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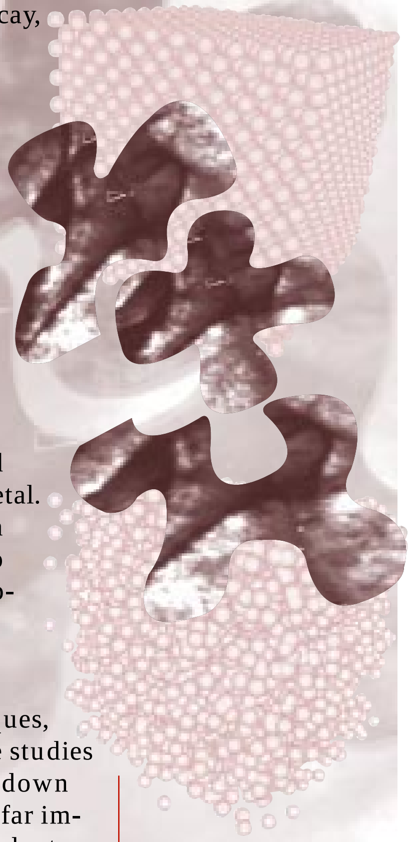
In This Issue

Aging Effects in Plutonium Metal Pose an Interesting Puzzle

Aging effects in plutonium metal are of metallurgical and practical interest. The Dismantlement Program has provided a wide variety of plutonium samples from a large number of retired nuclear weapons, enabling the development of a comprehensive database of materials properties as a function of age. Data are providing a substantive statistical foundation for these properties as well.

The primary cause of aging in plutonium is radioactive decay, which alters composition and deposits energy into the lattice. Three major consequences of this decay are annealing, changes in phase, and in-growth of helium.

Daughter species produced during radioactive decay alter the composition of the metal directly with age, in a known way. Composition may in turn affect phase, or lead to metastability. Impurities can affect mechanical properties by controlling the mobility and distribution of defects. Ingrowth of daughter species during radioactive decay may restabilize a metastable condition, as some of the daughters are known to serve the same function as gallium in stabilizing the delta phase.



Energy deposited in the metal lattice produces disordered regions, including dislocations and vacancies. The number and movement of dislocations govern the plastic properties of a metal. Impurities or lattice defects produced by radioactive decay can block the passage of a dislocation, making the crystal harder to deform. Vacancies in the crystal can migrate to form small loop-shaped dislocations that can take part in plastic deformation. Thus, radioactive decay can alter the mechanical properties of a metal through a complicated set of competing processes.

Age-induced changes are measured by a variety of techniques, as part of the Enhanced Surveillance Program and surveillance studies on plutonium metal of various ages. LIBS (laser-induced breakdown spectroscopy) gives a measure of annealing by indicating how far impurities have moved within the metal. Precision density and indentation testing (hardness measurement) may be sensitive enough to indicate the degree of annealing. Metallography reveals any large-scale structural perturbations resulting from annealing. Changes in phase are best revealed by x-ray diffraction measurements and by examination of the density of the material. Indentation testing may also indicate phase changes. Helium bubbles within the metal reduce the

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Aging Effects in Plutonium Metal Pose an Interesting Puzzle *(continued)*

This article was contributed by **Roberta N. Mulford** and **Wendel Brown** (NMT-15)

density, and any bubbles present are usually observable in metallographic images. Helium dispersed in the lattice can be measured only by provoking helium release using differential scanning calorimetry (DSC) or helium evolution analysis.

A damage cascade is the region of the lattice disrupted by the decay of a radioactive atom within the lattice. (See Figure 1.) In a damage cascade the daughter products from the decaying atom knock neighboring atoms out of position. These atoms in turn move other atoms within the lattice. The entire "collision phase" process produces a large region in which the lattice has been disrupted, producing dislocations, interstitial atoms, vacancies, and other lattice defects.

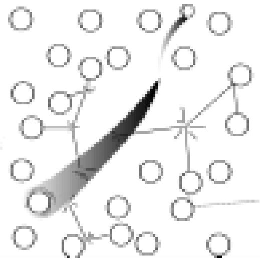


Figure 1. The primary cause of aging in plutonium is radioactive decay. This figure shows the beginning of a damage cascade, the region of the lattice disrupted by the decay of a radioactive atom within the lattice.

The kinetic energy in the lattice is dissipated as heat and rapidly conducted away, producing the "thermal spike phase," in which the lattice rearranges, and many of the disrupted atoms settle back into nearby equilibrium lattice positions, leaving a few disruptions. (See Figure 2.) The volume affected depends on the energy of the decay and can be both calculated and measured. In plutonium the affected volume is as large as several million atoms.

The energy of the decay may be sufficient to induce local melting in the region of the lattice affected by a damage cascade. Low-energy cascades may produce structural damage and minimal melting. In plutonium, the cascade energy is high, and local melting reduces the concentration of primary defects caused by damage cascades.

Annealing occurs when radioactive decay deposits energy in the lattice. Local recrystallization drives the microstructure toward equilibrium, usually toward a more-ordered condition, raising the purity of the ordered region and localizing impurities, usually moving them to grain boundaries. Thus, annealing can cause local compositions to vary from the bulk.

