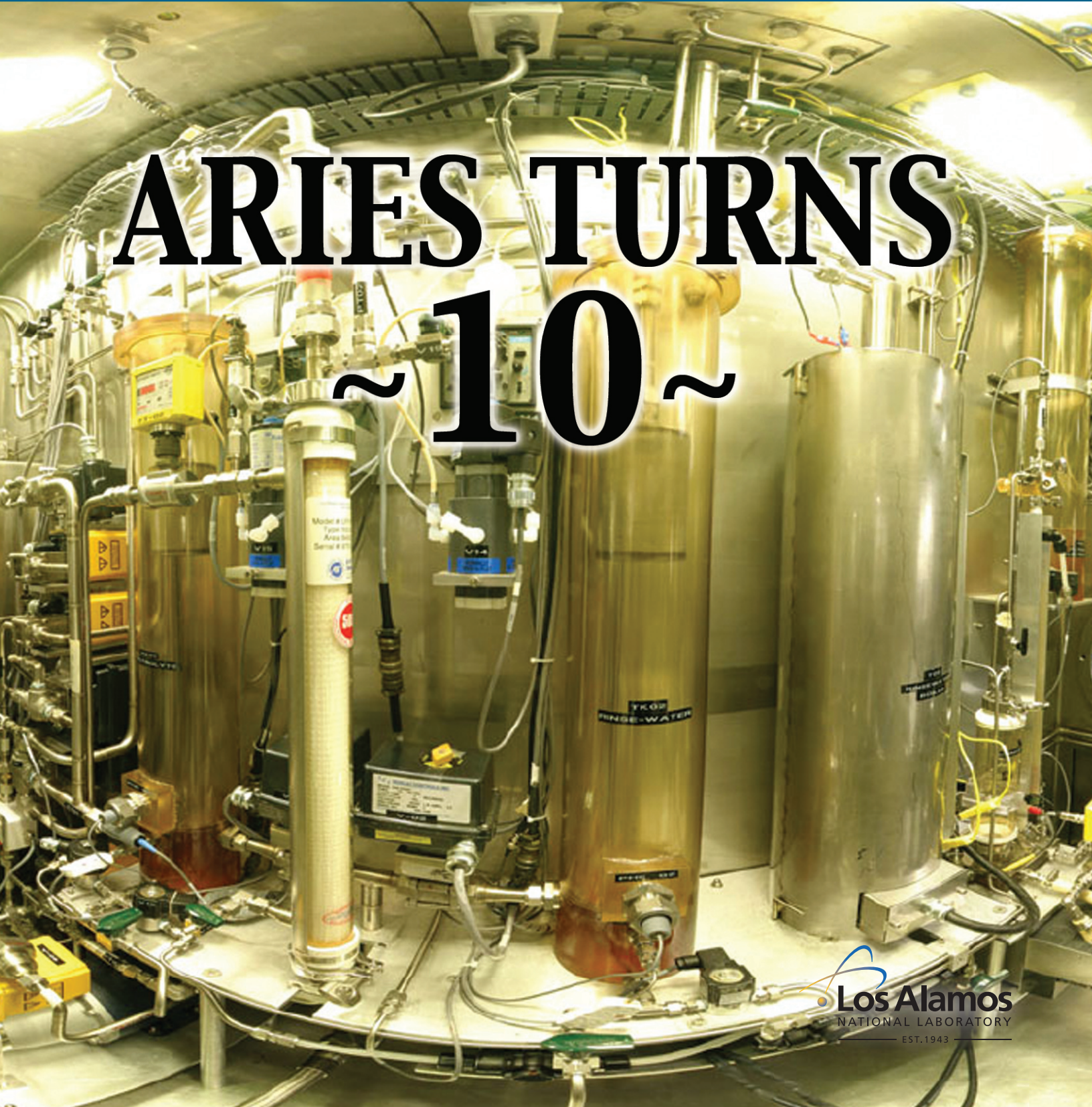


LOS ALAMOS NATIONAL LABORATORY

ACTINIDE RESEARCH QUARTERLY

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ARIES TURNS ~10~



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ARIES AT 10



This article was contributed by Steven McKee, Nuclear Nonproliferation Division.

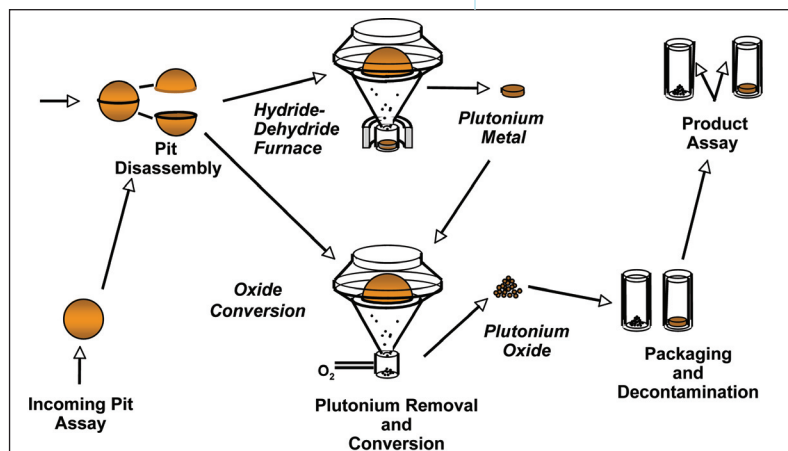
Secretary of Energy Bill Richardson dedicates the Advanced Recovery and Integrated Extraction System (ARIES) line on Sept. 11, 1998.

Ten years ago Secretary of Energy Bill Richardson toured TA-55 and cut the ribbon, officially allowing the start-up of the Advanced Recovery and Integrated Extraction System (ARIES) line. We have experienced many changes in personnel, equipment, and mission in the past decade, but the future of the technology remains bright. This issue of *Actinide Research Quarterly* highlights the science behind ARIES and celebrates the system's first decade.

Background

In 1995, President Clinton stated that the United States would “withdraw 200 tons of fissile materials from the defense stockpile, never again to be used for nuclear explosives.” In 1998, New Mexico Senator Jeff Bingaman reiterated the president’s message, stating that the United States is poised to make meaningful progress on arms control issues in the next few years, but it will depend in part on the success Los Alamos and other national laboratories achieve in addressing technical issues related to arms control.” On Sept. 1, 2000, the United States and Russia agreed to permanently dispose of 68 metric tons of weapons-grade plutonium.

In response to government commitments to reduce the U.S. stockpile of weapons-usable plutonium, the DOE announced a strategy for the disposition of surplus plutonium by irradiation as mixed-oxide (MOX) fuel in existing



The ARIES disassembly and conversion process. The process dismantles the weapon, removes the plutonium from the pit, and converts the metal into the oxide form, which is then milled, blended, subjected to physical and chemical analyses, and packaged for long-term storage.

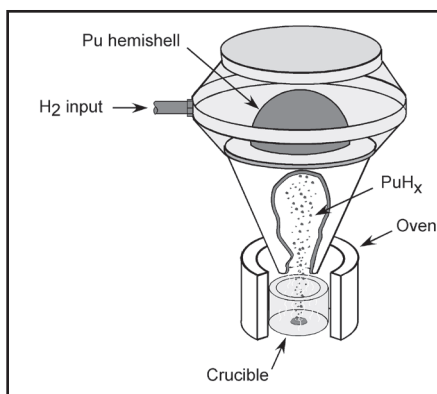
domestic, commercial reactors. The DOE has committed to providing the nation a capability to dispose of surplus weapons-usable fissile materials (see the Records of Decision for the Storage and Disposition of Weapons-Usable Fissile Materials Final Programmatic Environmental Impact Statement and the Surplus Plutonium Disposition Final Environmental Impact Statement).

In support of this mission, Los Alamos was given the task of developing the technology needed to dismantle the weapons and dispose of the excess plutonium. The result was ARIES. ARIES is being demonstrated at Los Alamos in support of the design of the Pit Disassembly and Conversion Facility (PDCF) planned for construction at the Savannah River Site (SRS) in South Carolina.

The ARIES process

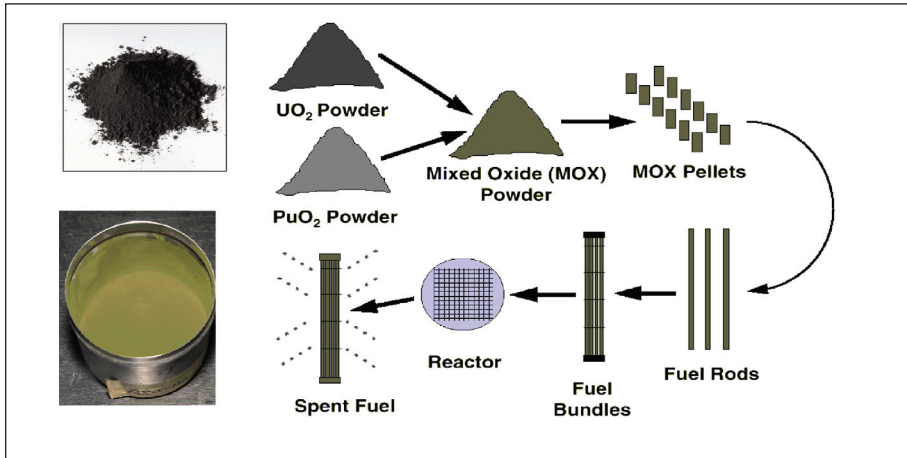
Most nuclear weapons consist of multiple explosive components that derive their power from both nuclear fission and nuclear fusion processes. The primary, or “pit,” is a machined plutonium sphere that is detonated via nuclear fission to trigger a far more powerful secondary fusion reaction. The ARIES process focuses on the pit, and uses eight modules: pit disassembly (PDIS), direct metal oxidation (DMO), packaging (ICAN), nondestructive assay (NDA), hydride/dehydride (HDH), parts sanitization (PSF), highly enriched electrodecontamination/conversion (HEU), and special recovery (SRL). These modules dismantle the weapon, extract the plutonium from the pit, and convert the metal into the oxide form, which is then milled, blended, subjected to physical and chemical analyses, and packaged for long-term storage. The oxide form is preferred for long-term storage because it is relatively stable. It is also easy to convert into a MOX fuel for use in nuclear power plants.

The use of former weapons-grade plutonium in MOX nuclear fuels has an additional benefit: it results in a final form (spent nuclear fuel) that is very



A schematic of the hydride-dehydride or hydride-oxidation chamber for converting excess weapons plutonium. In the hydride-dehydride process, hydrogen (H_2) is continuously recycled. The hydride falls into a crucible that is heated, driving off H_2 gas, and leaving molten plutonium in the crucible. The H_2 refluxes to the top of the chamber, where it removes additional plutonium from the component. For conversion to an oxide, the crucible is not heated, and either oxygen (O_2) is admitted and the hydride burned to release H_2 and leave an oxide powder, or nitrogen (N_2) is used and burned to a nitride powder. The nitride can then be converted to the oxide by burning with O_2 .

From “Plutonium,” Chapter 7, The Chemistry of the Actinide and Transactinide Elements, D. Clark, S. Hecker, G. Jarvinen, and M. Neu, Springer, 2006.



Fabrication of mixed-oxide (MOX) fuel using uranium (top left) and plutonium oxide (lower left).

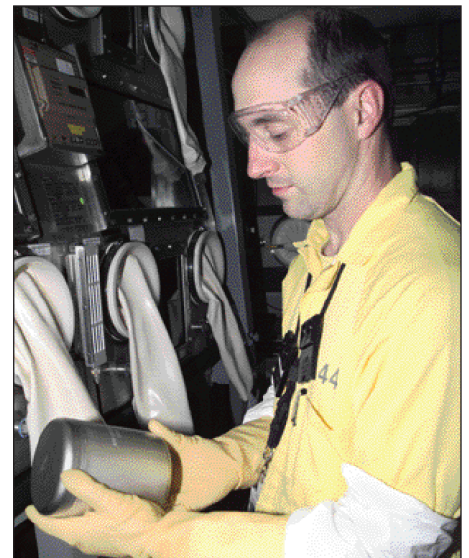
proliferation resistant. In other words, spent nuclear fuel is extremely difficult to reconvert into a form that can be used in another nuclear weapon.

