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of the Nuclear Materials Technology Division

In This Issue

Zircon Promises to be A Host Phase for the Immobilization of Excess Weapon Plutonium

One of the new and daunting challenges in nuclear waste management is the disposition of plutonium recovered from dismantled nuclear weapons. Under the first and second Strategic Arms Reduction Treaties, as well as unilateral pledges made by the United States and Russia, several thousand nuclear weapons will be dismantled. Dismantlement will result in an estimated 100 metric tons of excess weapon plutonium that will require long-term disposition. The disposal strategy should not only protect the public and the environment, but should also ensure that, in the interests of nonproliferation and denying plutonium to terrorists, the plutonium is not readily recoverable.

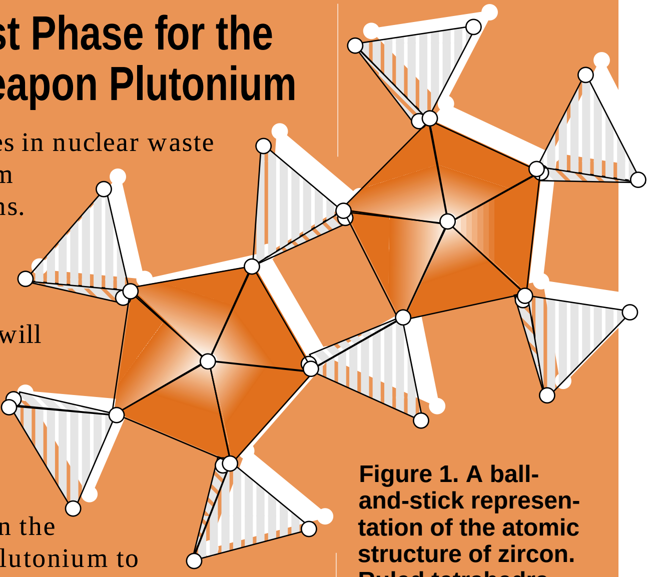

Figure 1. A ball-and-stick representation of the atomic structure of zircon. Ruled tetrahedra are SiO_4 groups, and shaded polyhedra are ZrO_8 groups.

Two recent reports by the National Academy of Sciences presented “promising alternatives”: 1) partial consumption of Pu in a mixed oxide fuel in existing or modified reactors and final disposal as spent nuclear fuel without reprocessing; 2) vitrification as a borosilicate glass mixed with radioactive high-level waste and disposal as “glass logs.” In both cases the waste will be so highly radioactive that it cannot be handled by terrorists or potential proliferators. A third, less studied, option involves deep burial (4 to 6 kilometers).

A key consideration for any disposition strategy is the form of the immobilized waste. The deep burial option will require a waste form durable at elevated temperatures (>150°C) that result from the normal geothermal gradient and with a waste loading sufficiently high so as to reduce the volume of material. Additionally, the higher the ^{239}Pu content, the more important are concerns for criticality. A highly durable waste form can retain not only the Pu but also other nuclides to serve as neutron “poisons.”

With such considerations in mind, researchers at the University of New Mexico (UNM) are initiating a collaboration with the Nuclear Materials Technology Division (NMT) of Los Alamos National Laboratory to investigate zircon ($ZrSiO_4$), a highly durable, naturally occurring mineral, as a host for the immobilization of Pu. This project is also part of a collaboration with Battelle Pacific Northwest

This article was contributed by Regents Professor **Rodney C. Ewing,** who is in the Department of Earth and Planetary Sciences at UNM. Collaborators on this research effort are Ewing and **W. Lutze** at UNM, **W. J. Weber** at PNL, personnel in the NMT Division at Los Alamos, and **E. Alexandrov** and **B. Burakov** at the Khlopin Radium Institute, Russia.

“A key consideration for any disposition strategy is the form of the immobilized waste.”

Laboratory (PNL) and the Khlopin Radium Institute in St. Petersburg, Russia.

Zircon occurs in nature with uranium and thorium in its structure in concentrations up to 5,000 ppm. It is an extremely durable accessory mineral in igneous and metamorphic rocks and is often found as a heavy mineral in stream sediments. After transport over great distances, it shows limited chemical alteration or physical abrasion. Some of the oldest zircons occur in sedimentary rocks that have been through numerous cycles of erosion, transport, and deposition, all the while retaining the original U/Pb systematics of the rock in which the zircon crystallized. Because of zircon’s importance in geochronological and geochemical studies of the earth’s crust, there are already numerous studies that have analyzed the variations in the U/Pb systematics as a result of radiation damage, thermal events, and alteration. Most recently, zircon with high actinide concentrations (10 wt % UO_2) have been identified in the Chernobyl “lavas,” molten materials resulting from the Chernobyl reactor meltdown accident.