Phase changes may arise from these local changes in alloy composition, if there is a nucleation site to start the process. Phase changes may be nucleated by damage cascades arising from radioactive decay or they may be nucleated by lattice strain. Plutonium may be stabilized in the delta phase by addition of approximately 1 wt % of gallium. Radiation-induced changes in the distribution of the gallium can result in regions of the lattice that are no longer stabilized.

Without nucleation to provide a kinetic driver promoting transformation, metastable delta plutonium can persist for long times at room temperature or above, since the atoms lack sufficient thermal energy to move en masse into the new equilibrium lattice positions in the monoclinic alpha-phase lattice. The transition itself is martensitic.

Damage cascades may promote transformation by simply rearranging the lattice, by increasing the presence of interstitial decay products in the metal matrix, or by producing dislocation loops. (See Figure 3.) Interstitial atoms and decay products incorporated into the metal matrix produce disordered or strained regions in the lattice, which can more easily reorder themselves into the lowest energy state, the new phase. Time scales can be rapid, such as recrystallization during the damage cascade, or slow, such as the progress of thermal annealing at the edge of an existing disordered region or at a frontier between two phases. It has also been demonstrated that during decay, the trapped alpha particle produces an effective local pressure in the lattice as high as 2 GPa, which is sufficient to drive a pressure-induced transformation to the alpha phase.

Helium is produced in the lattice by alpha decay of the ^{239}Pu, reaching a predictable concentration as a function of age. Very little helium is released by the metal, even though the solubility of the helium in the metal is expected to be low. Helium that is not dissolved is evidently trapped in the lattice, in configurations that have varying degrees of stability. Configurations of low stability may permit mobility of trapped helium and a potential for accumulation into small bubbles leading to void swelling, a phenomenon seen in other metals in radiation environments. Void swelling may reduce the density and produce softening of the metal.

The migration of the helium is affected by its location. Helium atoms from alpha decay may come to rest in lattice sites, as interstitials, in vacancies, in dislocation cores and voids, in other large distortions of the lattice, or at grain boundaries. Different locations of helium atoms will produce different lattice energies, different lattice strains, and different barrier energies to mobility of the helium. Calculating the mobility of helium assuming simple energy barriers between helium occupation sites gives a rate for helium agglomeration that is linear or decreasing in time. Void formation generally begins only after an induction time, indicating that location and barrier height are not the only factors governing the process.

Either migration of vacancies to form the voids or dislocation mobility in the metal may be the rate-determining phenomenon in this process, as seen in other metals. The induction time for void formation in plutonium near room temperature is evidently as long as 50 years. Thus, data from particularly old metal is valuable; such metal may be further into the induction time than the majority of such samples examined to date.

A proper model of the process evidently should include the interaction of the helium with dislocations, voids, impurities, and other structural perturbations as well as with the uniform structure of the bulk metal. Disruption or melting of the lattice during the decay cascade introduces strong perturbations into any description of the migration of the helium.

Since significant changes in density are thought to occur only during void formation and not during the accumulation of dispersed trapped helium in the metal, it is important to be able to correlate subtle changes in lattice structure with precise measurements of helium in the lattice, made by observing evolution as a function of temperature.

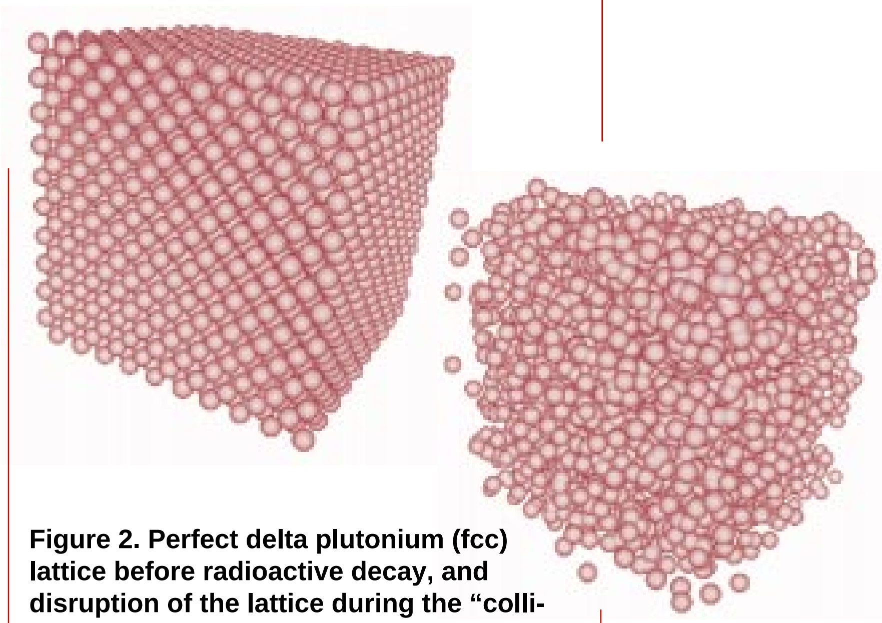


Figure 2. Perfect delta plutonium (fcc) lattice before radioactive decay, and disruption of the lattice during the “collision” phase of the decay cascade. The lattice will relax back to near perfection during the “thermal spike” phase of the cascade.