The operations within ARIES have produced significant results. To date, ARIES has executed two technology demonstration runs. These have demonstrated the applicability and versatility of the process used to dismantle a wide variety of nuclear weapons and efficiently convert the plutonium into plutonium oxide. The processes associated with pit disassembly, while classified, have yielded significant information to support the design of the PDCF. This element of the program represents significant technology transfer, and provides extensive process validation. ARIES was also designed to incorporate processes that generate very little waste, and permit the application of automation. Automation is highly desirable for ARIES processes, as it significantly decreases the potential for radiation exposure to workers.

The future

The U.S.-Russia agreement, first negotiated and signed during the Clinton Administration, has subsequently been reaffirmed during the Bush Administration and is still advancing, despite the political realities that have evolved since the end of the Cold War. Since the agreement was signed, there has been significant progress in the overall plutonium disposition program, and many of its most positive and tangible elements are the direct result of the work performed at Los Alamos.

As for the future, the ARIES platform is being upgraded for a final technology demonstration. Los Alamos will then initiate a significant production mission to produce plutonium oxide to feed the start-up of the MOX Fuel Fabrication Facility (MFFF). Currently under construction at SRS, the MFFF will be operational in 2017. The PDCF is being designed and is planned to be operational in 2022. Internationally, the Russians have embarked on a disposition pathway that will remove 1.5 metric tons of plutonium annually, starting in 2012.



Automated packaging of special nuclear material allows for significant reductions in worker dose and greater safety.



THE ARIES ROBOTIC LATHE

This article was contributed by Wendel Brown, Yvonne Rivera, Ernie Montoya, and Danny Gallant, Plutonium Manufacturing and Technology Division.

The ARIES robotic lathe (ARL) is used in the first step of the plutonium disposition line: pit disassembly (PDIS). This first process dismantles and separates metal parts into individual components for declassification and conversion. The parts include both special nuclear material (SNM) and non-SNM DOE Standard 3013 storage cans and other metal items.

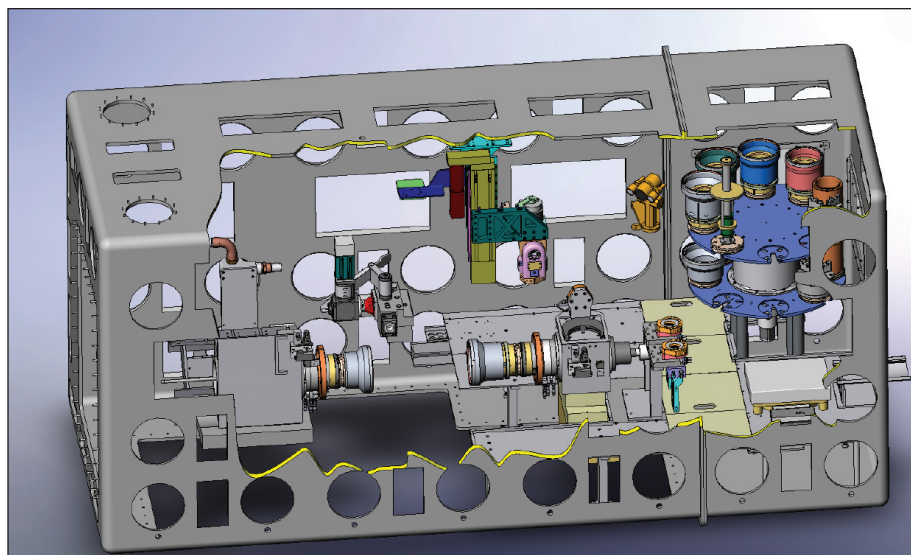
The ARL is used for a variety of machining tasks within the glovebox enclosure. The primary task is to bisect and disassemble pits into hemishell subassemblies as a first step in the extraction of SNM. In addition, the ARL is used to cut open material-storage containers and for other general machining tasks.

The ARL, which was manufactured for Los Alamos by the Moore Tool Co. in Bridgeport, Conn., consists of a robot with a custom-made lathe machine tool. The technology significantly decreases the amount of manual operations performed and uses improved cutting tools to minimize both the generation of fine material and worker exposure, as compared with methods and equipment previously used. A major goal of the robotic lathe is to demonstrate that the dose to the worker can be reduced by a factor of 10 through the application of automation and robotics.

The robot performs many material-handling operations with minimal human interaction. The robot loads items onto the lathe and can aid in dismantling according to item-specific procedures. The robot also can be used to move items to the trolley tray. Several safety features facilitate automated lathe operations. For example, glovebox gloves could easily be torn or breached if they got caught in the robotic arm, the lathe, or the cutter. In the ARL, glovebox gloves are protected by a light beam interlock that automatically stops all operations if a light beam in the plane of the gloveports is broken.

Some operations still require a modicum of human intervention, depending on the type of item being machined and the type of work being carried out.

Schematic of the ARIES robotic lathe and glovebox.



For example, one expected manual operation involves fine adjustments to the vacuum seating of the pit in the chuck when pit disassemblies are carried out.

Two primary tools, a beveled-edge rotary parting tool and a milling tool attachment, are used for cutting pits and/or containers. The main component of the rotary parting tool is a hardened cutting wheel similar to what you would see on a metal pipe cutter or pizza cutter. A small amount (about 150 milliliters) of hydraulic oil is applied to help hold the tools on the lathe. The cutting wheel penetrates the environmental shell of the pit with a maximum of 1500 pounds of force. The rotary parting tool is driven inward automatically toward the center of the pit to a depth predetermined by the item-specific procedure. The purpose of the rotary parting tool is to bisect the pit while minimizing cutting waste.



Milling tool attachment mounted in the tool post inside the glovebox.

A milling tool attachment also can be used to cut pits and other SNM containers. For this milling attachment, the item spins very slowly, at about 3 revolutions per minute (rpm), while the milling tool spins up to 2000 rpm and cuts into the item to a predetermined depth.

A vacuum system located directly below the cutting area is used for collecting lathe turnings and chips. A cyclone separator located between the vacuum nozzle and the vacuum source aids in the separation of metal pieces. This collection system is necessary; the Laboratory's operational philosophy is to account for the total mass of an item as it enters a process, as well as the total mass of an item's separate components as they exit the process.

The robot is operated by an Aerotech U600 Controller System, which is a personal-computer-based machine control and monitoring system. The U600 controls the lathe and the robot's movement and monitors the condition of

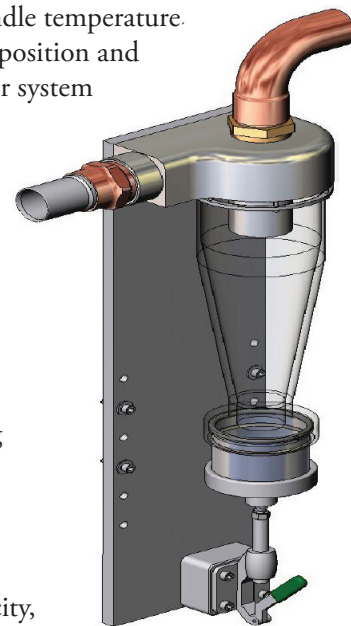
Schematic of chip collection and cyclone separator (shown at right).

critical lathe operation parameters including spindle temperature, vacuum quality on head- and tailstocks, and the position and status of the gripper and the robot. The controller system uses software profiles for each robotic operation, from tool picking through various lathe operations, and has a built-in software-profiled safe corridor in which the robot can be operated.

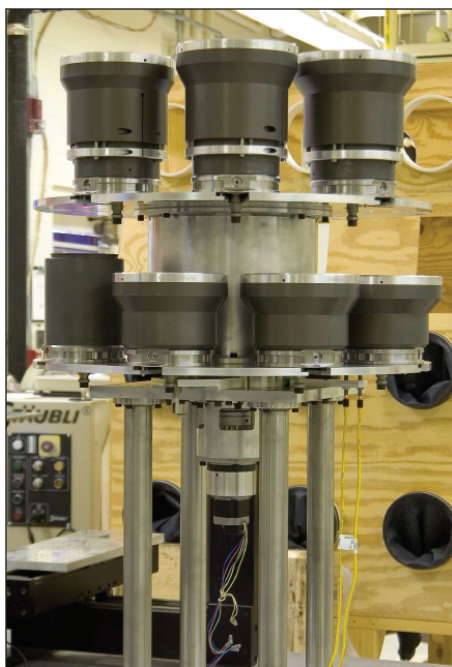
Before initiating robotic lathe operations, the operator preprograms the software profile. While operating the robot, the operator observes the process and can stop (E-stop) the operation at any time. Once the E-stop button is pressed, the operator can restart the robot without having to restart the software controller program from the beginning. The same capability exists if the plane of a gloveport covered by the light-beam interlock is broken inadvertently.

Because of limited in-line SNM storage capacity, a 16-station fixture storage carousel was designed to contain the pit-holding fixtures and the tooling for holding DOE Standard 3013 storage containers. Many of these fixtures weigh as much as 15 kilograms and are difficult to manipulate by hand inside the glovebox. The robot and carousel are programmed to mount and dismount these fixtures on the lathe without the intervention of the operator. All of these capabilities will significantly reduce worker occupational dose and the potential for worker injuries, as well as contribute to improved ergonomics during disassembly operations.

Funding for the development, procurement, and installation of the ARL module has been provided by DOE's Office of Fissile Materials Disposition (NA-26) with NNSA.



Novel 16-station fixture storage carousel.



DECONTAMINATION AND CONVERSION OF HIGHLY ENRICHED URANIUM

Throughout the DOE Complex there are holdings of highly enriched uranium (HEU) contaminated with plutonium and americium. Contaminated HEU cannot be sent to the Oak Ridge Y-12 National Security Complex in Tennessee, the nation's uranium disposition site, until its surface is cleaned to a smearable transuranic (TRU) alpha activity level of below 20 disintegrations per minute (dpm)/100 centimeters squared (cm^2) or to an oxide contamination limit of 60–210 parts per billion (ppb) TRU. Together, the Plutonium Manufacturing Technology and Applied Engineering Technology Divisions at Los Alamos have developed and deployed advanced methods for the decontamination and conversion (D&C) of HEU.

The D&C process

The first stage of the D&C process, electrolytic decontamination, dissolves uranium and TRU contaminants from the surface of HEU parts. The process is similar to the common industrial practice of electropolishing and is accomplished by passing a constant current from a stainless steel cathode to the HEU anode through an electrolyte solution. The dissolved contaminants are subsequently removed from the solution by precipitation and filtration. The surface contamination on the HEU parts can be reduced by this method to below either of two acceptance criteria: 20 dpm/100 cm^2 TRU contamination or TRU activity less than one-fiftieth of the uranium alpha activity. Initial experiments were run on a fully manual electrodecontamination system.

The second stage of the D&C process involves direct metal oxidation (DMO) and blending to produce Y-12 acceptable oxide. Oxidation is performed in a static box furnace with no supplied oxygen. This setup, although not optimized, allows for a slow, steady oxidation of uranium metal to U_3O_8 and avoids the risks associated with a supplied oxygen system.

ARIES demonstration review

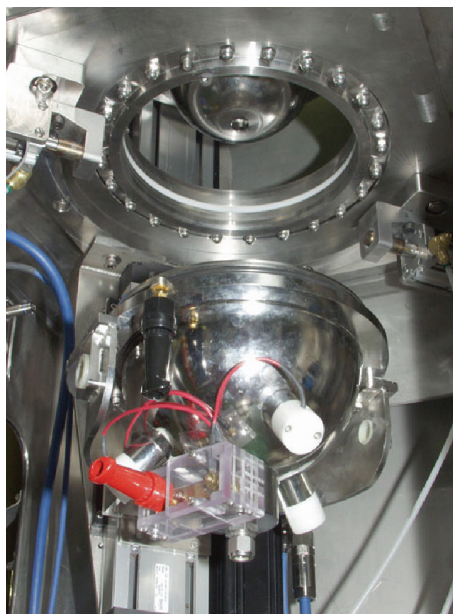
The ARIES HEU decontamination and conversion process is proven to generate Y-12 acceptable material. Demonstrations of the D&C module, performed with all-manual instrumentation and material transfer, validated the D&C processes and supported the development of bounding Pit Disassembly and Conversion Facility (PDCF) production rates. To date, Los Alamos has hot-tested an experimental D&C system that is functionally equivalent to the system designed for the PDCF. This includes testing of a fully manual flow loop and two different clamshell fixtures.

