, ^{233}Pa, is highly radioactive so the entire process must be performed within a shielded hot cell.

The necessary equipment to melt and cast the ^{237}Np metal remotely will be installed in a hot cell "alpha box" to process the alpha-emitting materials and to contain potentially high levels of contamination. The alpha box will be purged with nitrogen gas, which provides a low-oxygen-concentration atmosphere to allow work with the neptunium metal, a pyrophoric material.

The ^{237}Np sphere will be cast in a vacuum within a graphite mold designed by MST-6 to eliminate voids in the sphere volume. The mold design (shown in Figure 3) has been successfully tested by MST-6 using depleted uranium to cast a sphere to within about 0.010" on the diameter and very near theoretical density. MST-7 personnel encapsulate the ^{237}Np sphere first within tungsten hemispheres and then within two pure, welded Ni spheres for cladding. This project provides a tool to the Los Alamos Critical Experiments Facility staff for experiments for fundamental research.

Sanitization Project

Plans are being formulated to initiate a sanitization project at Los Alamos to remove classified aspects from actinide-contaminated, nonnuclear weapon components generated from disassembly of nuclear weapons and other weapons-related activities. The system designed for accomplishing this activity consists of an induction furnace for melting metallic components and a jaw crusher for pulverizing brittle, nonmetallic items. This system is tentatively planned for installation in Wing 9 of the CMR Building and consists of several glove boxes containing the processing equipment and a HEPA-filtered environmental housing surrounding the glove box system. Discussions with DOE sponsors are currently underway to establish the funding and scope of the operation.

ULISSES Program

To achieve the next generation of uranium processing technology, Los Alamos has developed the Uranium Line for Special Separation Science (ULISSES), a modular test-bed for chemical process optimization. This new processing line will include state-of-the-art dissolution chemistry and separation science, on-line sensors coupled to automated instrumentation, and recycling with waste minimization based on environmentally benign chemicals. This technology base will be used to develop, design, and demonstrate advanced chemical processing technology that minimizes hazardous wastes.

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Figure 2. The Wing 9 fabrication line produces uranium targets that will be used for production of ^{99}Mo.

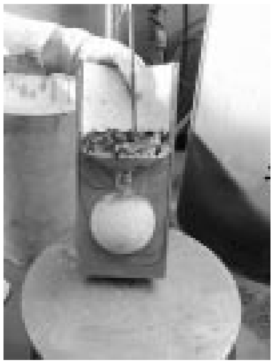


Figure 3. Mold design test casting using depleted uranium. Eventually a clad ^{237}Np sphere will be used for criticality measurements.

Wing 9 Hot Cells Support Work Involving Highly Radioactive Materials *(continued)*

This article was contributed by **Stan Bodenstein, Suzanne Helfinstine, Robert Romero,** and **Jim Ledbetter,** all of NMT-11.

Remote-Handled Transuranic Waste (RH-TRU)

The RH-TRU Program involved characterizing and packaging remote-handled TRU waste for ultimate WIPP disposal. Because many of the waste cans packaged thus far have typical radiation readings between 400 and 1000 R / hr at contact, a unique drum-loading and canister-handling system was designed and constructed for loading, welding, packaging, and transportation of the RH-TRU Waste (see Figure 4). To date, sixteen WIPP-approved canisters have been characterized, packaged, and placed into retrievable underground storage at Los Alamos (Area G) awaiting shipment to WIPP. This capability in the hot cell facility is operational to support the eventual disposition of RH-TRU waste at WIPP. The technology will also serve to resolve RH-TRU packaging problems at other sites within the DOE complex.

Magnetic Isotope Separation (MIS) Program

The MIS process is designed to separate isotopes for medical applications and the stockpile stewardship program. The process is performed in a separator system with the potential to process many different elements. The isotopes being separated are radioactive and have an activity of up to approximately 20 Ci. Radioisotopes of interest to be separated are ^{82}Sr / ^{85}Sr, ^{32}P / ^{33}P, ^{184}Re / ^{186}Re, ^{168}Tm, ^{170}Tm / ^{171}Tm, ^{173}Lu / ^{174m}Lu / ^{174g}Lu, ^{88}Zr / ^{93}Zr / ^{95}Zr, ^{190}Ir / ^{192}Ir, and possibly others. These radioisotopes are limited to nonactinide materials.