Experimental data include measurements of helium evolution as a function of temperature, and observation of expansion of the solid metal with increasing temperature. The temperature dependence of mobility may itself provide structural information. Past studies of this type have observed nonuniform evolution of helium with increasing temperature, possibly helium released from the various sites listed above.

Aging effects in plutonium pose a complicated problem involving many interrelated variables, and dependencies on quantities such as dislocation mobility that are relatively difficult to measure or to estimate. Careful statistical analysis of a large database of measurements on varying samples may enable us to make meaningful extrapolations both forward and backward in time, permit analysis of the relationships between properties, and indicate the relative importance of several physical effects that are being explored. A clear understanding of the aging mechanisms in plutonium metal will enable us to more confidently predict the safety and reliability of our nuclear deterrent well into the twenty-first century.

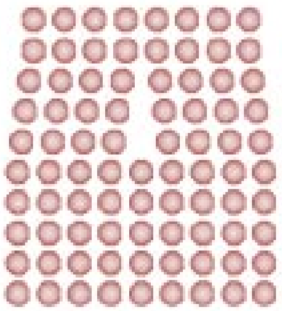


Figure 3. Energy deposited in the metal lattice produces disordered regions including dislocations and vacancies. Such damage can alter the mechanical properties of a metal through a complicated set of competing processes. This figure shows dislocation in the lattice as a result of a damage cascade.

NMT-9 Recycles Heat Source Fuel

This article was contributed by Gary H. Rinehart (NMT-9).

Los Alamos has been involved in the fabrication of heat sources to power experiments for space exploration since 1979. With the shutdown of the Savannah River reactors, the United States has purchased $^{238}PuO_2$ from Russia to supplement US heat source material for NASA's future space missions. These missions include the Pluto / Kuiper Express, Europa / Orbiter, Solar Probe, and a number of Mars surface science and sample return missions. A significant fraction of the heat source fuel in the Los Alamos inventory, including the Russian material and fuel from disassembled milliwatt heat sources, requires purification before it can be fabricated into new heat sources.

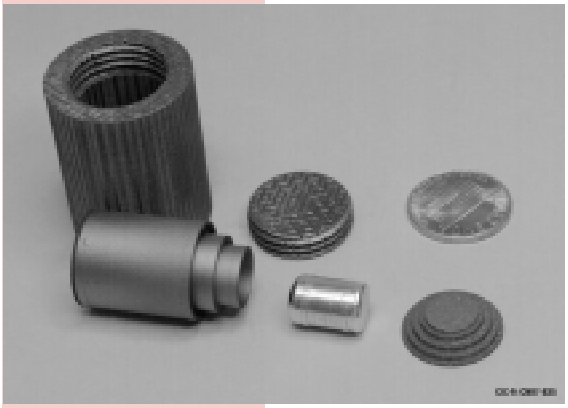


Figure 1. Welding a capsule of $^{238}PuO_2$ fuel to power experiments in space exploration. The Power Source Technologies Group is currently quantifying a ^{238}Pu aqueous recovery process to provide feed for the heat source fabrication process.

Heat source fuel was previously recycled and purified in facilities at Savannah River. For future missions, members of the Power Source Technologies Group (NMT-9) are currently qualifying a ^{238}Pu aqueous recovery process to provide feed for the heat source fabrication process. A new glove box line is currently being installed for this work, and the full-scale aqueous recovery process is expected to become operational in FY01.

The impure oxide is first dissolved in a nitric / hydrofluoric acid solution and then follows one of two routes for purification. If the feed contains only impurities that can be successfully removed by oxalate precipitation, the solution is reduced to Pu(III) using hydroxylamine nitrate as the reductant and sulfamic acid or urea as a holding agent. This solution is precipitated as plutonium oxalate by adding oxalic acid to the solution.

If the feed contains impurities that cannot be removed by oxalate precipitation, the solution is oxidized to Pu(IV) with sodium nitrite and loaded onto a nitrate anion exchange resin (Reillex-HPQ). The eluate from this process is reduced to Pu(III) and precipitated as plutonium oxalate.

Plutonium oxalate is converted to purified plutonium oxide by calcining the oxalate in the presence of oxygen enriched in ^{16}O at 750°C. The use of ^{16}O is very important in the processing of $^{238}PuO_2$. The ^{17}O and ^{18}O that are present in atmospheric oxygen have large (α,n) cross sections. The neutron emission rate of $^{238}PuO_2$ fabricated from atmospheric oxygen has a neutron emission rate of about 16,400 n / s / g ^{238}Pu (13,700 n / s from atmospheric oxygen and 2,700 n / s from spontaneous fission of ^{238}Pu). During the calcining process the ^{17}O and ^{18}O atoms are exchanged with ^{16}O atoms leading to a neutron emission rate as low as 4000 n / s / g ^{238}Pu. This lower neutron emission rate reduces personnel radiation exposure and lowers the risk of radiation damage to sensitive electronic equipment on the spacecraft.