The zircon structure is well known, and compositions of $ASiO_4$, for which A4+ = Zr, Hf, Th, Pa, U, Np, Pu and Am, were synthesized in the early 1960s. Four of these compositions, hafnon ($HfSiO_4$), zircon, coffinite ($USiO_4$) and thorite ($ThSiO_4$), occur naturally. There is complete miscibility between $ZrSiO_4$ and $HfSiO_4$. Zircon with 10 wt % Pu has already been synthesized. The fact that a pure, end-member composition, $PuSiO_4$, has been synthesized suggests that extensive substitution of Pu for Zr is possible.

“Radiation damage studies of zircon began in the early part of the century.”

Because of the large number of previous studies of zircon, a number of the issues that are inevitably raised in the evaluation of a nuclear waste form can already be addressed. Radiation damage studies of zircon began in the early part of this century. Zircon undergoes a radiation-induced transformation from the periodic to the aperiodic state at doses over the range of $10^{18 \text{ to } 19}$ α–decay events/g (= 0.2 to 0.6 displacements per atom) with a density decrease and a corresponding volume expansion. Previous studies provide a firm basis for predicting the microstructure of the radiation-damaged zircon as a function of dose and temperature. Based on these data, for a waste loading of 10 wt % of ^{239}Pu, the zircon will reach the saturation value of damage (1.2 x 10^{19} α—decay events/g or 0.8 dpa) in approximately 1,700 years.

Alteration of natural zircons has important implications for the use of U/Th/Pb techniques in geochronology; thus, there is an extensive literature that describes alteration. These studies show that even under extreme geologic conditions, the alteration is usually minor.

“One of the main advantages of zircon may be its high durability in an open system in which ground waters are present.”

Leaching studies of natural zircons in laboratory experiments have confirmed the loss of U, Th and Pb under hydrothermal conditions. However, at lower temperatures (< 80°C) and near neutral pH values, i.e., conditions more pertinent to nuclear waste disposal, zircon is extremely insoluble.

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Guest Editorial

Reducing the Nuclear Danger, Securing the Nuclear Future

Los Alamos has adopted the words of this title to be at the core of its institutional vision and mission for the future. How can we at the Laboratory make an impact? The answer lies in the responsible management of nuclear materials in the broad context: from guns, gates, and guards through stabilization and disposal of excess weapons materials, and, ultimately, through nuclear power. Three major issues surrounding our mission are nonproliferation, the environment, and national security.

Nonproliferation: Reducing the risk of proliferation requires secure storage and careful disposition of fissile materials. New treatment technologies and standards are needed to meet the requirement for long-term storage. For example, the ARIES (Advanced Recovery and Integrated Extraction System) hydride-dehydride system—an R&D 100 winner—removes plutonium from pits in an unclassified form for subsequent international verification.

The environment: Process residues and wastes remain and must be stabilized at various Department of Energy nuclear sites. Transportable modular systems employing advanced processing technologies offer the potential for cost-effective and timely eradication of this difficult Cold War legacy.

National security: The world is a safer place with a dependable U.S. stockpile than with one that is uncertain or nonexistent. Stockpile management, including pit surveillance and rebuilding, is essential to assure the continued safety and reliability of our nuclear arsenal.

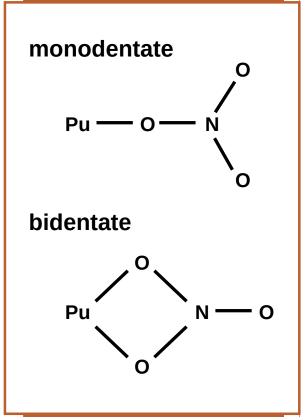
What about the global picture? Experience has shown that U.S. leadership through example can influence other nations in areas such as arms control and disarmament (if not so effectively in nuclear power and the nuclear fuel cycle). There is concern today, not only over the status of weapon plutonium in the former Soviet Union, but also over the growing number of nations separating plutonium in their commercial nuclear power fuel cycle. The U.S. now has opportunities to exercise leadership in disposition technology that may ultimately lead to minimization of world plutonium inventories while extracting maximum energy benefits needed for future growth.

In all of these areas, the capabilities and facilities of the Los Alamos plutonium programs are vital. We should be proud of our contributions in the past and confident of our ability to lead in the future. The Laboratory goals—reducing the nuclear danger and securing the nuclear future—will guide our priorities as we approach the next century.

Paul Cunningham, Program Director of the Nuclear Materials & Reconfiguration Technology Programs at Los Alamos National Laboratory

Collaborators Determine the Structure of Plutonium Nitrate Complexes in Nitric Acid

Figure 2.a. A monodentate plutonium nitrate. b. A bidentate plutonium nitrate.