Results of the HEU decontamination tests indicate that the process can reliably meet the Y-12 acceptance requirements for bulk HEU oxide. Los Alamos has demonstrated that HEU can be oxidized in a static furnace, but no rate or product purity data are available for the PDCF-style conversion furnace.

This article was contributed by David Costa, Brad Schake, Minnie Martinez, Phillip Banks, Jim Rocha, and Mannie Valdez, Plutonium Manufacturing and Technology Division; and Kirk Weisbrod, Applied Engineering and Technology Division.



Panoramic view of the interior of the Auto-HEU glovebox.



Fixture overview with upper and lower bowls and glovebox central section.

The first and second ARIES demonstrations have been run with non-prototypic decontamination fixtures. The decontamination fixture used in the second ARIES demonstration was designed for manual operation. It required rotation for draining, manual opening and closing, did not have any features to prevent cross contamination of material, and did not allow for selective decontamination of the inner or outer hemishell surfaces.

Ongoing development and testing

The results of ARIES demonstration tests revealed deficiencies in both the components and in the process maturity of the manual HEU system. These gaps in mechanical maturity represent a troubling level of risk to the PDCF facility. Because HEU processing is a low-dose operation, hot testing is not required to close the component maturity gaps.

The Auto-HEU system is designed to mitigate the problems of cross contamination between the clean part and the plutonium-contaminated region of the glovebox and to allow selective decontamination of the inner and outer hemishell surfaces. The system components include the fixture, the supporting automated flow loop, and the solution cleanup systems. This system is designed to test the PDCF fixture and includes the split hot–cold design, electrical isolation of the inner and outer cathodes, a retractable dip tube, and Lexium drive motors and controls. The system is integrated with the prototypic conversion furnace. Flow-loop operations will be automated by a programmable logic controller, and all flow rates to and from the fixture will mimic those expected in the PDCF.

The Auto-HEU system has been thoroughly cold tested. Fixture testing included mechanical testing of the Lexium drives for the upper and lower bowls, helium leak testing of the closed fixture, repeated pressure cycling of the fixture to test structural deflection and locking pin loads, and tests of the electrical isolation of the upper and lower bowls. Acceptance testing also validated the automated flow-loop operations.

The first and second ARIES demonstrations validated the D&C concept through the production of Y-12 acceptable U_3O_8 . Demonstration testing also revealed deficiencies in the manual system in a production environment. The Auto-HEU system is designed to correct many of the difficulties associated with manual operation. Implementation of these design features into the PDCF design will allow for the reliable production of Y-12 acceptable uranium oxide on a production scale.

DIRECT METAL OXIDATION: A PROCESS-ORIENTED APPROACH

The furnace system used in the direct metal oxidation (DMO) module was developed to oxidize plutonium, bonded plutonium, and highly enriched uranium (HEU) hemishells to produce oxide that meets specifications at conversion rates for Pit Disassembly and Conversion Facility (PDCF) throughput requirements. The furnace design has evolved over time to become a system that is automated to minimize operator handling of metal and oxide.

The DMO-2 furnace body is made of stainless steel and the overall design features a top-loaded upper furnace section holding a rotating basket. The basket is loaded manually outside of the furnace and then loaded into the furnace by means of an overhead hoist. The lid is placed on top of the furnace to provide containment. It is sealed with a high-temperature fluorocarbon elastomer O-ring seal. The weight of the lid and a negative operating pressure with respect to the glovebox environment provide a sealing pressure on the O-ring. No mechanical clamping forces are used, thus the seal is immediately broken if the internal furnace pressure rises much above glovebox pressure.

An auger located in the lower furnace moves oxide into the calciner. Another auger section, directly linked to the furnace auger, transports oxide through the calciner. Both of the auger sections, calciner tube, basket assembly, and sprocket and axle assemblies are made of a nickel–chromium alloy that resists oxidation and corrosion at high temperatures.

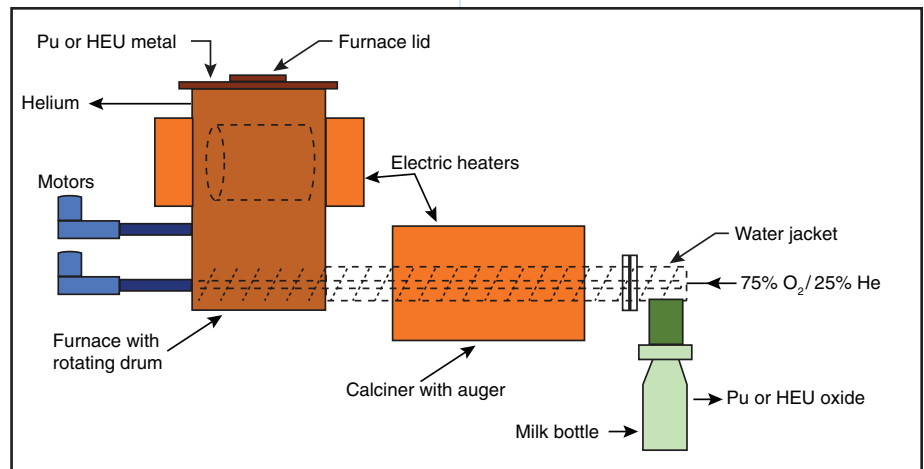
Plutonium and uranium both burn at nominally the same rate. Plutonium oxidation is dependent upon metal surface area or particle size, oxygen concentration in the reaction zone, temperature, alloying, and surface conditions such as oxide coating and surface imperfections. Plutonium metal at room temperature will form an oxide coating, but it will not normally ignite and burn.

Plutonium ignition

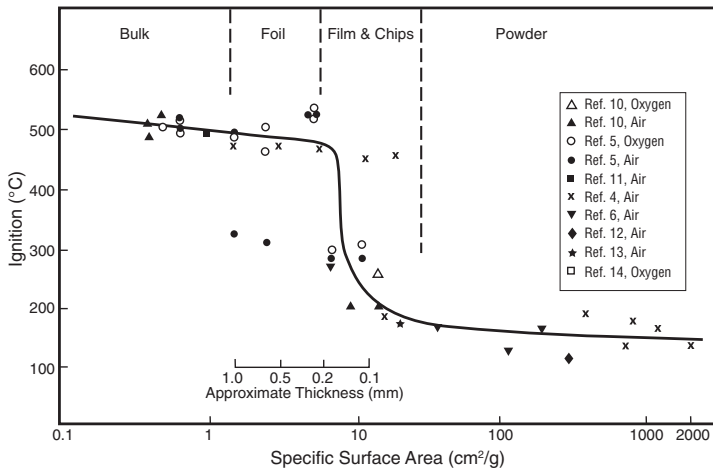
Ignition occurs when plutonium metal begins to rapidly oxidize. Ignition temperature for metal pieces in air is typically above 475 degrees Celsius (C). Several variables affect ignition: surface area, alloying, surface condition, oxygen concentration, mechanical stress, and heat transfer. A brief description of the variables follows.

Surface area: Plutonium chips and powder ignite at lower temperatures than do bulk or foil samples because of the greater surface area of the individual particles.

This article was contributed by Elizabeth Bluhm, Kane Fisher, Kenny Martinez, and Lowell Christensen, Plutonium Manufacturing and Technology Division; and Mike Keddy, Applied Engineering and Technology Division.



DMO-2 schematic showing basket, augers, and water-cooling system.



The effect of surface area on plutonium ignition temperature. From Plutonium Pyrophoricity, J. Stakebake, RFP-4517.

Alloying: Depending on the material alloyed with plutonium, the ignition temperature can either be raised or lowered. The addition of 2 atomic percent gallium increases the ignition temperature by approximately 30 degrees C. Adding nickel or cobalt lowers the ignition temperature by 30–40 degrees C. Unalloyed plutonium and some alloys oxidize much faster and with more spallation of oxide particles from the surface than the plutonium/gallium alloy. The higher the oxidation rate, the lower the ignition temperature of the sample.

Surface condition: Plutonium dioxide on the surface retards oxidation until oxide spallation begins.

Oxygen concentration: Metal coupons, or test specimens, ignite at 510 degrees C in 10% and 20% O₂ but do not ignite in lower oxygen concentrations. The

ignition temperature falls below 450 degrees C when the metal is oxidized in pure O₂.

Mechanical stress: Internal stress in the metal has been shown to reduce ignition temperature. Mechanical stress can create fractures in the oxide layer.

Heat transfer: Heat transfer away from the plutonium metal results in higher heat input being required for ignition. Once plutonium ignites, the oxidation rate slows and eventually stops if there is sufficient net heat flow away from the reaction.

Plutonium oxidation

When plutonium oxidizes at high temperature, the metal appears very bright and glows like burning charcoal. Oxidation rate is strongly dependent on temperature. Plutonium metal oxidation is an exothermic reaction in which 1 kilocalorie (kcal) of heat is released per gram of plutonium. Plutonium metal and oxide absorb some of this heat, while the remainder is released to the environment.

As the temperature of the metal increases, its oxidation rate increases exponentially, releasing more heat. Initiation of the self-sustaining oxidation in the DMO furnace is indicated by a decrease in percent power needed to maintain the upper furnace set-point temperature. A noticeable drop in furnace temperature indicates that oxidation is complete. This drop in temperature is followed by a gradual increase in percent power needed to maintain the oxidation furnace set-point temperature as the amount of metal to oxidize decreases until the amount reaches zero.

The equilibrium temperature at which plutonium oxidizes depends on a balance between heat generation and heat dissipation. Heat generation is controlled by factors that influence the oxidation rate, such as O₂ availability and temperature. Heat dissipation is controlled by normal heat-transfer mechanisms, such as

conduction, convection, and radiation. Bulk oxidation of the metal in air (or O_2) is controlled by diffusion of oxygen or oxygen-containing species through an adherent and coherent oxide layer present on the metal surface.

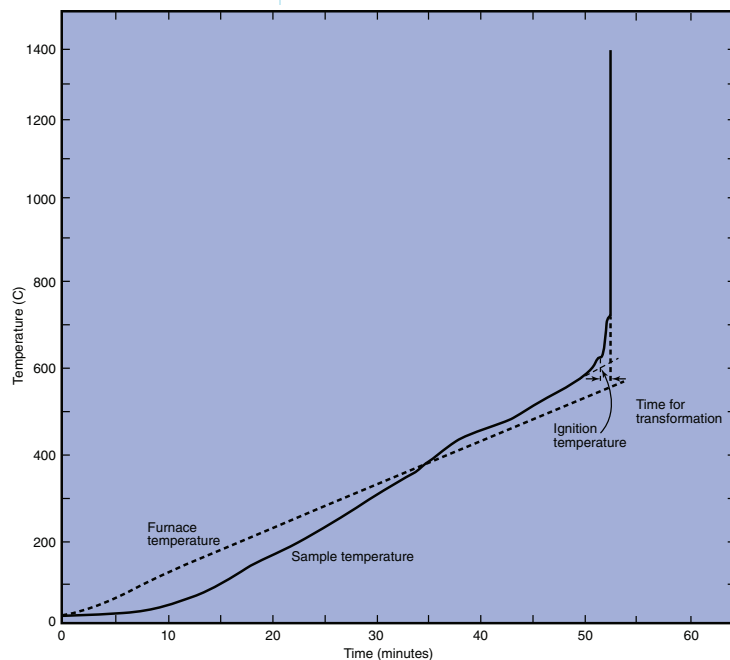
A typical DMO run consists of heating plutonium metal between 475 and 575 degrees C, at which point it ignites and begins to oxidize when a mixture of 75% O_2 and 25% helium (He) flows into the furnace at about 2 liters per minute (L/min). This temperature range prevents plutonium from melting, which occurs at 640 degrees C. Exact temperature of the metal surface is not known, but it is kept below the melting temperature of 640 degrees C. If 100% oxygen is used, the reaction is rapid, and the plutonium may develop hot spots and localized melting because of the exothermic reaction. If low concentrations are used, the reaction is not self-sustaining.

Recent ARIES tests showed that the 75% O_2 /25% He mixture worked well to produce plutonium oxide at a controlled rate. Once the furnace temperature reached ignition temperature, O_2 was slowly added to the furnace at about 2 L/min, and the flow continued until the reaction was complete and the oxide was augered through the calciner.