Effluents and filtrates from the aqueous processing steps are precipitated as hydroxide cakes, and the hydroxide filtrates undergo ultrafiltration / polymer filtration before they are sent to TA-50 for disposal.

The pure oxide feed from the aqueous processing is formed into sintered granules for most terrestrial heat sources and into hot-pressed pellets for space heat sources. Most terrestrial heat sources are designed to contain the helium that builds up as a result of alpha decay. The terrestrial heat sources use a granular fuel that has a relatively low power density. They have significant free space within the capsule to accommodate the helium inventory and are heavy-walled and usually triply encapsulated to withstand the helium pressure. Space heat sources need to have high power-to-mass ratios because of the high cost of launching items into space. These heat sources are normally singly encapsulated and contain vents to let helium escape to the atmosphere.

Fabrication steps involved in forming the oxide feed into a completed 62-W_{th} general-purpose heat source (GPHS) or a light-weight radioisotope heater unit (LWRHU) are nearly identical. The most important step in obtaining the desired microstructure in the hot-pressed pellet is the sintering temperature of the <210-mm diameter granules. A total of 60% of the granules are sintered at 1100°C and the remainder at 1600°C. During the hot pressing at 1500°C, the high-fired, non-reactive granules form a microstructural skeleton, around which the more active low-fired granules sinter during pressing and post-press sintering. The graphite die that is used to contain the fuel during the hot pressing reduces the fuel to a stoichiometry of about $PuO_{1.9}$. The post-press sintering of the pellet at 1527°C oxidizes the fuel to $PuO_{2.00}$ and increases the grain size within the pellet to about 20–30 mm. This larger grain size helps prevent the formation of respirable fines during a launch accident. The sintered pellet has a density that is 85% of theoretical density and has connected porosity to assist the release of helium from its interior.

Assembly and welding of both the GPHS and LWRHU capsules are performed in helium atmosphere glove boxes. The LWRHU welding fixture is shown in Figure 2. An empty Pt-30% Rh capsule is loaded into the fixture, and a fuel pellet is placed in the capsule. Then a weld shim is placed over the fuel pellet and an end cap is placed on the capsule. The welding fixture rotates the capsule around a vertical axis in front of a welding torch held at an angle of 45° to the axis of rotation. The weld current and rotation speed are computer controlled. The entire welding cycle requires 1.9 s of arc time.

Welded capsules are then decontaminated in a nitric/hydrofluoric acid solution and subjected to numerous nondestructive tests to assure the heat sources meet specifications. Tests include helium leak testing, neutron emission rate and calorimetry measurements, radiography and ultrasonic examination of the weld, and dimensional measurements. The GPHS capsules are normally shipped to the Mound Facility for assembly into radioisotope thermoelectric generators (RTGs), but the LWRHU capsules are assembled into their final configuration for use on the spacecraft before they are shipped to the Kennedy Space Center. Figure 2 is a photo of the LWRHU assembly, which consists of the fueled capsule, three layers of pyrolytic graphite thermal insulation, and the outer, impact-resistant graphite fiber aeroshell. The complete assembly weighs 40 g. Three of these heat sources were recently used to keep electronics at normal operating temperature on the Mars Pathfinder rover, and 117 are on their way to Saturn on the Cassini spacecraft. Figure 3 is a photo of the GPHS iridium-clad capsule showing the equatorial tungsten arc weld and the iridium frit-vent at the pole, which releases helium from the alpha decay of plutonium. The Cassini spacecraft employs 216 of these heat sources to provide thermal power for the three RTGs on the orbiter.

Figure 2. The LWRHU assembly consists of the fueled capsule, three layers of pyrolytic graphite thermal insulation, and the outer, impact-resistant graphite fiber aeroshell. Three of these heat sources were recently used to keep electronics at normal operating temperature on the Mars Pathfinder rover, and 117 are on their way to Saturn on the Cassini spacecraft.

Figure 3. Photo of the GPHS iridium-clad capsule showing the equatorial tungsten arc weld and the iridium frit-vent at the pole, which releases helium from the alpha decay of plutonium. GPHS capsules are normally shipped to the Mound Facility for assembly into RTGs. The Cassini spacecraft employs 216 of these heat sources to provide thermal power for the three RTGs on the orbiter.

Nuclear Literacy:
Is It Keeping Up with Nuclear Technology?

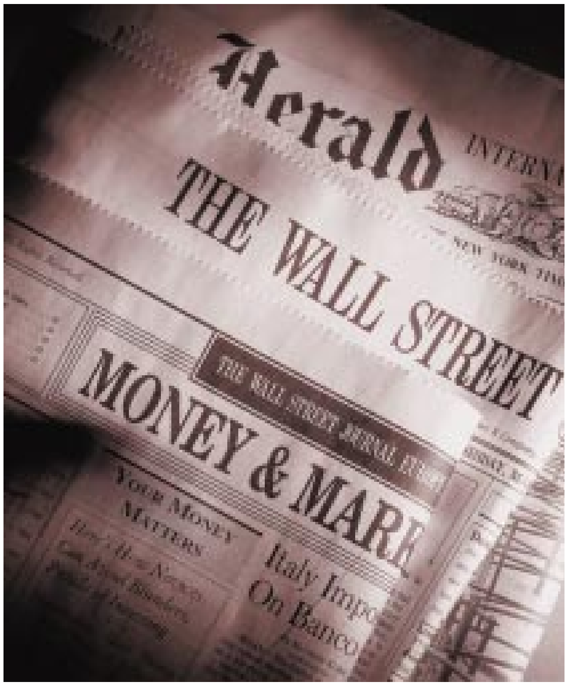
This editorial was contributed by **Kyu C. Kim,** NMT chief scientist and **K. K. S. Pillay,** Project Leader for Waste Minimization.