The first determination of the structure of a plutonium complex in solution has been carried out by scientists in NMT Division in collaboration with the Materials Science and Technology (MST) Division and the Glenn T. Seaborg Institute at Lawrence Livermore National Laboratory (LLNL). The experiments were conducted at the Stanford Synchrotron Radiation Laboratory (SSRL) in Palo Alto, California in the summer of 1992. The geometry of a series of plutonium-nitrate complexes that form in nitric acid has been studied. The experiments were enormously successful in demonstrating that radioactive substances such as plutonium can be studied safely at SSRL and in establishing the structure of plutonium nitrate complexes in nitric acid.

An immediate benefit of the present study is its application to plutonium processing. The nitric acid anion exchange process used to purify plutonium at TA-55 is based on the capability of Pu(IV) to form both anionic (negatively charged) and cationic (positively charged) nitrate complexes. An anionic nitrate complex is found on anion exchange resins in contact with 8 M nitric acid. A cationic nitrate complex is eluted from the resin with 0.4 M nitric acid. Understanding the speciation of the Pu(IV) ion in such nitric acid solutions is fundamental to understanding this process. The questions we wish to answer about these chemical species are 1) How many water and nitrate ligands are coordinated to the central plutonium ion in each of the six possible species that can form in nitric acid? and 2) Are the nitrates coordinated bidentate (i.e., two of the nitrate oxygen atoms bound to the plutonium) or monodentate (i.e., one nitrate oxygen bound to the plutonium)? Obtaining a detailed structure for the complexes involved could, for example, allow new ion exchange resins to be designed to provide more efficient means of processing nuclear materials. New bifunctional anion exchange resins based in part on these results have been synthesized at Texas Tech University and tested at Los Alamos. These new resins, covered by a recent patent application, have shown much higher efficiencies in the nitric acid anion exchange separation of plutonium. The structures of plutonium nitrate complexes in solution had not been accurately characterized previously even though the nitric acid anion exchange process has been used and studied for nearly forty years.

Extended x-ray absorption fine structure (EXAFS) is a modern experimental technique used to obtain the distances and the numbers of atoms that surround a central x-ray-absorbing atom, in this case plutonium. This is the technique that was used at SSRL. It is based on interferences between light rays scattered off of the surrounding atoms and those passing directly to the central atom. These distances are small, on the order of Angstroms, so the wavelength of light needs to be small in order to see the interferences, thus x-rays are used. The effect is also quite weak so a bright x-ray source, such as found at modern synchrotron sources, is required. The EXAFS technique is element-specific because it uses the natural resonance absorption edge of the element of interest. The medium, in this case water and nitric acid, does not give rise to any signal, and data analysis is relatively straightforward.

We chose to study the species formed in 3 M, 8 M, and 13 M nitric acid. We knew from earlier studies that the principal species in 3 M nitric acid contains two nitrate ligands and an unknown number of water ligands. We also knew that in 13 M nitric acid the principal species contained six nitrate ligands and no water ligands. The structure of this plutonium hexanitrato complex was reasonably well known because 1) the structures of the plutonium hexanitrato anion in solution and in single crystals are the same and 2) single crystal structures of thorium and neptunium hexanitrato compounds have been determined and are thought to be the same as the structure of plutonium hexanitrato compounds. We chose to study the complexes in 8 M nitric acid because 8 M nitric acid is used in the anion exchange process and because we knew there are almost equal concentrations of the dinitrato complex, the hexanitrato complex, and a third complex tentatively assigned as the tetranitrato complex (contains four nitrate ligands and an unknown number of water ligands). We wanted to study the tetranitrato complex because the efficiency of sorption of Pu(IV) onto anion exchange resin is proportional to the concentration of this species.

Examination of the three curves in Figure 3 allows us to answer our questions. There are three principal peaks in the spectra at 1.8 Å, 2.5 Å, and 3.6 Å. (The smaller wiggles are artifacts of the data analysis.) The peak positions and intensities correspond to the position and the number of atomic nearest-neighbors of Pu respectively. The largest peak represents the nearest-neighbor oxygen atoms (or first-shell oxygen atoms), which are part of the nitrate and water ligands. The peaks at 2.5 Å and 3.6 Å arise from second-shell nitrogen atoms and third-shell noncoordinated oxygen atoms of the nitrate ligands. The positions of these peaks indicate that the nitrate ligands are bound bidentate as shown in Figure 2b. If the nitrates were monodentate, then the positions of the peaks would have been different. The intensities of the outer two peaks are proportional to the number of nitrates bound to the plutonium. Systematic increases in the intensities of the nitrogen peak and the noncoordinated oxygen peak clearly demonstrate the trend of increasing nitrate ligation as a function of increasing nitric acid concentration. At the same time the intensity of the first peak does not appear to change substantially indicating that the nitrate ligands are replacing the nearest-neighbor water ligands. A quantitative analysis indicates that the number of first-shell oxygen atoms is either 11 or 12. Therefore, the dinitrato complex which has 4 first-shell nitrate oxygen atoms must also have 7 or 8 nearest-neighbor water ligands to bring the number of first-shell oxygen atoms to 11 or 12 and the tetranitrato complex must have 3 or 4 nearest-neighbor water ligands.