Total O_2 quantities were well in excess of the stoichiometric amount to convert plutonium to plutonium oxide (PuO_2). Plutonium oxidation is exothermic, but the energy produced varies as the reaction progresses and there is less metal to oxidize. After plutonium ignition and oxidation, furnace power typically drops from 100% to about 50% and then gradually increases to about 70% as the mass of plutonium oxidizing decreases at the end of the run. As the last of the plutonium is converted to oxide, the furnace temperature begins to drop. The temperature-control system increases heater power to maintain temperature. An increase in heater power serves as an indicator that the reaction is near completion.

Uranium oxidation

Factors that affect the ignition and oxidation of plutonium also affect uranium. However, there are differences in ignition and oxidation behavior. Uranium has an ignition temperature of almost 600 degrees C, at which temperature it oxidizes rapidly, with the temperature rising exponentially if there is sufficient oxygen to sustain oxidation. However, uranium can be ignited in a heated furnace at about 350 degrees C and oxidized in a controlled reaction at temperatures between 400 and 600 degrees C. Uranium in O_2 shows unique ignition behavior: above 400 degrees C, the temperature of the uranium sample



The unique ignition behavior of uranium in O_2 . From *Ignition Behavior and Kinetics of Oxidation of the Furnace Metals, Uranium, Zirconium, Plutonium, and Thorium, and Binary Alloys of Each*, J. Schnitzlein, et al., ANL-5974.

exceeds the furnace temperature. Uranium is oxidizing at this point, but it is not a self-sustaining reaction. As with plutonium, if there is sufficient net heat flow away from the uranium-oxidation reaction, the oxidation rate will slow and eventually stop.

Under controlled temperatures between 400 and 600 degrees C, HEU burns at nominally the same rate as plutonium. The HEU temperature must be controlled after ignition so that it does not reach temperatures at which the DMO furnace would be unable to remove enough heat to control the reaction. Just as in burning plutonium, the furnace internal temperature is controlled by cutting heater power as the heat of reaction increases. The flow rate of O₂ to the furnace may need to be shut off if the temperature gets too high.

The Pit Disposition Science and Technology Group (PMT-4) and the ARIES program recently demonstrated HEU oxidation using DMO-2 equipment. HEU was heated between 475 and 575 degrees C, where it ignited and oxidized with an addition of flowing 75% O₂/25% He mixture. The final U₃O₈ product was augered into the “milk bottle” at the end of the furnace and transported to another glovebox for packaging and shipment to Y-12.

Oxidation rate

Observed conversion rates for metal oxidation during ARIES tests are affected by several factors: metal characteristics, preheating, oxygen pressure, ignition temperature, oxidation temperature, oxide coating, and mechanical stress and imperfections.

Metal characteristics such as mass and mass/surface area ratio: Pieces of metal have different masses and shapes, and their ignition and oxidation behavior are different; the higher the mass/surface area, the harder it is to ignite and burn.

Preheating: A nominal 20-minute soak time allows the plutonium metal to heat evenly so that there will be no hot spots when oxidation begins. The soak time during ARIES testing was timed, starting when the temperature was 90% of ignition temperature. Soak time is essentially a set limit to ensure the metal “equilibrates” under a high temperature.

Oxygen partial pressure: O₂/He gas combined with oxidation furnace pressure control establishes a concentration of oxygen in the furnace that supports oxidation.

Ignition temperature: The ignition temperature is a limit below which oxidation will not be self-sustaining.

Oxidation temperature: The higher the temperature, the higher the oxidation rate, but the temperature must be controlled to prevent melting, in the case of plutonium, and to prevent rapid temperature rising, in the case of uranium.

Oxide coating on the metal and the spallation rate of this oxide layer: Plutonium burns and forms an oxide coating, which has been shown to inhibit further oxidation. Oxide will spall, with or without agitation, but the DMO

furnace has been designed with a basket that rotates to enhance spallation from the metal pieces.

Mechanical stress and surface imperfections in the metal: Internal stress in the metal has been shown to reduce ignition temperature. Mechanical stress can create fractures in the oxide layer.

In the second ARIES demonstration, the observed conversion rates varied from run to run, but the effect of each of these factors in contributing to the variation is not clear. Experimental test plans for the third ARIES demonstration have been developed in which key operating parameters will be bounded with more certainty.

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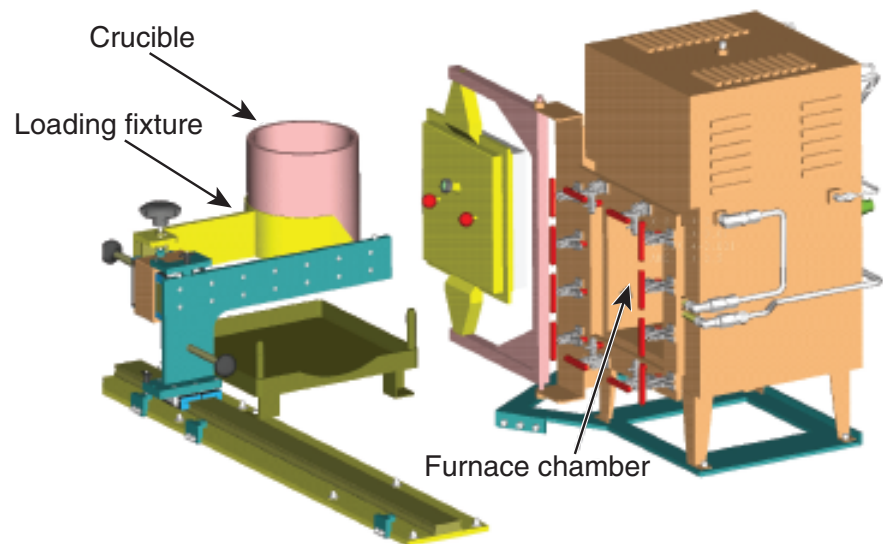
Plutonium Pyrophoricity, J. Stakebake, Rocky Flats Plant, RFP-4517, June 2, 1992.

REACTION MELTING IN THE PARTS SANITIZATION FURNACE

This article was contributed by Rene Chavarria and David Kolman, Plutonium Manufacturing and Technology Division.

Besides producing 34 metric tons of plutonium metal, weapons disassembly will also produce a stream of stainless steel, aluminum, beryllium, and other metal parts that must be destroyed. The disposition of these nonnuclear byproducts will be handled by the parts sanitization furnace (PSF). The PSF team is developing the process technology that will be used to dispose of the nonnuclear material weapon components.

The chief technical challenge for PSF is the elimination of the innate classified information of each metal component. Classified information can reside in the part geometry, its mass, and its composition. Furthermore, any evidence of the metallurgical processing performed on the part may also be classified. Parts sanitization will remove all classified information from the parts by using an inexpensive, commercially available high-temperature furnace to melt the classified pieces.



Schematic of the parts sanitization furnace.

Design

Several alternative methods, such as grinding, shredding, and dissolving, were investigated before the ARIES project team chose melting as the least expensive and least difficult solution. Melting provides the cheapest, safest, and most reliable sanitization. Several technologies are used to melt metals in industrial settings, including resistance heating, microwave heating, and induction heating. Of the variety of melting technologies available, we chose to implement a molybdisilicide element resistance furnace for parts sanitization.

Resistance furnaces work by running current through inefficient conductors that act as the heating elements. The heating elements, which can reach temperatures up to 2000 degrees Celsius (C), radiate energy inside the furnace and heat anything inside it. This is very different from what happens in induction or

microwave furnaces, in which the energy transfer is accomplished by some form of coupling.

In induction and microwave furnaces, the material being heated must first have a favorable geometry. It must also be conductive or susceptible to heating by microwaves. For both microwave and induction furnaces, the technical development needed to situate a functioning furnace inside a glovebox was deemed too time consuming and costly. By contrast, resistance furnace technology is relatively simple, cheap, and well characterized, so it was favored for installation in the ARIES glovebox line.

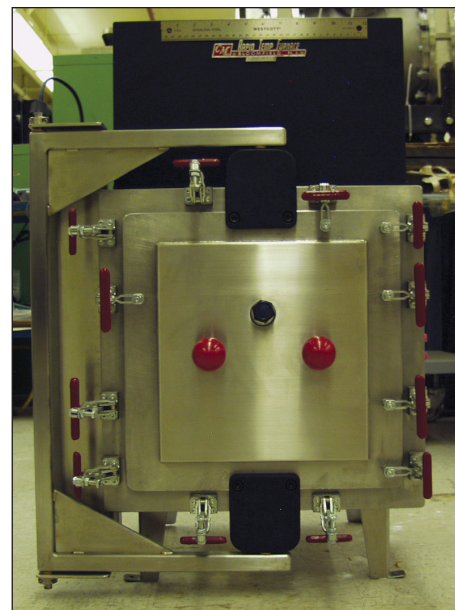
Installing a furnace that can reach temperatures of up to 1600 degrees C in a glovebox within a Category 1 nuclear facility is not a simple task. Although we were able to procure a standard off-the-shelf furnace, some modifications were necessary for better glovebox functionality. A cooling water-flow interlock and a glovebox over-temperature interlock were added to the basic furnace. Because floor space and glovebox space are very valuable inside the Plutonium Facility (PF-4), we repackaged the power supply and process controllers into a smaller cabinet to minimize the system footprint. We also designed a new crucible-handling aid that allows workers to safely handle loads of up to 40 pounds inside the glovebox, greatly facilitating the loading and unloading of crucibles from the furnace.

Technical challenges

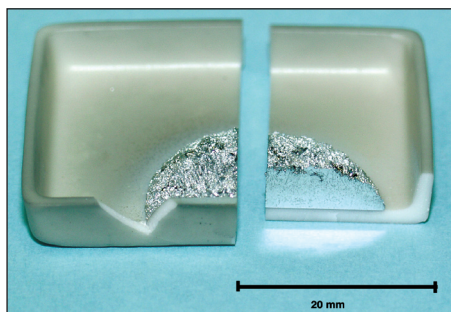
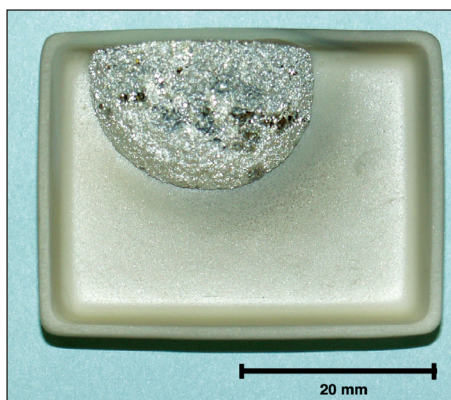
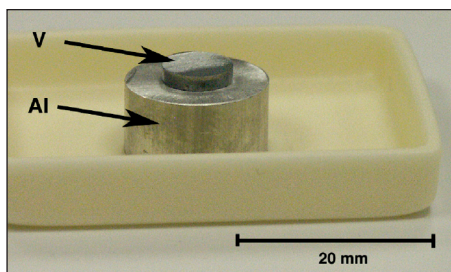
We also had to develop each of the metal-specific sanitization processes for use in PF-4. In many ways, this was the most challenging aspect of the work. For example, the dismantling process creates quantities of vanadium, a metal with a very high melting point (1910 degrees C). Vanadium sanitization in the PSF presents challenges because the maximum temperature achievable within the furnace is 1600 degrees C. One possible solution is to lower the melting point of vanadium.

Due to the complexity of ternary (three-metal) systems, the investigation focused on binary (two-metal) systems. One possible solution was to co-melt vanadium with another metal that has a lower melting temperature. Unlike a eutectic reaction that depends on a single, very narrow composition range to lower the melting point, co-melting works in a wider range of compositions at a given temperature. This is an important detail because it produces a more predictable process. In a eutectic reaction, a small change in composition could result in a very large change in the melting temperature. Any errors could leave parts incompletely melted, and partially melted parts would still contain classified information.

Co-melting works by dissolving the solid phase into the liquid molten phase. Industry has used this type of reaction melting to successfully melt other refractory metals such as titanium and nickel. However, the dissolution reaction is so exothermic that care must be taken to control the rate of the reaction.



Front view of the furnace.



From top to bottom: Alumina crucible with vanadium (V) and aluminum (Al) samples; co-melted Al and V at 1300 degrees Celsius (C) for 4 hours; sectioned co-melted Al and V at 1300 degrees C for 4 hours.