The recent nuclear accident in Japan illustrates a widespread nuclear illiteracy worldwide. The news media descriptions of this accident and the public's response compounded this illiteracy, further mystifying things nuclear rather than illuminating them. One is left with a sense of astonishment that this kind of accident can happen in a country that is widely known as technologically advanced.

As we learn more about the nature of this accident, newspaper accounts allege that the plant operators fed "bucket full" loads of nuclear fuels into the processing vessels. As in ordinary fire, too much fuel at one time in an undesirable configuration can cause uncontrolled burns. The threshold for the uncontrolled nuclear burn or reaction is called criticality. The nuclear energy is released in the form of highly energetic radiation instead of heat energy as in ordinary fire.

A serious consequence of an uncontrolled nuclear reaction is that it is frequently accompanied by release of radioactive debris when the reaction is not completely contained. Radioactive debris is like ashes and smoke in a wood-burning stove. The difference is that these nuclear ashes themselves are highly radioactive so that they have to be contained and disposed of by suitable methods. In addition, the volume reduction resulting from fuel to spent fuel is not very big compared to that of a wood fire, and the hazard associated with handling nuclear waste is much greater from the human health point of view.

One important aspect of a nuclear reaction in terms of criticality is that it is totally predictable. We know how much material is safe to handle in what material types, shapes, and forms. The physical environment immediately adjacent to nuclear materials also affects nuclear reactions. The radiation from interactions with surrounding materials can cause another reaction indiscriminately. Nuclear reactors worldwide are operated under conditions finely tuned to maintain a sustained level of fuel burning while taking these effects into account.



The technology of harnessing nuclear energy has been around for a few decades, and it does not fall in the realm of some exotic nuclear reactions only nuclear physicists can comprehend. Since nuclear energy is vast compared to ordinary chemical energy, the harnessing of nuclear energy has played, and will undoubtedly play, a major role in meeting the world energy demand for generations to come. Presently, nuclear energy contributes to significant percentages of world energy consumption in many developed countries. It is therefore prudent that we be more in tune with its associated technologies and require training of its practitioners in all relevant fields. Members of the public, who benefit from the technologies, need to be better informed.

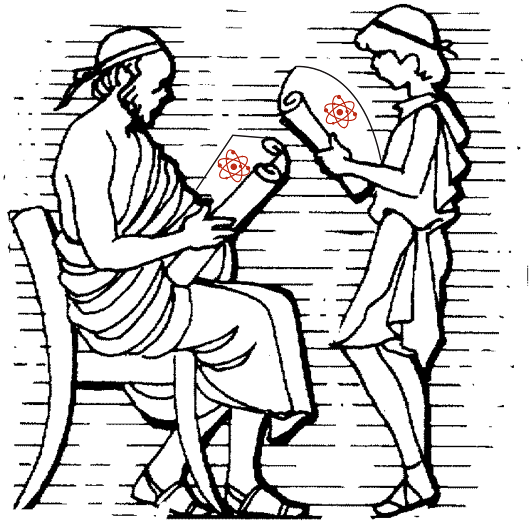
Information on peaceful applications of nuclear technologies should be shared among nations. Unknown to many, applications of nuclear technologies enhance many aspects of modern-day living. While it may never be possible for a majority of the public to learn the technology base of everything in our daily lives, it is a paramount social responsibility that the practitioners of nuclear technologies should be well trained and knowledgeable in the technologies with which they are entrusted.

Here in the United States the number of students enrolled in nuclear engineering and other nuclear science fields has been declining for some time. During the last decade, the enrollment in nuclear programs in U.S. universities has declined at all levels by 50% with the undergraduate enrollments decreasing the most (Manpower Assessment Brief Number 44, May 1999, U.S. Department of Energy). During the same period the applications of nuclear technologies in energy production, medicine, agriculture, food safety, and numerous other areas have steadily increased. The basic reason for the drop in students' interest may be the perception of horrors connected with nuclear weapons and the association of those horrors with nuclear energy and all nuclear technologies.

If we closely examine some of the nuclear mishaps of recent years, their root causes can be traced to inadequate education and training of operators and / or the operator's negligence, especially the failure to follow procedures. It is now common for certain safety cultures to rely upon operational procedures developed by knowledgeable individuals other than the operators, thus, without the experience-based knowledge of the operators themselves. In the absence of that basic knowledge, any deviation from well documented procedures or use of ill-conceived procedures will invite a disaster to occur, particularly in the nuclear business. It is highly unlikely that the type of accident that happened in Japan would happen in the Unites States. Even so, neither the industry nor the government here seems to have a strategy to remedy the current general illiteracy problem confronting nuclear science and technologies.