The complete text of a journal article describing this work and the quantitative analysis can be found on the World Wide Web at http://w3.anl.gov:8002/people/dsanchez/public/public.html or on the NMT-6 Home Page when its construction is complete.

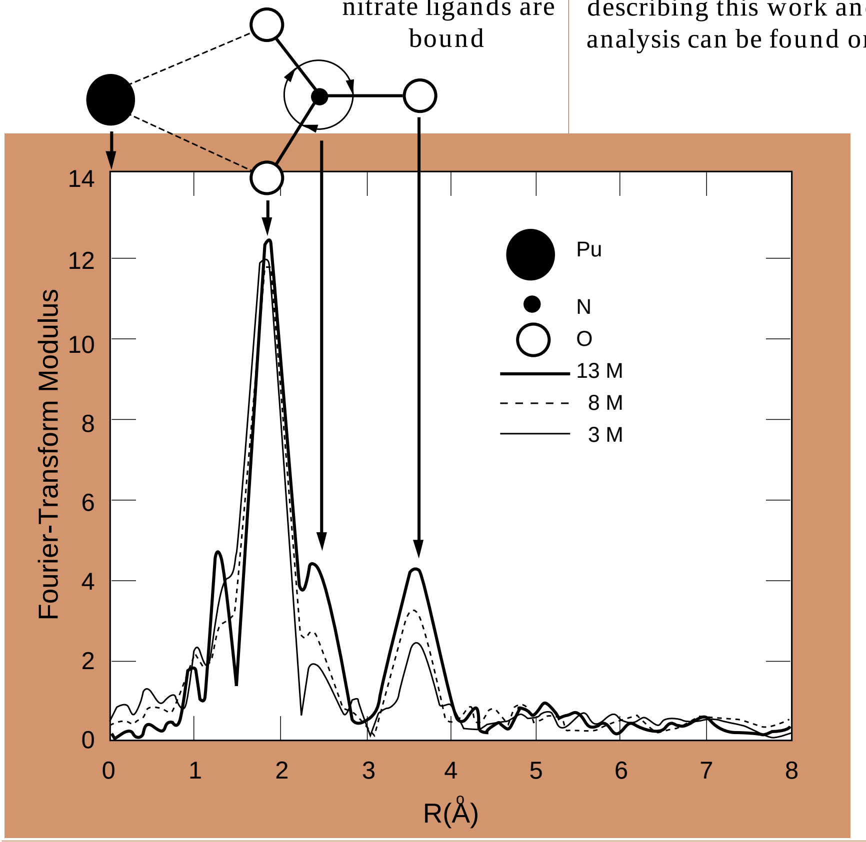


Figure 3: For the first time in nearly 50 years, scientists have determined the structure of a plutonium complex in solution. Qualitative information can be obtained from Fourier-transform analysis of raw EXAFS data as shown here.

Contributors to the project include Pat Allan of the Glenn T. Seaborg Institute for Transactinium Science at LLNL, **Kirk Veirs** and **Coleman Smith** of NMT Division, Steve Conradson of MST Division, and Fred Marsh of Sandia National Laboratory. The experiment required the help of a large number of TA-55 groups and individuals. **Bill Haag** in NMT-4 handled the requirements for proper transportation containers and made sure that the samples were received at Stanford. Radiation Control Technicians (RTCs) from TA-55 made the trip to Stanford to provide radiation protection as Stanford does not have properly trained RCTs. Experiments were conducted day and night. For our samples **Arch Nixon** provided the RCT coverage.

Molten Salt Chemistry Plays a Prominant Role in Accelerator-Driven Transmutation Systems

The nuclear system being studied by the Accelerator-Driven Transmutation Technology (ADTT) project is an accelerator-driven, subcritical nuclear reactor with molten salt used as the working fluid.

The nuclear system being studied by the Accelerator-Driven Transmutation Technology (ADTT) project is an accelerator-driven, subcritical nuclear reactor with molten salt used as the working fluid. Such a system could be used to destroy weapon plutonium and plutonium contained in Defense Program (DP) wastes and spent nuclear fuel. Briefly, a proton beam generated by an accelerator is directed at a target material of high atomic number (e.g., Pb, Bi) resulting in the production of neutrons by spallation. These neutrons are used to cause the plutonium to fission and to enhance the destruction of fission products. The plutonium, present as plutonium trifluoride dissolved in 2 ^{7}LiF - BeF$_2$ fuel salt at 600 °C–750 °C, is circulated through the core of the system, heat exchangers, and fission-product-removal modules. The system shares some common characteristics of the reactor designs developed and studied at Oak Ridge National Laboratory (ORNL) during the 1960s and 1970s. The fuel-preparation process and the removal of fission products are essential elements in the operation of these systems. The following paragraphs describe an overview of the chemistry associated with the conceptual fuel-preparation process and the conceptual separation process for the transtion metal fission products, lanthanide fission products, and gaseous fission products.