Candidates for the co-melting process must lower the melting temperature at least 350 degrees C and must have a melting temperature lower than 1600 degrees C. A literature search revealed three practical candidates that already required sanitization: aluminum, nickel, and uranium. Nickel is a component in the Type 304 stainless steel parts that are also part of the PSF waste stream. The melting temperature of 304 stainless steel is between 1400 and 1450 degrees C; thus, according to our criteria it is a possible candidate for co-melting with vanadium. Uranium, with a melting temperature of 1135 degrees C, was also considered. We chose aluminum in the end, however. Of the three metals we considered, aluminum has the lowest melting temperature (660 degrees C).

The primary reason for selecting a metal with a melting temperature below 1600 degrees C is to obtain a solid/liquid interface. Having a solid/liquid interface increases the reaction surface area by having the molten metal liquid completely wet the solid vanadium. The phase diagrams from the literature clearly pointed to the existence of aluminum and vanadium liquid phases at temperatures below 1600 degrees C. However, we lacked data on the kinetics, or speed, of the predicted dissolution reaction. Maximizing the reaction surface area increases reaction kinetics and will therefore greatly increase the viability of the co-melting process.

Experiment

The furnace used is a resistance-heated furnace with molydisilicide heating elements and a digital controller. The heating chamber is an extruded alumina tube 90 millimeters (mm) in diameter and 1.25 meters in length. The vanadium samples were cut from 99.8 percent vanadium slugs with a diamond-edge blade. The samples were 5 mm in diameter, 2 mm in length, and weighed approximately 0.2 gram (g). The aluminum samples were cut with a carbide blade from 99.8 percent aluminum rod. The aluminum samples were 12 mm in diameter, 10 mm in length, and weighed approximately 2 g. The aluminum-to-vanadium contact surfaces were abraded with 800-grit carbide paper to remove burrs and to provide a relatively flat contact surface. Finally, the samples were cleaned with acetone and distilled water.

The samples were placed in an alumina crucible with the vanadium slug on top. Stainless steel foil was used inside the furnace tube hot zone to act as an oxygen-getter. Argon was introduced into the furnace before and during the heat cycle, and the samples were heated to 1300 degrees C for four hours. The temperature ramp rate was 10 degrees C per minute during heat up. Samples were furnace cooled following the four-hour exposure.

Results

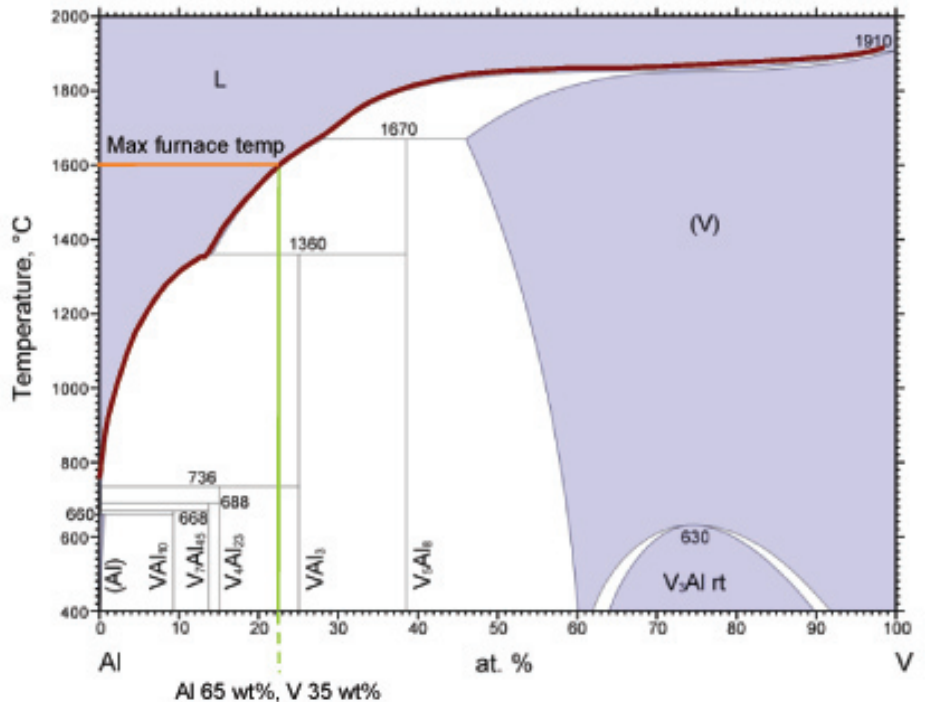
The vanadium and aluminum metal melted and formed an irregularly shaped ingot. The silver-colored ingot fused to the alumina crucible. Sectioning of the fused mass did not reveal any trace of the original vanadium sample. The interior of the product alloy was silver without any evidence of oxide inclusions or voids (in the macro scale), and there was no material segregation evident. The experiment showed that reaction melting is a viable way to melt the vanadium parts from ARIES pit disassemblies.

The phase diagram showed that up to 35 weight percent (wt%) vanadium could be melted in aluminum at the maximum operating temperature of 1600 degrees C. Increasing the vanadium concentration would increase the melting temperature beyond the furnace capabilities.

Lowering the temperature to 1400 degrees C would still allow us to melt up to 25 wt% vanadium. What this means is that, if some changes in concentration or temperature are necessitated by room or glovebox conditions, the vanadium part will still be sanitized. The experiment clearly showed that reaction melting is a viable way for ARIES to melt the vanadium parts. If needed, the same approach could also be used with other high-temperature metals.

The future

At this time, Los Alamos has no path forward for the disposal of plutonium-contaminated, classified, nonspecial nuclear material components. The current agreement between the Waste Isolation Pilot Plant (WIPP) and Los Alamos requires that the waste generated at PF-4 not contain any classified information when shipped to WIPP. In the past, the only solution has been to store the classified parts, but PF-4 space is in high demand and expensive and storage is not an option. Installing the sanitization furnace in PF-4 will give us the ability to dispose of these parts. This will not only benefit ARIES, but the entire Plutonium Facility as well, where a significant accumulation of parts has occurred over time.



Aluminum-vanadium phase diagram.
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CHARACTERIZATION OF PLUTONIUM OXIDE POWDERS

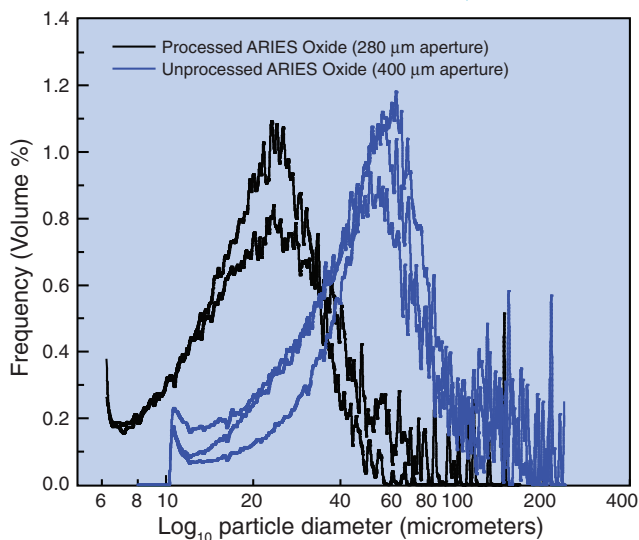
This article was contributed by David Wayne and Carl Martinez, Plutonium Manufacturing and Technology Division; Brian Bluhm, International and Applied Technology Division; David Gallimore and James Dyke, Chemistry Division; Donna Smith, Science Program Office; and Angelique Neuman, Materials Science and Technology Division.

In process chemistry, physical and chemical analyses are used to demonstrate that the final product meets the customer's specifications. This is an extremely important component of the ARIES program, which produces plutonium oxide (PuO_2) intended for use in mixed-oxide (MOX) nuclear fuel rods for commercial nuclear reactors. For example, the particle-size distribution of the plutonium oxide in a MOX fuel must be carefully controlled so that isolated, abnormally radioactive "hot spots" do not occur. Over time, such features may damage the fuel-rod cladding and cause less-efficient power production. Similarly, trace-element analyses are required because seemingly insignificant impurities of certain elements, typically in the parts-per-million (ppm) range, may adversely affect the long-term behavior of a MOX fuel.

In addition to their utility for quality control and quality assurance purposes, data obtained from the physical and chemical characterization of plutonium oxide powders also provide a wealth of information about the nature of the starting materials, the processes used to produce the plutonium oxide, and any subsequent processing that the oxide might have undergone. For example, ARIES plutonium oxide, which is produced by direct metal oxidation (DMO), has physical and chemical characteristics that are very different from those of MOX plutonium oxide, which is produced by a chemical precipitation process. These characteristics are of great interest to scientists working on other aspects of nuclear materials research, such as nuclear forensics and industrial hygiene.

Plutonium characterization is not a stand-alone module of the ARIES line, but is a process used as part of the packaging (ICAN) module. At TA-55 we perform several physical characterization analyses on plutonium oxide powders: particle size distribution (PSD), specific surface area (SSA), bulk/tap density, solid ("true") density, loss on ignition, and isotopic analysis by nondestructive assay (NDA) techniques. Samples are also sent to the Chemical and Metallurgical Research (CMR) Building for wet chemical plutonium assay, trace-element analysis by inductively coupled plasma mass spectrometry (ICP-MS) and inductively coupled plasma atomic emission spectroscopy (ICP-AES), and high-precision isotopic composition analysis by thermal ionization mass spectrometry (TIMS). Transuranic decay products such as americium and neptunium are also analyzed at the CMR, using gamma spectroscopy.

PSD measurements (ESZ technique) of ARIES oxide before (blue) and after (black) processing.



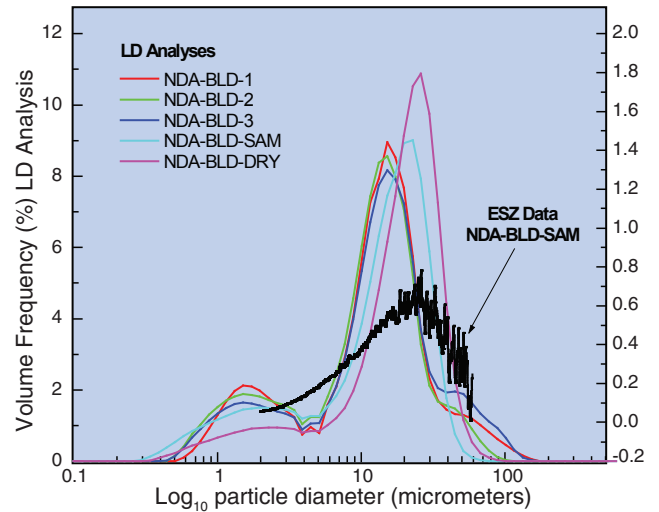
Particle size distribution

Particle size distribution data were obtained for several samples of ARIES plutonium oxide before and after the material underwent bulk-processing steps identical to those to be used at the Plutonium Disposition and Conversion Facility (PDCF). These steps included sieving to 180 micrometers (μm),

milling the oversize fraction, cross blending, and sampling by rotary riffing. The first batch of processed oxide was set aside for use in new Los Alamos NDA standards.

Complicating factors relevant to all PSD measurements are the high material density—11.5 grams per cubic centimeter (g/cm^3)—of plutonium oxide and the tendency for individual oxide particles to form loosely bound agglomerates that dis-aggregate continually during analysis. Thus, PSD results change over time and never reach a stable value. A reasonably accurate picture of the PSD in the dry powder can be obtained by quickly analyzing a preslurried sample, with minimal or no ultrasonic agitation.

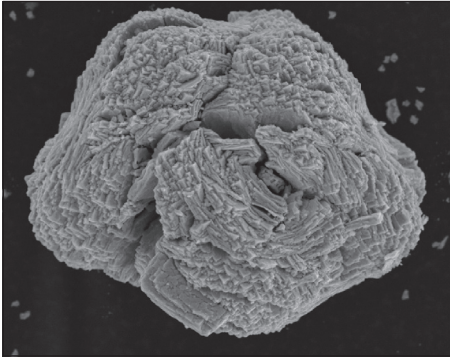
Typical results using the electronic sensing zone (ESZ) method—or Coulter Counter (*see sidebar*)—for ARIES oxides before and after processing show that mean particle size varies from 31–80 μm before processing and decrease to 21–27 μm after processing. Processing also decreases the modal and median particle sizes of the PSD, and decreases the diameter at d_{90} and d_{10} (*see sidebar*). Treatment in an ultrasonic bath de-agglomerated some of the larger particles and increased the populations of smaller particles. However, unprocessed ARIES oxide treated in an ultrasonic bath may still contain a significant number of particles with diameters greater than 200 μm , even after 10 minutes of intense agitation.