Astronauts are educated in basic aerospace science and are trained to operate spacecraft. Imagine them consulting manuals and procedures on the way to the moon or experimenting with various rocket thrusts to see if they can get there faster or trying to take a shortcut! We are on this Earth ship together traveling through space. Now is the time for us to learn the science and technology that will allow us to use and consume our available resources properly and safely during our journey.

> **"...it is a paramount social responsibility that the practitioners of nuclear technologies should be well trained and knowledgeable in the technologies with which they are entrusted."**



NMT-2 Responds as National Priorities Change

This article was contributed by **Stephen B. Schreiber** and **Stephen L. Yarbro** (NMT-2).

Overview of Plutonium Processing at Los Alamos

The mission of the Actinide Process Chemistry Group (NMT-2) is to effectively manage nuclear materials and supply plutonium for defense and materials disposition programs for the Department of Energy (DOE). (See Figure 1.)

NMT-2 has 70 technical personnel including 45 technicians and 25 staff members working at the Los Alamos Plutonium Facility at TA-55. They include chemists, chemical engineers, chemical process technicians, mechanical technicians, and drafting support. The technical staff are supported by a small core of project management specialists who formally track budgets, schedules, and quality.

NMT-2 currently encompasses four major technical areas: 1) providing high-quality-specification plutonium metal to the pit manufacturing program and high-purity oxide for standards, long-term storage, and disposition programs; 2) managing the vault to allow safe, secure storage of special nuclear materials (SNM); 3) providing special isotopes for dynamic experimentation testing; and 4) the proper management and disposition of secondary residues and wastes. To accomplish these objectives, NMT-2 is organized around major technology areas: pyrochemical processing; aqueous processing, which includes chloride and nitrate based aqueous recovery as well as dry powder handling; vault operations; and advanced projects.

The pyrochemical processes are performed in molten chloride salt at high temperatures. First, plutonium oxide is reduced to metal by reacting the oxide with calcium metal. After it is converted to metal, americium is removed by sparging chlorine gas through the molten plutonium metal. The americium is oxidized into $AmCl_3$, which is then absorbed and removed in a small salt blanket. The impure plutonium metal is cast into cylindrical anodes and electrorefined. The highly-pure (99.9% wt. Pu) plutonium metal is available for pit manufacturing while the secondary salt and crucible residues along with the anode heels are sent on to the aqueous operations for further recovery.

Residues are dissolved in either hydrochloric acid for pyrochemical items or nitric acid for impure oxides, ashes, and other miscellaneous compounds. After the plutonium is in solution, impurities are separated using solvent extraction or ion exchange. The solvent extraction process is a modified PUREX process in centrifugal contactors. The ion exchange process uses a polyvinylpyridine resin developed 10 years ago in collaboration with Reillex industries. The purified plutonium is removed from solution via a Pu(III) oxalate precipitation, and the remaining liquid effluents are either neutralized and discarded or reduced in volume in an evaporator. The lean evaporator overheads are currently sent to the Radioactive Liquid Waste Treatment Facility while the concentrated bottoms are fixed in Portland cement to be discarded as transuranic waste.

The PF-4 vault safely stores the majority of SNM. The vault operations are also closely tied to those of the nearby nondestructive assay laboratory, which performs measurements of the SNM before storage. The PF-4 vault maintains adequate space to receive materials from off-site and store them separately from the Plutonium Facility processing operations.

Advanced projects include the research, development, and deployment of new technologies and process unit operations to decrease waste and improve product quality. Projects include an enhanced pyrolysis unit with off-gas treatment, an acid fractionator for nitrate removal and nitric acid recovery, a glass vitrification unit to increase transuranic waste loading and minimize the final waste volume, and a low-pressure salt still for the efficient separation and recovery of monovalent chloride salts from metal oxides. In addition, an aqueous chloride processing line, CLEAR, will use enhanced organic extractants to reduce the amount of radioactivity discharged in the liquid caustic effluents by several orders of magnitude.