The essential step in fuel preparation consists of the hydrofluorination of plutonium in the presence of excess hydrogen to produce plutonium trifluoride dissolved in the fuel salt. Excess hydrogen is used to prevent the formation of volatile plutonium hexafluoride. Conversion of weapon plutonium to ADTT fuel requires no preprocessing. However, converting the plutonium contained in DP wastes and spent nuclear fuel to ADTT fuel requires additional processing, as described below.

Treatment of DP wastes consists of the addition of the waste form (e.g., contaminated graphite or ceramic) to a secondary ^{7}LiF-NaF-KF molten salt, where the plutonium is hydrofluorinated in the presence of excess hydrogen. The waste matrix (e.g., graphite or ceramic) is removed from the secondary salt by physical means, such as filtration. The secondary salt is used in the hydrofluorination step instead of the fuel salt so that when the waste matrix is removed from the salt, it is not contaminated with BeF$_2$ and may be discarded as nonhazardous waste. Finally, the secondary salt is blended with the fuel salt before it is transferred to the nuclear system.

Spent nuclear fuel consists of plutonium dioxide, uranium dioxide, fission products, and metal cladding. The spent fuel is decladded (although decladding is not essential), and the oxide fuel matrix is crushed, added to the fuel salt, and hydrofluorinated to produce soluble fluorides. Excess hydrogen prevents the formation of volatile fluoride species (e.g., MoF$_6$, UF$_6$, PuF$_6$). Gaseous fission products (e.g., Xe, Kr) released during the process must be adsorbed on activated charcoal or molecular sieves. Next, an electrowinning process is used to remove the transition metal fission products and uranium from the fuel salt. The plutonium remains in the fuel salt because its reduction potential falls below that for beryllium, a major constituent of the fuel salt. The electrowinning process is described in more detail later in the article. Finally, the fluoride potential of the fuel salt is adjusted before it is transferred to the nuclear system.

After the fuel salt has been introduced into the nuclear system and as the fission process occurs in the system, the fission product concentration increases in the fuel salt. A class of transition metal fission products that poses several problems is the so-called “noble metal” fission products: Mo, Nb, Ru, Rh, Ag, Cd, etc. These could potentially become plated onto surfaces in the heat exchanger and elsewhere in the system. They also affect the neutron economy of the system. The on-line removal of the noble metals from the molten salt is crucial to the operation of the system. Electrowinning is envisioned for removing the noble metal fission products.

Electrowinning methods have been used extensively to produce pure metals from oxide or halide feed material dissolved in molten salt

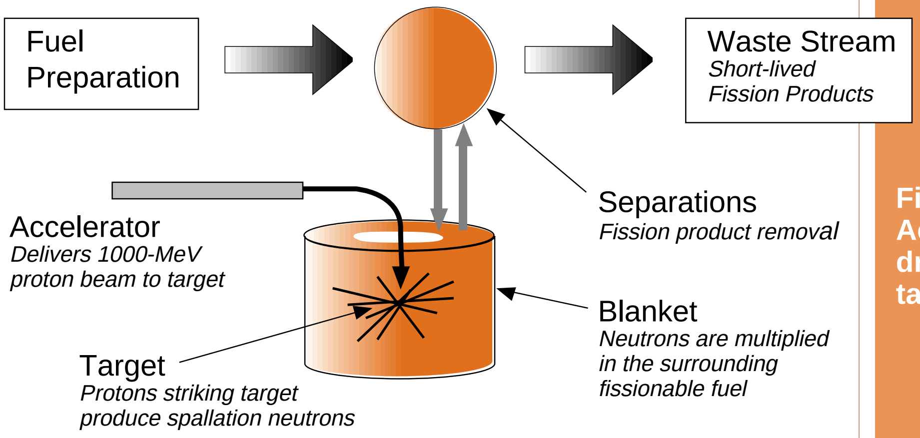


Figure 4: Accelerator-driven transmutation of waste.

(e.g., Al, Mg production). In this application of the method, one is interested in purifying the molten salt and not in producing a pure metal. The electrowinning cell consists of a consumable Be anode and a nickel cathode onto which the metals are deposited. The reaction is spontaneous because of the difference in free energy between metal fluorides. Therefore, in principle, the cell could be operated passively (i.e., no external voltage source required). However, the efficiency of the cell would be enhanced by applying an externally generated voltage. Plutonium, higher actinides, lanthanides, Sr, and Cs remain in the fuel salt. The cell could be located ahead of the heat exchanger system to prevent the deposition of fission products on the heat exchanger tubes. Such deposition poses no safety threat to the system in terms of material interactions, but it would degrade the efficiency of the heat transfer system. Maintenance of the cell would consist of the periodic, remote replacement of the anode and cathode. The metals collected on the cathode could be oxidized, blended with silica, and vitrified.