PSD measurements of processed ARIES oxide acquired, using both the LD method (various colors; see legend) and ESZ (black) techniques.

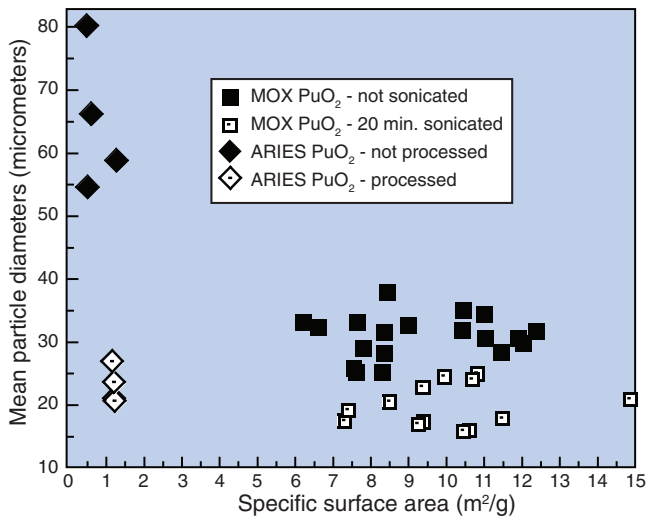
Sample	Ultrasonic Time (m)	Mean μm	Mode μm	d_{10} μm	d_{50} μm	d_{90} μm	% <200 μm
Before processing (Oxide from direct metal oxidation)							
ARIOXB02S3A	1.0	66.3	62.6	27.6	57.5	121	98.2
ARIOXB05S4A	1.0	54.7	54.0	21.8	49.0	90.5	99.4
ARIES020906	0	80.3	50.4	26.4	68.5	157	96.5
ARIES020906	10	73.0	33.0	39.8	68.8	105	98.9
ARIES1023	0	58.9	39.1	16.4	48.3	122	
ARIES1023	1.0	56.9	51.1	18.8	47.7	112	99.4
ARIES1023	5	51.9	34.4	18.5	41.7	105	
ARIES1023	10	44.7	29.2	18.2	37.6	80.5	
ARIES1023	10	31.4	19.6	10.8	27.2	57.8	
After processing							
NDA-BLD-SAM1	0	21.0	38.3	5.56	18.4	40.8	
NDA-BLD-SAM2	0	20.6	26.4	5.51	18.0	40.1	
NDA-BLD-SAM3	1	23.6	23.1	10.3	21.3	35.6	
NDA-BLD-SAM4	1	26.9	23.6	10.2	22.7	47.5	

Particle-size data obtained using the laser diffraction (LD) technique (*see sidebar*) includes particle populations down to 0.2 μm in diameter, a range that cannot be analyzed using the ESZ method. The LD data compare well to the ESZ data. All samples analyzed using the LD technique contain significant particle populations smaller than the 1- μm limit attainable using the smallest ESZ aperture. The “dry” LD data refer to a sample slurried and analyzed immediately



SEM photomicrograph of a typical polycrystalline plutonium oxide particle taken from a MOX sample. Note the complex, highly convoluted surface.

Specific surface area and mean particle diameters of ARIES (diamonds) and MOX (squares) plutonium oxide (PuO₂) powders.



after addition to the reservoir, with no ultrasonic agitation. The remaining LD samples were ultrasonically agitated for 30 to 60 seconds before data were collected. The ESZ sample shown by the black line in the figure on the previous page was not ultrasonically agitated, and the resulting data correspond most closely to the unsonicated dry run from the laser diffraction analyzer.

Sample	Mean μm	Mode μm	d_{10} μm	d_{50} μm	d_{90} μm	% <0.5 μm	% <1.0 μm	% <100 μm	% <200 μm
NDA-BLD-SP1	15.94	14.24	1.62	12.61	30.67	0	3.096	99.5	100
NDA-BLD-SP2	13.56	14.13	1.45	11.64	26.04	0.134	5.114	99.998	100
NDA-BLD-SP3	17.41	14.20	1.61	13.01	38.95	0.128	4.488	99.5	100
NDA-BLD-SAM	14.56	21.14	1.50	14.17	27.57	1.212	5.972	100	100
NDA-BLD-'DRY'	19.31	24.28	2.98	19.08	33.48	0.445	2.95	100	100

Specific surface area

The DOE has not set forth a specification for the specific surface area of ARIES plutonium oxide. Even so, plutonium oxide specific surface area data are of great interest to the nuclear materials community. Plutonium oxide particles readily adsorb water vapor from the surrounding atmosphere, and the amount they can adsorb is dependent on the oxide's specific surface area: the greater the surface area, the greater the amount of adsorbed water. It is important for scientists to be able to anticipate the moisture content of plutonium oxide powders because, in the presence of plutonium, water can likely be transformed into highly reactive and potentially explosive hydrogen (H₂) gas due to a radiation-caused process known as radiolysis.

Using the dynamic Brunauer–Emmett–Teller (BET) technique (*see sidebar*), we quantified the SSA of approximately 15 ARIES plutonium oxide samples.

The SSA of unprocessed ARIES plutonium oxide varies from approximately 0.3 square meter per gram (m²/g) to approximately 1.7 m²/g, while SSA measurements from the processed oxide indicate that the material is much more uniform, with an average SSA of 1.21 ± 0.02 m²/g.

Both the mean particle size and specific surface area of processed and unprocessed ARIES plutonium oxide is distinct from those of MOX plutonium oxide. A picture of a typical MOX oxide particle taken using a scanning electron microscope shows the complex, porous surface of the polycrystalline MOX plutonium oxide aggregate, which explains its significantly greater SSA relative to the ARIES plutonium oxide. Taken together, physical characterization data can help nuclear forensics experts trace the ultimate source of an unknown plutonium oxide and provide information on the nuclear processing capabilities in foreign countries.

Trace elements

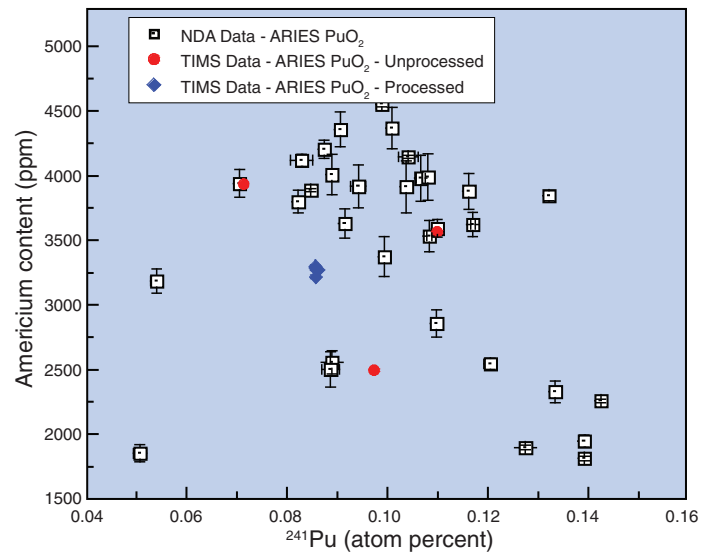
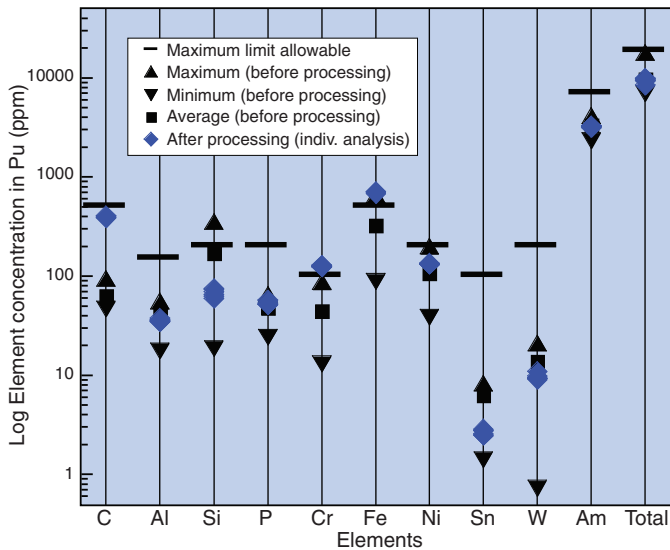
Trace elements are impurity elements with an average concentration less than 100 ppm. Most impurity elements in ARIES plutonium oxide are present in quantities less than 100 ppm, and well below the maximum ARIES limits agreed to by DOE. Americium and gallium are by far the most abundant impurities in ARIES plutonium oxide. Gallium is an important alloying element added to the plutonium metal. Actinide and transuranium elements, such as americium, uranium, and neptunium, are expected to be present in concentrations far greater than 100 ppm because they are byproducts of the radioactive decay of the various plutonium isotopes. Other impurity elements that are often present in amounts exceeding 100 ppm, such as carbon, iron, and silicon, may occur in such quantities because they are ubiquitous in the glovebox environment needed for the safe handling of plutonium.

An area of particular concern for the ARIES program is the potential for adding extra impurities to the plutonium oxide during processing. As the oxide is sieved, milled, blended, and sampled, it comes into contact with a wide range of materials, each a potential source of contaminants. Another area of concern is our ability to create a chemically and physically homogenous blend from chemically different sublots of material. We analyzed ARIES plutonium oxide before and after processing to investigate the possible addition of impurities to the finished material and to see if ARIES processing was capable of producing a homogenous lot.

Analytical results for select trace impurities before and after ARIES processing show that the material was rendered chemically homogenous with regard to trace and minor impurities. For example, before processing, the variation in americium concentrations over six samples exceeded 20% relative standard

Below left: Selected impurities in ARIES oxides before (black symbols) and after ARIES processing (blue symbols). Maximum element contents specified by DOE for ARIES oxides are also shown (horizontal bars).

Below right: Plot of americium-241 content vs. plutonium-241 for ARIES oxides analyzed either by NDA (hollow squares) or by TIMS (blue and red filled symbols).



deviation (RSD). After processing, the between-sample variation in americium was less than 2% RSD. Similarly, the average measured total impurity content in the PuO_2 changed by approximately 0.7% after processing, while the between-sample variation for total impurities decreased by almost an order of magnitude (from 34.2% to 5.6%). Individual elements that were in excess of the specifications (for example, silicon) for certain samples were averaged out by the intensive sieving, milling, and blending regimen.

After processing, the material showed significant net increases in mean concentrations of carbon, chromium, and iron, along with a slight gain in mean nickel concentration. Because the material was milled in a stainless steel rod-milling jar with tungsten carbide rollers, it is quite possible that the material acquired iron, chromium, carbon, and nickel via contamination from the walls of the steel milling vessel. Interestingly, an increase in tungsten concentration did not occur in the processed material, indicating that little, if any, material was lost from the rollers.

Plutonium isotopic composition data from PuO_2 sampled before and after ARIES processing also indicates that the material was effectively homogenized. The extremely high precision and accuracy of isotopic data acquired using TIMS is illustrated in the figures shown below, where the error bars on the individual analyses are smaller than the symbols used to denote the data points.

Conclusions

Besides providing key quality data for the ARIES program, physical and chemical analyses of nuclear materials also provide a wealth of information that scientists can use to reveal the age, origin, and processing history of nuclear materials. Many of these tests are relatively simple and can be conducted rapidly, even in a glovebox environment.

We also devised a powder-processing regimen for ARIES oxides and tested it on ARIES oxide intended for disposition in four new Los Alamos NDA standards. Physical and chemical characterization data were collected both before and after processing. These data indicate that the ARIES processing scheme produced a continuum of plutonium oxide with near-uniform chemical and physical properties.