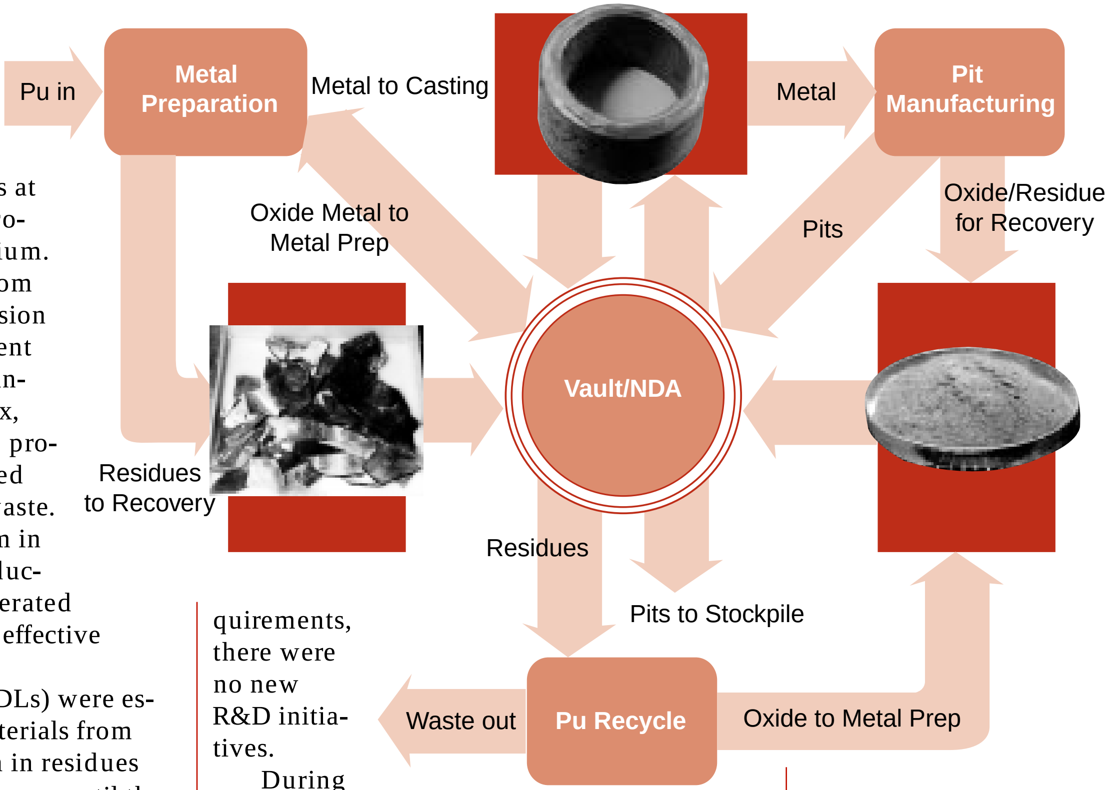


Figure 1. NMT-2 manages the division's nuclear material holdings, allowing for an efficient flow of materials into and out of the production lines. Historically the group has adapted its work in response to changing national priorities. In the future the group will continue to produce products that meet or exceed all specifications using processes that minimize waste.

Tying Plutonium "Value" to National Priorities

Historically, nuclear reactors at Hanford and Savannah River produced "weapons grade" plutonium. The plutonium was extracted from the spent fuel and associated fission products using the PUREX solvent extraction process. Because the intense radiation required complex, remotely operated facilities, this process was expensive and generated large quantities of radioactive waste. Often the value of the plutonium in the residues from weapons production exceeded that of newly generated plutonium, and it was thus cost effective to recover it.

Economic discard limits (EDLs) were established to segregate waste materials from recoverable residues. Plutonium in residues was aggressively recovered for reuse until the concentration fell below the EDL, at which point the residue was declared a waste and packaged appropriately for disposal.

Before the end of the Cold War, Los Alamos provided pure plutonium metal to support the Rocky Flats Plant operations. The capacity for metal production exceeded that of the related scrap recovery operations so only the richest residues were recovered. Lean residues that accumulated throughout this period were stored in the PF-4 vault. Supporting research and development (R&D) focused mainly on methods to improve the metal and oxide production steps. In addition, we made reactor-grade plutonium oxide feed for mixed oxide (MOX) fuel to be burned in the Fast Flux Test Facility breeder reactor at Hanford.

Transitions in Plutonium Processing Programs

By the early 1990s, no defense or civilian program needed plutonium. There was no longer a need to produce additional pure plutonium metal or oxide and no programmatic driver to recover plutonium from scrap materials. Consequently, with no processing requirements, there were no new R&D initiatives.

During this period, the DOE reduced the size of the national weapons complex, shutting down, decontaminating, and decommissioning aged facilities. Los Alamos was selected to carry on plutonium pit programs. The Defense Nuclear Facility Safety Board Recommendation 94-1 mandated complex-wide programs to stabilize the legacy scrap holdings. International treaty agreements mandated reduced amounts of available weapons grade plutonium. In response, the DOE initiated a materials disposition program with the dual approaches of immobilizing and isolating plutonium or blending it into a MOX fuel for burning in commercial or government-owned reactors. As a result of these national changes, the focus at the Plutonium Facility and in NMT-2 changed from plutonium recovery to waste stream treatment and polishing, waste minimization, and pollution prevention.

We stopped using metal production methods that generated large volumes of waste. We optimized the balance of the pyrochemical processes to minimize the amounts of secondary salt wastes by recycling reagents or switching to new ones.

NMT-2 Responds as National Priorities Change *(continued)*

For the aqueous chloride processes, we designed and produced better extractants to remove residual activity from the secondary liquid waste stream. For the aqueous nitrate processes, we developed nitric acid recycling to remove nitrates to meet ground-water discharge standards at the Laboratory outfall and to reduce residual activity in the liquid waste stream leaving the facility. We developed pyrolysis to reduce the volume of combustible rags and plastics without incineration.

Plutonium Processing Programs Today

As we approach the new millennium, the objectives for NMT-2 are again changing. The original goal of the 94-1 program was to stabilize high-risk or "high-priority" legacy residues for worker safety reasons; after three years all the high-risk items have been processed and disposed of. The focus is now on reclaiming vault storage space to allow the facility greater programmatic flexibility. We have identified approximately 700 items as potentially unnecessary for national strategic reserves. These may be consolidated and potentially disposed of as waste (transuranic or other) freeing valuable vault space. Resources are being allocated to develop new EDLs and apply them to a broader range of residue types and thus more effectively dispose of both legacy and newly generated residues.