Another class of fission products that must be removed from the fuel salt is the lanthanides. These must be removed to maintain the neutron economy in the system. Also, they must be removed because of the similarity of their solubility and physical properties with trivalent plutonium. The ADTT system has two options for the removal of the lanthanide,s and each is being evaluated on the basis of waste minimization and its resistance to plutonium proliferation. The options are described below.

One option for removing the lanthanides is liquid-metal extraction coupled to a liquid-metal centrifuge system. Scientists at ORNL studied the extraction of lanthanides and plutonium from fluoride molten salt using liquid bismuth that contained a small concentration of lithium. In this application, the liquid-metal extraction step is used as a feed for the centrifuge system. The lithium concentration is chosen so that both the lanthanides and plutonium are extracted into the liquid bismuth. The bismuth is used to transport the species to the centrifuge system, where they are separated from each other. Plutonium contained in one of the bismuth streams that comes from the centrifuge system could be back-extracted into the fuel salt and fed back into the nuclear system. Lanthanides contained in the other bismuth stream could be removed from that stream by decreasing the temperature of the bismuth, collecting the solids that are formed, blending the solids with silica, and vitrifying the waste material. The bismuth from each stream could be recycled. The centrifuges could be operated at modest speeds and moderate temperatures while still providing adequate separation.

Alternatively, the lanthanides and plutonium could be separated by a multiple-stage, liquid-metal extraction process. In this process, the lithium concentration in the bismuth is selected so that the plutonium is preferentially extracted from the fuel salt in the first extraction stages. A different lithium concentration is selected for use in subsequent extraction stages so that the lanthanides are removed from the fuel salt. The two bismuth streams produced by this option could be processed in the same way as the two streams that come from the centrifuge system.

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Participants in this phase of the ADTT project include **N. Li**, **F. Venneri**, and **M. Williamson.**

NMT Team Profile

TA-55's Cold Machine Shop Provides Valuable In-house Fabrication

“If it's in a glovebox in PF-4, we probably made it.”

Scientific research is central to TA-55’s functions, and the technical articles resulting from this research are central to our quarterly publication. This article, however, salutes different sort of TA-55 asset: the Nuclear Materials Technology Division (NMT) Cold Machine Shop (There is a “Hot” Shop in PF-4 that works with radioactive materials, hence the distinction “Cold.”) The Cold Machine Shop, or “Shop 55,” has been an important part of TA-55 since the Plutonium Facility opened in 1978, and its in-house fabrication work contributes to many technical programs both within and outside of TA-55.

From its founding in 1978, the shop was part of the Mechanical Division (MEC) until October 1, 1992, when MEC Division was dissolved. The shop then became part of NMT. The seven machinists in the shop work with groups in NMT and other divisions to supply parts for lab experiments as well as for use in gloveboxes in Building PF-4 (the Plutonium Facility). They fabricate a wide variety of components and assemblies of varying complexity, using all the standard machine shop equipment, as well as two Computer Numerical Controlled (CNC) mills, and one CNC lathe. “If it’s in a glovebox in PF-4, we probably made it,” says Assistant Supervisor Richard Lemler.

Other Shop 55 employees are Supervisor Leonard Busch, David Archuleta, Angela Baca, Jose Garcia, John Mosley, and Robert Valdez. Some were trained through the Laboratory’s apprenticeship program, and together they boast a combined 181 years of machining experience. Archuleta and Garcia operate the CNC mills, which are “not user friendly,” in Archuleta’s words. Baca operates the CNC Quickturn Lathe, turning out parts that are accurate to within .0001 of an inch. Mosley is currently learning to operate this machine, though for the most part he operates the more conventional equipment, as do Lemler and Valdez. The shop also has some welding capabilities.


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Figure 5. The TA-55 machinists (L to R): Archuleta, Valdez, Garcia, Mosley, Busch, Baca, and Lemler.

The machinists at Shop 55 handle jobs ranging from the production of one specialized prototype part to the thousands of parts being used in the Cassini Project. The more standard-size jobs can often be completed in just one day. In addition to making parts, the machinists often work closely with the scientists, engineers, and other personnel who are their customers to help design or modify parts. In a survey of Shop 55 customers taken in August 1994, customer Larry Tellier wrote, “I really appreciate the machinists’ point of view when I have a problem to solve before I even submit a drawing.”

Shop 55’s customers particularly value the machinists’ willingness to work together and make midprocess changes, thus saving time and money. In the survey, customer Wayne Smyth explained, “On many occasions changes must be made to a drawing because of an unforeseen obstruction in the original design, or because of an engineering change in scope by the project supervisor or technician. Such changes may cause substantial problems with an outside organization because of contract agreements leading to excessive cost escalation.” On the other hand, Shop 55 can more easily accommodate its customers because it is internal. Many customers agree that having to use an external shop would greatly slow down their projects and increase costs.