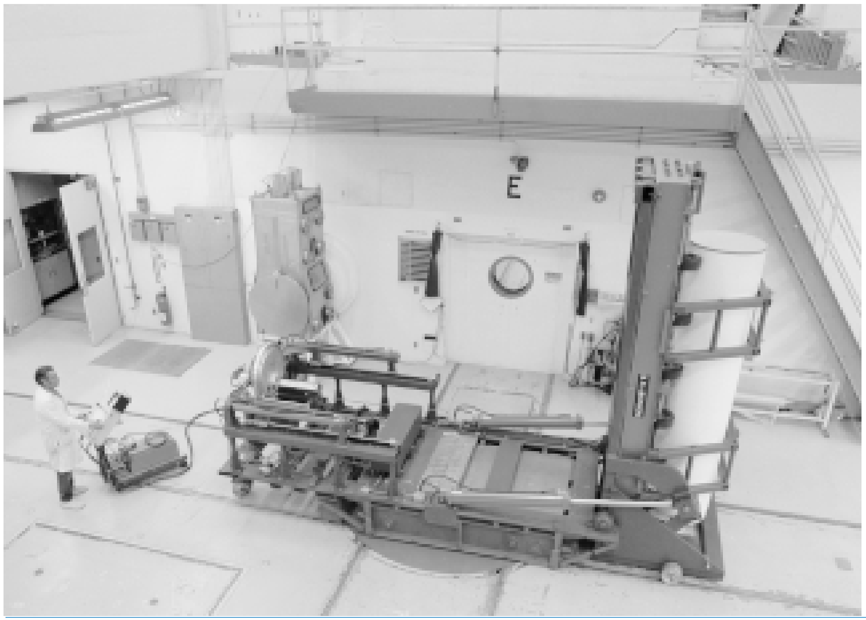


Figure 4. Drum-loading, canister-handling system for loading, welding, packaging, and transportation of waste destined for WIPP.

Accelerator Production of Tritium (APT) Program

This Wing 9 capability has been developed to support the APT Program by evaluating and testing candidate materials that have been irradiated (Alloy 718, 316L and 304L stainless steel, modified Fe9Cr1Mo(T91), Al6061-T6, Al5052-0) for use in the APT target and blanket. The specimens have been irradiated for fewer than 3,600 hours to a maximum proton fluence of 4 x 10^{21} p / cm^2 in the center of the proton beam. Specimens are then carefully removed from the packages in the hot cells in Wing 9 to begin mechanical testing, which includes tensile, bend and compact tension tests, metallographic examination, and transmission electron microscopy. Specimens will yield data on the effects of proton irradiation under a number of test conditions.

Actinide Source-Term Waste Test Program (STTP)

The Actinide STTP is designed to measure time-dependent concentrations of actinide elements from actual, contact-handled TRU waste immersed in brines that are chemically similar to those typically found in the underground formations of WIPP. The STTP will determine the effect of TRU waste matrices and brine chemistry on the concentrations and behavior of actinides under WIPP bounding conditions.

Several actual TRU waste types typical of DOE waste inventories have been characterized and loaded into specially designed test containers filled with brine containing additives to enhance the action of each influencing variable. The test containers are then placed in environmental chambers in Wing 9 to simulate in-situ conditions found at WIPP. Analysis is then performed on the brine leachate and headspace gas in the test containers.

Publications and Invited Talks
(December 1998–February 1999)

Editor's note: We get information from the library when papers have received LA numbers. However, it may be many months before the papers are published. Since ARQ *lists only those papers that are published or have been accepted for publication, we have no way to track them from the time they first receive numbers. We must rely on authors to tell us when their papers are accepted for publication in journals or proceedings. So when your paper is accepted, we'll tell your colleagues: please let us know! Ann Mauzy, 667-5387, mauzy@lanl.gov.*

Invited Talks

E. A. Blum, "Inorganic Membranes for Metal Ion Separations: Surface Effects on Cation Transport across Porous Alumina Membranes," IE&C Division, American Chemical Society, Anaheim, CA, March 21–25, 1999.

A. C. Lawson, B. Cort, J. A. Roberts, and B. I. Bennett, "Neutron Diffraction and the Physical Properties of the Light Actinides," American Nuclear Society, Washington, D.C., November 16, 1998.

Publications

D. K. Dennison, D. Gallant, D. C. Nelson, L. A. Stovall, and D. E. Wedman, "Automated Nuclear Material Recovery and Decontamination of Large Steel Dynamic Experiment Containers," in "Proceedings of the 8th International Topical Meeting on Robotics and Remote Systems," American Nuclear Society (ANS), ANS Pittsburgh Section, Monroeville, PA, in press.

P. A. Leonard, E. A. Strietelmeier, M. E. Pansoy-Hjelvik, and R. T. Villarreal, "Microbial Characterization for the Source-Term Waste Test Program (STTP) at Los Alamos," in WM '99 Proceedings, HLW, LLW, Mixed Waste and Environmental Restoration - Working Towards a Cleaner Environment, WM Symposia, Inc., Tucson, AZ, in press.

R. Marshall, A. S. Wong, and D. Thronas, "Lawrence Livermore National Laboratory Working Reference Material Production Plan," Los Alamos National Laboratory report LA-13552-MS (December 1998).