The crucial first step in several defense programs is to supply pure plutonium metal that meets stringent quality specifications. Recent electrorefining runs have produced metal that exceeds purity and production requirements. The highly pure (99.9 wt %) plutonium metal is available for pit manufacturing. Using sealed interim storage containers has allowed the product metal to be properly stored and safely stockpiled. Unfortunately, the program funding does not concurrently support processing of the associated residues; low-grade residues are once again accumulating in the PF-4 vault.

A final objective is to supply pure plutonium oxide as feed to the Materials Disposition Program for processing into MOX fuel. NMT-2 is responsible for dissolving and purifying plutonium oxide for the demonstration lead test assembly. Process performance requirements and waste acceptance criteria are driving the current R&D in actinide science. Electrorefining is the only process that can consistently meet the exacting specification (99.9 wt %) for plutonium metal. Yet yields are averaging only 70% so residue recovery is necessary to maintain the plutonium stockpile. We will accomplish this through salt distillation and improved americium extraction techniques. Plutonium oxide purity must exceed 87.5% plutonium of a theoretical maximum of 88.2% in PuO_2. In addition, the oxide must have less than 100 ppm of any element impurity and must be a reactive (nonsintered) oxide to allow for either complete dissolution or reduction to metal. Presently, only aqueous recovery can do this. Finally, there can be no hydrogenous transuranic waste, according to the Waste Isolation Pilot Plant's very restrictive waste acceptance criteria. Waste vitrification with the associated off-gas scrubbing is being developed to address these criteria.

The Future of Plutonium Processing

Research is just beginning on the basic structure and chemistry data for the use of ionic liquids for both pyrochemical and aqueous operations. Advanced materials manufacturing might be achieved through the use of emulsion membranes for fabricating oxides with special physical properties. Similarly, metallic powders and foams might be generated for advanced metal-shaping techniques. We are investigating improved equipment and techniques for existing plutonium processing operations. Finally, we are currently looking for improvements in waste forms, waste treatment methods, and engineering models of the individual unit operations as well as their interactions in the overall process.

The bottom line is that through all the programmatic transitions, by using better technology and more reliable process controls that have resulted in numerous process improvements, the amount of waste per unit of product has steadily declined and will continue to do so. The Actinide Process Chemistry group is thus moving into the twenty-first century with the integrated goals of producing pure plutonium products while reducing secondary waste and residues and by fully complying with all regulatory and environmental constraints.

Publications and Invited Talks (September–November 1999)

Ames, R. L., F. Coriz, and E. M. Ortiz, “Aqueous Fluoride Unit Operations: Demonstration and Testing of Reducing Agents During Hydroxide Precipitation,” Los Alamos National Laboratory report, LA-13640-MS (September 1999).

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Park, J. J., J. J. Buksa, M. G. Houts, and E. D. Arthur, “Estimates of Service Lifetime of 304 Stainless Steel for Use in the Blanket region of the LANL ABC System,” *Nuclear Engineering and Desig*n **194** 151-159 (1999).

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Wayne, D. M., “Glow Discharge Mass Spectrometry: Fundamentals and Potential Applications in Stable Isotope Geochemistry,” chapter in *Handbook of Stable Isotopic Analytical Techniques*, Pier A. de Groot, ed., ElsevierScience BV, Amsterdam, the Netherlands (in press).

Newsmakers

NMT Names New Division Director

Steve Younger, Associate Laboratory Director for Nuclear Weapons appointed long-time NMT employee **Timothy G. George** as the new NMT Division Director, effective Dec. 6, 1999. George came to the Laboratory as a metallurgist in 1983. In 1993 he became group leader of the Actinide Ceramics Group. Shortly thereafter, he was appointed the Radioisotope Heat Source Program Manager and Cassini Heat Source Production Project Leader. In October 1997 George became the project leader for the Nuclear Fuels Demonstrations portion of the Department of Energy's Nuclear Fuels Technologies Project. In this capacity he coordinated efforts to demonstrate the fabrication of mixed-oxide fuels. In April 1999 he accepted the position of acting Deputy Director of the division.

The new director says, "I'm honored that upper management has placed their confidence in me. The NMT facilities, people, and programs are truly national treasures."

"The NMT facilities, people, and programs are truly national treasures."
NMT Division Director Tim George

Calls for Papers

Plutonium Futures–The Science: Submission deadline for papers is February 1, 2000, Information on the conference and on-line submission is on the Web at http://www.lanl.gov/orgs/nmt/nmtdo/PuConf2000/PuConf2000.html . The conference will be held in Santa Fe July 10–13, 2000.

American Nuclear Society–2000 Annual Meeting and Embedded Topical Meetings: Instructions and forms for preparation of paper summaries can be found on the Web at http://www.ans.org/meetings/. Completed summaries should be sent to subject category 10 (Materials Science and Technology). The conference will be held in San Diego June 4–8, 2000.

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