Distinct trace element and isotopic signatures in the starting PuO_2 lots were homogenized; particle size, specific surface area, and other physical characterization measurements were uniform and well within ARIES specifications. Rod milling in a steel milling jar may have increased the concentrations of several trace impurity elements, such as iron, chromium, and carbon. However, none of the postprocessing samples had total impurity concentrations in excess of the ARIES specification. The amount of impurities introduced during processing may be decreased by reducing milling time, reducing the amount of material that is milled, or both. A ceramic (either Al_2O_3 or ZrO_2) milling jar and media may also be used instead of steel.

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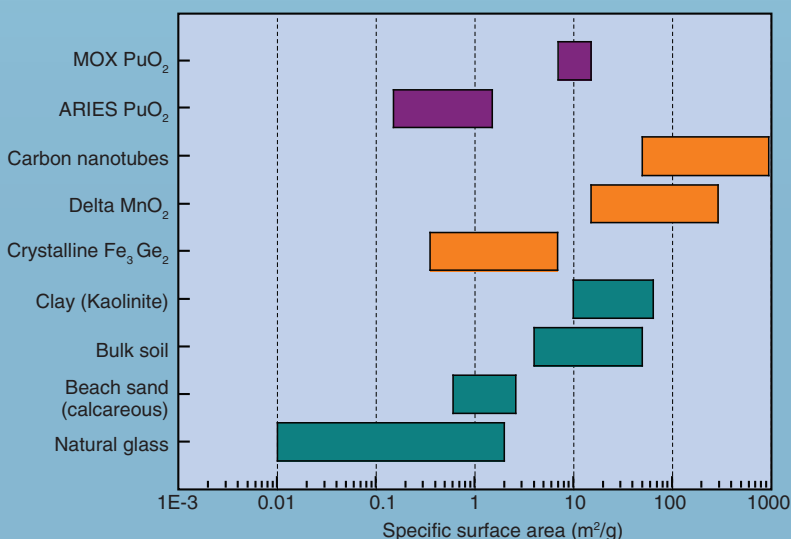
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Specific surface area measurement

The evaluation of the specific surface area (SSA) of a powder is based on the Brunauer–Emmett–Teller (BET) model for inert gas physisorption onto a solid surface. BET is a well-characterized analytical method that has been used since the late 1930s. It assumes that the surface is completely covered with a monolayer of gas molecules before a second layer of gas molecules is sorbed.

SSA measurement of a bulk particulate involves gently heating (outgassing) the sample to approximately 200 degrees Celsius (C) to rid the surfaces of contaminants. The sample is then immersed a stream of a high-purity molecular nitrogen (N₂) and helium (He) gas mixture and cooled to the boiling point of liquid N₂, which is 77.4 kelvin (K). At liquid N₂ temperature, a monolayer of N₂ from the gas stream is sorbed onto the sample surface. Upon warming, the sorbed N₂ is released and detected. The integrated N₂ is detected, and its peak height is proportional to the surface area of the sample. We use the multipoint method of surface-area determination, in which the analysis is repeated at different N₂/He ratios.

The accompanying figure shows the variation of SSA measurements for a variety of natural and man-made materials. SSA varies as a function of particle size, surface roughness, and the degree of interconnected porosity. At the low end of the SSA spectrum are nonporous, crystalline materials with relatively smooth surfaces, such as beach sand and natural glass particulates. Materials with high SSA are porous and have highly complex and irregular surface topologies. Naturally occurring materials with high SSA include clays and biogenic iron and manganese oxides. Man-made compounds such as carbon nanotubes and aerogels may have extraordinarily high SSAs, in excess of 1000 square meters per gram (m²/g).



Specific surface area data for various natural and man-made materials.

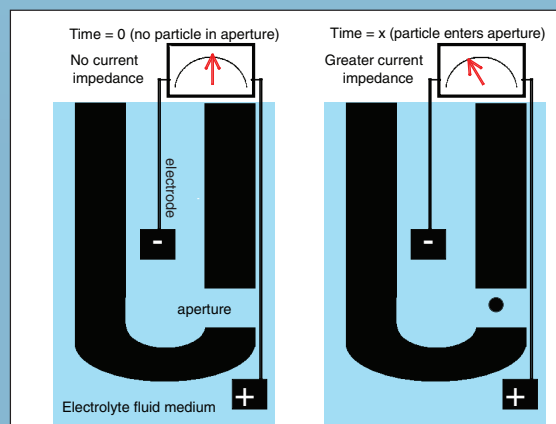
Particle size distribution analysis

The particle size distribution (PSD) of a granular aggregate is a quantitative measure of the number of particles present, sorted according to their diameter. For actinide materials, particle size data are used to determine the respirable fraction of fine bulk powders, to characterize the properties of product materials from various TA-55 process lines, and to optimize processing parameters for quality-control purposes.

In its simplest form, PSD analysis assumes that all particles are spherical in size. In real life, this is often not the case. As a result, the data from a PSD measurement can vary significantly based on the type of measuring technique used. Knowledge of PSD is extremely important for many industrial processing applications where the flowability, chemical reactivity, mechanical strength, and thermal properties of a powdered or granular aggregate must be tightly controlled.

The archetype for PSD measurement is sieve analysis, in which the powder is passed through a stack of sieves, each with a different mesh size. Each sieve will “stop” the particles that are too large to pass through. At the end of the analysis, the content on each sieve is weighed to yield the proportion of material in each size bin—for example, “percent of sample between 180 micrometers (μm) and 200 μm .” PSD data are expressed in two ways: as a binned analysis, in which the amount in each size range category or bin is listed in order; or in cumulative form, in which the total of all sizes passed through (under-sized) or retained by (over-sized) a certain bin size is given for the entire range of bin sizes. Although sieving is still used to determine the PSD of granular materials, an array of modern-day techniques provides narrower bins over far greater size ranges.

Currently, we use two methods to determine particle-size distribution in bulk plutonium oxide powders: the electronic sensing zone (ESZ) technique, also known as the Coulter Principle, and laser diffraction (LD). Both techniques require a small amount (approximately 100 milligrams) of sample to be suspended in a flowing electrolyte solution. In the ESZ method, the suspension is pumped through a tiny cylindrical aperture. An electrical current flows across electrodes located on either side of the aperture. As each particle passes through the aperture, it displaces a corresponding volume of conducting liquid, momentarily increasing the current impedance. This change in impedance produces a proportional current flow that is fed into an amplifier, which converts the current fluctuation into a measurable voltage pulse. The Coulter Principle states that amplitude of this pulse is directly proportional to the volume of the particle that produced it. The particle diameter is then calculated from the Coulter volume.



Schematic representation of the Coulter Principle.

For LD analysis, the suspension is pumped through a transparent flow cell. Light emitted sequentially from two different sources—a 632.8-nanometer (nm) helium-neon (He-Ne) laser, and a 405 nm monochrome blue tungsten lamp—passes through the fluid-filled flow cell where it interacts with the suspended particles. Dispersed and scattered laser light enters a condenser lens and is focused on the 75-element forward ring silicon-diode array detector. Scattered light from the tungsten lamp is sensed by a detector array, which converts the light intensities into electrical signals that are used to calculate the particle size distribution of the suspended solids.

The resulting data are processed and statistically analyzed. The most meaningful parameters are the mean, mode, median, d_{10} , and d_{90} particle sizes. The mean is the average particle size in the entire population. The mode is the particle size that occurs most often. If two values occur with the same greatest frequency, each is considered a mode, and the PSD is said to be bimodal. The median particle size is the diameter of a particle that has an equal number of larger particles and an equal number of smaller particles. The d_{10} and d_{90} particle sizes correspond to particle sizes in which 10% and 90% of the particles are smaller than itself. All of these statistical measures are used to describe the general trends in PSD within a granular or powdered sample.

Chemical and isotopic analyses

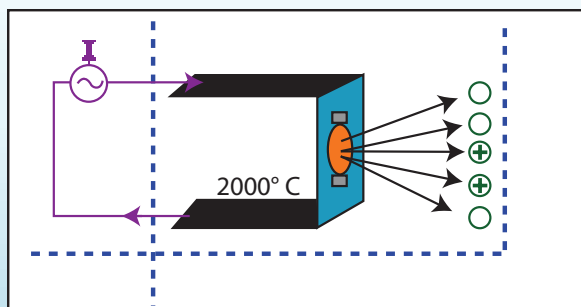


A working ICP-MS ion source.

Plutonium oxide powder samples are sent over to the Chemistry and Metallurgy Research (CMR) Building for dissolution and chemical analysis. Light elements (up to the first-row transition elements on the Periodic Table) are determined by using inductively coupled plasma atomic emission spectroscopy (ICP-AES), while heavier elements are quantified by using inductively coupled plasma mass spectrometry (ICP-MS). Isotope ratios for plutonium and uranium are measured by using thermal ionization mass spectrometry (TIMS).

Transuranium elements such as americium and neptunium are measured by using gamma counting techniques. Wet chemistry techniques are used to quantify carbon, nitrogen, sulfate, and halogen contents in plutonium oxides.

Isotopes are atoms of the same element that have different nuclear weights due to differences in the number of neutrons in their nuclei. The most well-established technique used to measure quantities of isotopes in an element (also known as the “isotopic composition”) is mass spectrometry. For mass spectrometric analyses, samples are dissolved in strong acids and then purified using ion-exchange resins and ultrahigh-purity acids to eliminate



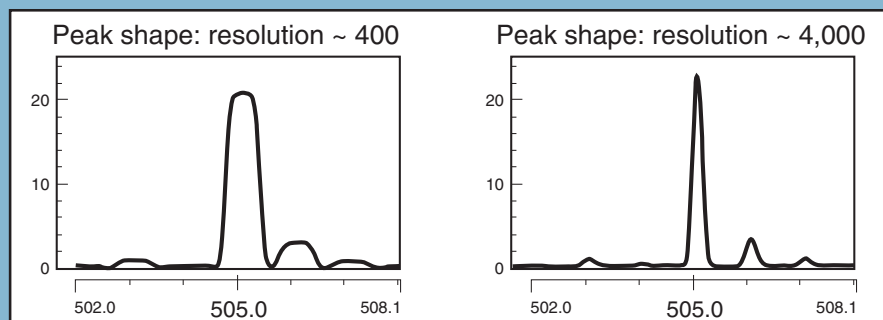
Schematic representation of a TIMS ion source.

all elements except for the element to be analyzed. The purified sample is dried to a tiny droplet and loaded onto the metal filament of a thermal ionization mass spectrometer. Under high vacuum in the TIMS sample chamber, the filament is heated until the sample vaporizes and then ionizes. The ions are accelerated and subjected to a curved magnetic field, which sorts them according to their mass-to-charge ratio. The ion signals are then detected, amplified, and quantified.

By contrast, samples for trace element analysis by ICP-MS are dissolved, diluted, and sampled directly into the ICP ion source. Unlike TIMS, which requires a very high vacuum, the ICP ion source operates at near-atmospheric pressure. The sample is simply introduced as an aerosol that is sprayed into a specially designed plasma torch. The ICP uses a very powerful radiofrequency field to ignite the aerosol and form plasma that reaches temperatures in excess of 7000 kelvin (K). The advantages of the ICP source are twofold: most elements are completely ionized in the ICP plasma, which makes mass spectrometric analysis very convenient, and the light emitted by the plasma can also be analyzed spectroscopically for characteristic wavelengths emitted by the elements in the sample. Both mass spectrometry and light spectroscopy from ICP sources are used to analyze trace elements at the CMR Building.

The different types of data that can be acquired using TIMS and ICP-MS have a lot to do with how the ions are produced and detected. Thermal techniques produce a stable, low-energy ion beam that also has a relatively narrow energy distribution that is ideal for the extremely precise and accurate analyses required for isotopic analyses. The ICP source produces copious amounts of high-energy ions with a much greater energy distribution.

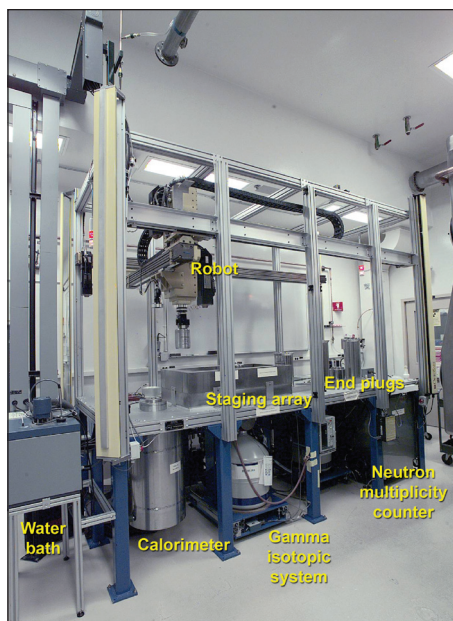
Consequently, ICP sources are more difficult to use in isotopic analysis, although recent advances in ICP-MS technology have made this possible. The main advantages of the ICP source are its incredible sensitivity and its ability to uniformly ionize all of the elements in a sample. Both of these properties make it well suited for trace element analyses.