Besides being convenient and cost-effective, TA-55 machinists are very easy to work with. While an outside contractor would demand complex official drawings, Shop 55 machinists are willing and able to work from sketches made on everything from napkins to paper lunch sacks, thus saving their customers time and money. Customer Gene M. Jacquez commented in the survey that shop employees have quickly completed many urgent orders by working overtime and doing whatever else was necessary to get the job done. Richard Lemler says that they always have more than enough work to do and often have to work overtime. “If we’re too busy or can’t do the job here in the shop,” he says, “we’ll take it to the appropriate shop that can handle it.” Customer Rudy Fernandez wrote in the survey that he knows the TA-55 machinists by their first names, and that the possibility of frequent contact with them makes his job much easier. Customers agree that Shop 55 and the machinists who work there are a valuable asset to NMT Division.

This article was contributed by Laura Linford.


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Figure 6: View of the Cold Machine Shop, with conventional mills in the foreground and the blue CNC lathe in the back.

Shop 55's customers particularly value the machinists' willingness to work together and make midprocess changes, thus saving time and money.

Recent Publications and Reports

Presentations to the 9th Annual Conference of the American Glovebox Society, Minneapolis, MN, July 19-22, 1995.

J. T. McFarlan, D. K. Veirs, C. A. Smith, and J. M. Berg, “Glovebox Well and Cuvette Transport Mechanism for UV VIS.”

J. T. McFarlan and T. O. Nelson, “Cryogenic Liquid Introduction System for Gloveboxes.”

T. O. Nelson, D. D. Hill, M. E. Huerta, L. Jaramillo, H. E. Martinez, J. T. McFarlan, A. N. Morgan, C. F. Prenger, W. R. Romero, and L. H. Stapf, “Demonstration of a Cryogenic Grinding System in a Glovebox,” Los Alamos National Laboratory Report LAUR 95-2183.

T. O. Nelson, L. Jaramillo, U. F. Gallegos, H. E. Martinez, W. R. Romero, and L. H. Stapf, “Electrolytic Decontamination of Gloveboxes,” Los Alamos National Laboratory Report LAUR 95-2184.

M. E. Evans, D. W. Gray, and G. Peralta, “Applications of LabVIEW Programming in a Glovebox Environment.”

Other Presentations

M. A. Williamson and F. Venneri, “Front-end and Back-end Electrochemistry of Molten Salt in Accelerator-Driven Transmutation Systems,” Global International Conference on Evaluation of Emerging Nuclear Fuel Cycle Systems, Versailles, France, September 11-14, 1995, Los Alamos National Laboratory Report LAUR 95-1296.

J. N. Dann, T. C. Huang, W. F. McClune, M. H. Mueller, A. C. Roberts, and P. L. Wallace, “Analysis of Metallic and Related Compounds Using ICDD Metals and Alloys Indexes,” European Powder Diffraction Conference IV, Chester, England, July 10-14, 1995.

C. W. Hoth, “Packaging of Nuclear Materials for Long-term Storage at Los Alamos,” Proceedings of the 36th Annual Meeting of the Institute of Nuclear Materials Management (INMM), Palm Desert, CA, July 9-12, 1995.

S. M. Long, S. S. Hildner, D. D. Gutierrez, C. J. Mills, W. E. Garcia, and J. N. Gladson, “Fabrication of 12% 240Pu Calorimetry Standards,” Proceedings of the 36th Annual Meeting of the Institute of Nuclear Materials Management (INMM), Palm Desert, CA, July 9-12, 1995.

J. R. Hurd, F. Hsue, and P. M. Rinard, “In-Plant Experience with Passive -Active Shufflers at Los Alamos,” Proceedings of the 36th Annual Meeting of the Institute of Nuclear Materials Management (INMM), Palm Desert, CA, July 9-12, 1995.

F. Hsue, J. R. Hurd, and P. M. Rinard, “Operating New 55-Gallon Drum Shufflers at Los Alamos,” 5th International Conference on Facility Operations-Safeguards Interface, American Nuclear Society, co-sponsored by the Institute of Nuclear Materials Management, Jackson Hole, WY, September 24-29, 1995.

D. J. Post, “Improving the Formality of Operations in a Nuclear Facility,” Master’s Degree Final Project, submitted to Prof. D. E. Mandeville, Oklahoma State University (National Technology University, MB 780-W) June 22 1995.

T. O. Nelson, M. E. Huerta, L. Jaramillo, and A. N. Morgan, “Electrolytic Decontamination of Conductive Materials for Hazardous Waste Management,” I&EC Special Symposium, ACS 7th Annual Symposium on Emerging Technologies for Waste Management, Atlanta, GA, September 17-20, 1995, Los Alamos National Laboratory Report LAUR 95-2474.