H. Nakotte, L. Laughlin, H. Kawanaka, Dimitri N. Argyriou, R. I. Sheldon, and Y. Nishihara, "Magnetic Properties of Single-Crystalline Mott Insulator $YV0_3$," *Applied Physics*, in press.

L. L. Romero, J. M. Hoffman, E. May Foltyn, and T. E. Buhl, "Operational Comparison of Bubble (Super Heated Drop) Dosimetry with Routine Albedo TLD for a Selected Group of Pu-238 Workers," accepted for publication in Operational Radiation Safety supplement to *Health Physics Journal*, in press.

S. B. Schreiber, S. L. Yarbro, E. M. Ortiz, F. Coriz, and S. Balkey, "Disposition of Mixed Waste Organics at the Los Alamos Plutonium Facility," Los Alamos National Laboratory report LA-13558-MS (January 1999).

S. M. Wander, I. R. Triay, P. S. Z. Rogers, S. T. Kosiewicz, D. R. Yeamans, A. R. Barr, J. D. Farr, J. Foxx, A. J. Montoya, M. A. Gavett, D. P. Taggart, S. E. Betts, G. R. Allen, H. D. Poths, D. R. Janecky , C. P. Leibman, G. I. Vigil, and J. J. Vigil, "Preparation of the First Shipment of Transuranic Waste by the Los Alamos National Laboratory. A Rest Stop on the Road to WIPP," in Proceedings of the Waste Management '99 Symposium, Laser Options, Inc., Tucson, AZ, in press.

O_2F in FOOF Holds Promise for Pu Decontamination *(continued)*

Success was immediate. The spectrum of O_2F was recorded without the interference of FOOF and analyzed. With this discovery we were able to study the properties of O_2F and its chemical relationship with FOOF, another Los Alamos first in laser photochemistry.
It was one of the most wonderful detective stories of an exotic chemical species and it helped to meet some of the important isotope separation program goals. The discovery of O_2F in FOOF found other applications in later years for recovery of plutonium from plutonium-containing residues and waste. In the future it may be a preferable alternative to acid dissolution of radioactive waste and can be used to recover plutonium from glove box systems and process lines in reactor vessels.

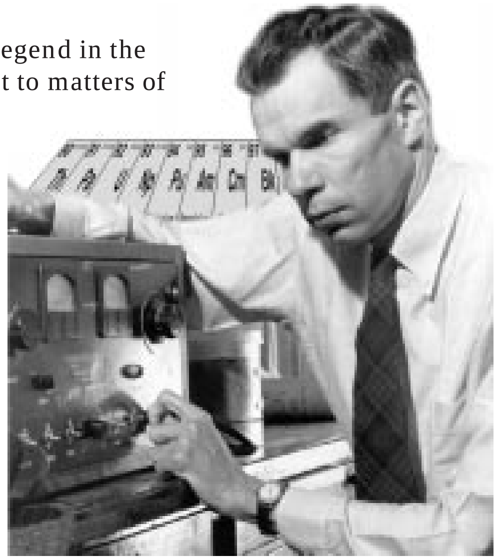
This article was based on the author's work and papers published in the mid 1980's.

NewsMakers

■ LBL Director Eulogizes Nobel Laureate Glen Seaborg, Who Died February 25, 1999.

Following are excerpts from Lawrence Berkeley Laboratory Charles V. Shank's memo to LBL employees:

> Dr. Seaborg was a true giant of the 20th Century, a legend in the annals of scientific discovery. His daily commitment to matters of the laboratory, even in retirement as associate director-at-large and as an active researcher, was an inspiration to us all…For his service to science, to education, and to our nation, we honor Dr. Seaborg's distinguished lifetime and will forever treasure his contributions to our institutions and to our lives. We who have been touched by his wisdom, his energy, and his tireless devotion to our profession will miss him… I encourage you to reflect on Dr. Seaborg's extraordinary life, much of it chronicled by him on an Internet Website accessible through our home page. Our thoughts and prayers are with the Seaborg family…



Dr. Seaborg's Life and Contributions are on-line at <http://www.lbl.gov/seaborg/> . Photographs are on-line at <http://www.lbl.gov/Science-Articles/Archive/seaborg-photo-gallery.html> . The 1951 Nobel Prize for Chemistry, presentation and acceptance speeches, are on-line at <http://www-library.lbl.gov/library/text/nobel/sea_mcmillan.html> .

■ Division Awards

Gordon Jarvinen has garnered the Division's 1999 R. D. Baker Award in Science and Technology; **Diedra Yearwood** will receive the 1999 William J. Maraman Award for Excellence in Facility Operations at an award ceremony to be held Thursday, May 13, during the Division Review.

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