K. W. Hench, L. M. Noell, J. D. Olivas, and P. R. Finch, “Modeling Pit Fabrication and Plutonium Recovery: an Integrative Approach,” Tri-Laboratory Engineering Conference on Computational Modeling, Pleasanton, CA, October 31-November 2, 1995.

K. W. Hench and Amjed Al-Ghanim, “The Application of a Neural Network Methodology to the Analysis of a Dyeing Operation,” Artificial Neural Networks in Engineering (ANNIE) Conference, November 12-15, 1995.

A. N. Morgan and T. O. Nelson, “Alpha Swipe Spectroscopy for Oralloy Disposition,” 19th Annual Actinide Separations Conference, Monterey, CA, June 12-15, 1995, Los Alamos National Laboratory Report LA-CP 95-52.

Publications

R. I. Sheldon, J. R. Ruble, L. E. Pence, and K. Bowman-James, “Potassium Nitrate,” submitted to *Acta Crystallographica*, July 10, 1995.

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Zircon Promises to be A Host Phase for the Immobilization of Excess Weapon Plutonium, *continued*

There are much less data than necessary for a full evaluation of zircon as a waste form, which will be the focus of the UNM-LANL collaboration. Crystalline zircon is stable to such an extent that the equilibrium concentrations of Zr and Si are in the order of 10^{-9} moles / L (0.1 ppb) at 25°C. The dissolution rate of amorphous zircon is still considerably lower than that of glass in stagnant, silica-saturated solutions. In an open system (e.g., moving ground water), the leach rate for zircon does not increase; however, the leach rate of borosilicate glass may increase by three orders of magnitude. Thus, one of the main advantages of zircon may be its high durability in an open system in which ground waters are present, as this attribute allows considerably greater flexibility in repository design.

Criticality is a concern for any Pu-waste form during both processing and final waste disposal. The possibility of criticality can be mitigated by adjusting the waste loading in the zircon or by including neutron “poisons,” such as hafnium and gadolinium, in the formulation. Natural zircons contain up to several thousand parts per million rare earths, including up to 5,000 ppm of Gd, and hafnium is a bothersome, major impurity in natural zircons. Thus, neutron-absorbing nuclides may be incorporated into the zircon structure as dilute solid solutions.

Processing and production technologies are always an important consideration in the adoption of a waste form. As already discussed, Pu-doped zircons have been synthesized with 10 wt % Pu. Zircon can be synthesized by sintering (1200°C to 1300°C), and yields of nearly 90% are obtained in less than one day. Although larger-scale pilot demonstrations are required, much is already known about the requirements for synthesis. In the case of weapon plutonium, one may use to advantage the essentially reagent grade of the Pu in engineering the process and the waste form. The final processing technology can draw directly on the extensive experience gained in producing mixed-oxide fuel from sintered powders.

NMT People

■ **Deborah Pudell,** a chemistry and mathematics teacher at Laker High School in Pigeon, Michigan, assisted **Jerry Foropoulos** of NMT-6 in his work on the destruction of volatile chlorocarbons. Her eight-week stay under the DOE/LANL TRAC program gave Pudell a wealth of information for her classroom and a unique opportunity to contribute to research at TA-55.

■ **Heather Hawkin**s has been working at TA-55 in the stabilization of solid residues since July 1 this year. She is a graduate student at the Pennsylvania State University's Materials Research Laboratory. **Nicholas Coppa** of NMT-6 is serving as her thesis advisor.

■ **Laura Linford**, a graduate student in humanities at the Florida State University, has been assisting Chief Scientist **K. C. Kim** with the division's Science and Technology Assessment since May 15 this year.

■ A team of some twenty Russian scientists and engineers toured the Los Alamos Plutonium Facility on August 16. The tour was arranged as part of the “Workshop on Non-Reactor Nuclear Safety and Waste Management,” held at Los Alamos under the sponsorship of the U. S. Government. The group toured the following laboratory areas: ^{238}Pu fabrication, hydride-dehydride process, MOX fuel fabrication, nondestructive assay, and aqueous processing of Pu residues. This is the second time within a year that a Russian delegation has visited TA-55.

Molten Salt Chemistry Plays a Prominant Role in Acelerator-Driven Transmutation Systems, *continued*

Finally, xenon and krypton isotopes produced during the fission process also affect the neutron economy of the system and must be removed from the fuel salt. A helium sparge gas could be used to remove the xenon and krypton from the fuel salt and to transport the gases to a fractional distillation system, where they would be separated from the helium and from each other. A similar system was proposed and designed by ORNL for use with the molten salt breeder reactor.

Several of the main chemical processes associated with the fluid-fueled system have been described in this article. Other significant issues are being studied, but these will be left for future discussion. As an ending note, scientists at ORNL have described the molten salt nuclear system as “a chemist’s reactor,” and it truly is.

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