Examples of isotopic data collected at low and high resolutions.

ARIES NONDESTRUCTIVE ASSAY INSTRUMENTATION

This article was contributed by Tracy Wenz, Nuclear Nonproliferation Division; and Gary Herrera and Becky Guillen, Plutonium Manufacturing and Technology Division.



The ARIES nondestructive assay system at TA-55.

The nondestructive assay (NDA) module is the final step in the ARIES process. This module assays the packaged plutonium oxide in a calorimeter and a gamma-ray isotopic system for Materials Control and Accountability (MC&A) before it is sent to storage in the TA-55 vault. The module also contains a passive/active neutron multiplicity counter (NMC) that can assay both plutonium and uranium oxides. The NMC will be the primary MC&A analysis method for uranium oxide at the Pit Disassembly and Conversion Facility (PDCF). The module demonstrates the measurement capabilities of the NDA instruments that will be used at the PDCF on identical materials.

The ARIES NDA instruments are located inside a metal frame that supports a robot used to handle the welded steel DOE 3013 containers and configure the instruments for measurements. A dedicated table top provides a mounting surface for all three instruments as well as temporary storage for the calorimeter baffle and the NMC end plugs while they are not in use. A shielded staging array is also located here to safely store cans awaiting measurement after they are introduced into the NDA system.

Items are manually brought to the input station on the NDA table where the robot moves them into the shielded staging array in preparation for measurement. Once all the items are introduced into the NDA system, the robot picks up one from the staging array and loads it into an instrument. Items are assayed in the different instruments and then returned to the sample staging array when the measurement is complete. Once all the assays are completed, the robot moves the item to the output station, and the 3013 can is manually removed from the system.

The assay of the cans in the NDA system can be performed manually from the robot and NDA computer consoles, or in a semi-automated fashion using the NDA host software, which has a scheduling capability that allows the operator to queue cans for measurement in the desired instruments. The host software also directs robot operations, including bar-code scanning and calorimeter baffle and end-plug movement, and monitors the work area through an array of position sensors. The host software stores the completed measurement results for each can and is able to combine calorimetry and isotopic results to generate summary reports with the elemental plutonium mass for each can.

Calorimetry

The heat-flow calorimeter has a gradient bridge design, which is the most compact of the available designs and is relatively inexpensive to fabricate. The calorimeter is designed to measure containers up to 13.02 centimeters (cm), or 5.125 inches, in diameter by 25.4 cm (10 inches) tall. The inner diameter of the calorimeter is fitted with a sleeve to precisely accommodate the nominal 126-millimeter (mm) diameter of the 3013 container as specified by the DOE. The precision sleeve allows us to perform NDA measurements without using a

“cal can” adapter (an over-pack container in which the nuclear material is placed to facilitate insertion and removal of the material from the calorimeter), and it can be modified to meet future changes to the 3013 Standard. Internal heater windings have been incorporated into the calorimeter for operation in servo-controlled mode, although it is normally operated in the passive mode without heaters.

The voltage measured across the Wheatstone bridge—an electrical circuit used to measure the change in resistance between the reference and sample chambers in the calorimeter—is converted to watts by using a calibration curve fitted to heat-flow data from plutonium-238 heat sources or plutonium samples with known mass and isotopic composition. The plutonium-238 heat sources have been calibrated against electrical standards that are traceable to the national measurement system.

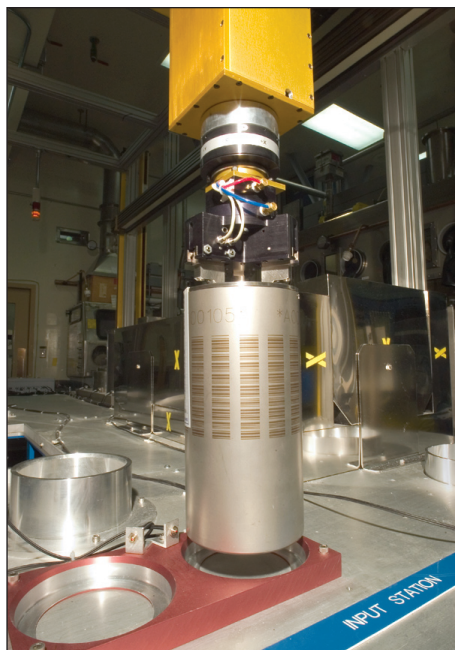
The plutonium samples are also traceable through destructive chemical and isotopic analysis results. The measured wattage of the unknown sample is converted to grams of plutonium using the parameter, effective specific power, P_{eff} (watts per gram of plutonium, or W/g Pu), from the gamma-ray isotopic analysis measurement.

For a set of plutonium oxide calibration standards ranging from 1.8 to 11.4 watts, the measurement bias (measured/true) = 1.0005 and the sample standard deviation (1σ) = 0.0026 for a set of 39 measurements.

Isotopic analysis system

The isotopic analysis system uses a high-purity germanium (HP Ge) detector mounted in a streamlined horizontal cryostat in a 30-liter liquid nitrogen Dewar. The detector is placed inside a 3.175 cm-thick (1.25 inches) tungsten shield that is mounted on a moveable detector platform. The detector also has a 1.27 cm (0.5 inch)-thick tungsten back shield. The detector platform is mounted on rails so that the detector-to-sample distance can be adjusted to optimize the count rate for the assay. The item container is placed on an elevator in front of the detector where it is rotated and vertically scanned during the measurement. The scanning is performed to average over any isotopic inhomogeneities that may be present in the sample.

In the past year, the original planar detector has been replaced with a 32% coaxial detector. The coaxial detector provides smaller uncertainties on the individual isotopic values and on specific power in the high-energy analysis region—200–1001 kiloelectron volts (keV)—for analyses of full 3013 cans. Minimizing the individual isotopic uncertainties is becoming more important as the role of the isotopic system expands to include characterization of the plutonium oxide for the fuel fabrication facilities. The expanded role of NDA will reduce destructive analysis costs. Also, going to a coaxial detector enables the system to collect prompt gamma spectra (800–5000 keV) used to quantify certain impurities in the 3013 oxides.



A robot picks up a 3013 can at the input station in preparation for measurement in the nondestructive assay system.

The isotopic analysis does not require standards for calibration, but it does use standards to verify measurement performance. The following table shows measurement uncertainties for a full 3013 can standard measured for two hours, along with the measurement requirement for the fuel processing facility.

ARIES Coaxial Isotopic Measurement Results								
	# of runs	Relative uncertainty at 2 standard deviations (%)						
		Pu238	Pu239	Pu240	Pu241	Am241	Pe _{eff}	Pu240 _e
Measurement	20	9.4%	0.14%	2.25%	9.84%	0.42%	0.26%	2.22%
MOX FFF Requirement	—	20%	0.2%	3.5%	13.5%	4.0%	—	—

Neutron multiplicity counter

The neutron multiplicity counter (NMC) is a cadmium-lined well counter containing eighty helium (³He)-filled tubes (at 4-atmosphere fill pressure) arranged in four rings embedded in polyethylene. The detector has a neutron counting efficiency of approximately 52%. The tubes are connected to twenty preamplifiers that feed to a derandomizer circuit for dead-time reduction before the signal pulses are analyzed by the multiplicity shift register from which the singles, doubles, and triples count rates are calculated.

The NMC has three sets of end plugs. One set is used for passive measurements of plutonium. A second set contains americium/lithium (AmLi) sources for active uranium assay. The third set is identical to the active set though it does not have the AmLi sources. The latter two end plug sets are used to determine the amount of uranium in uranium/plutonium mixtures, where the dummy active end plugs—those without the AmLi sources—are used to measure the background plutonium count rates in the sample.

Multiplicity analysis is the primary method used on ARIES plutonium. Multiplicity analysis results are independent of the chemistry of the sample, as plutonium oxides may contain low-Z (low atomic mass) impurities that affect its (α,n) neutron rate. For neutron assay of plutonium, the mass, multiplication, and (α,n) neutron generation rate are unknown, but with three measured quantities—the singles, doubles, and triples count rates—the mass of the sample can be calculated directly without making any assumptions about the sample.

Two other analysis methods are also used: known-alpha and passive calibration curve. These two methods require traceable calibration standards. Because these two methods rely on calibration curves, the results can be subject to bias due to changes in sample multiplication or composition; however, they are useful for identifying these changes in the material.

The plutonium-240-effective mass, a weighting of the even isotopes for plutonium, is the quantity calculated from all three neutron analysis methods. The plutonium-240-effective fraction is obtained from the measured isotopic fractions and used to convert the plutonium-240-effective mass to elemental

plutonium mass. The following table shows assay results from the previous ARIES demonstration for the three neutron analysis methods. Multiplicity counting results have the lowest uncertainty because they are least affected by impurity concentrations and density variations.

ARIES NMC Measurement Results		
	Bias (Measured/True)	Sample Standard Deviation (1σ)
Passive Calibration Curve	0.981	0.060
Known Alpha	1.005	0.089
Multiplicity Counting	1.000	0.025

Further reading

Application Guide to Gamma-Ray Isotopic Analysis Using the FRAM Software, T. Sampson, T. Kelley, and D. Vo, Los Alamos National Laboratory report, LA-14018, September 2003.

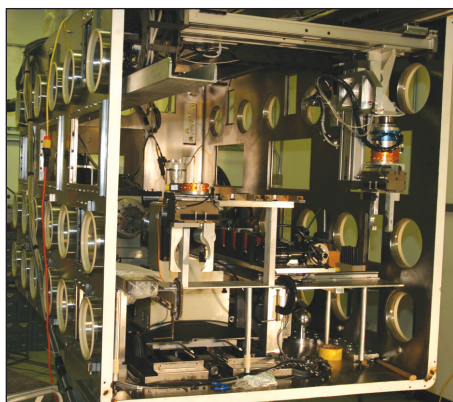
Application Guide to Neutron Multiplicity Counting, N. Enssline, W. Harker, M. Krick, D. Langner, M. Pickrell, and J. Stewart, Los Alamos National Laboratory report, LA-13422-M, November 1998.

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The ARIES Nondestructive Assay System: Description and Process Demonstration Results, T. Sampson, T. Cremers, T. Wenz, W. Hansen, N. Scheer, T. Kelley, W. Harker, D. Mayo, D. Bracken, G. Herrera, and B. Guillen, Los Alamos National Laboratory report, LA-14143, May 2004.

AUTOMATING ARIES

This article was contributed by Cameron Turner and Jane Lloyd, Plutonium Manufacturing and Technology Division.



Simulation models of the pit disassembly (PDIS) module were used to study the design, maintenance needs, and workspace of the PDIS automation systems.

ARIES has been a significant reason for the development of glovebox automation technologies. Only two of the original ARIES modules were initially automated. However, from the beginning, the ARIES project planned to incorporate additional automation by replacing the modular subsystems with more advanced designs that often included automation. The nondestructive assay (NDA) module was one of the first ARIES modules to be automated. Automation systems are being added to the pit disassembly (PDIS) and the robotic packaging (ICAN) modules. ICAN, which is also known as the robotic integrated packaging system (RIPS), is the most complex automation module yet deployed within ARIES.

Nondestructive assay module

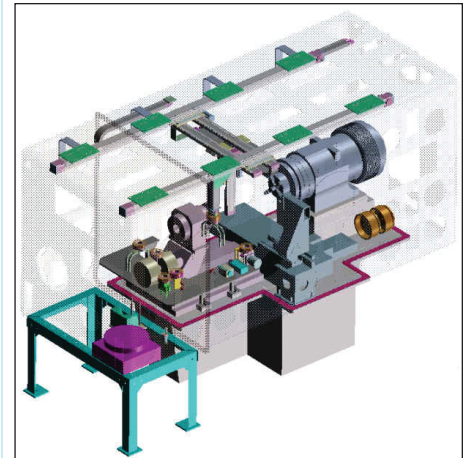
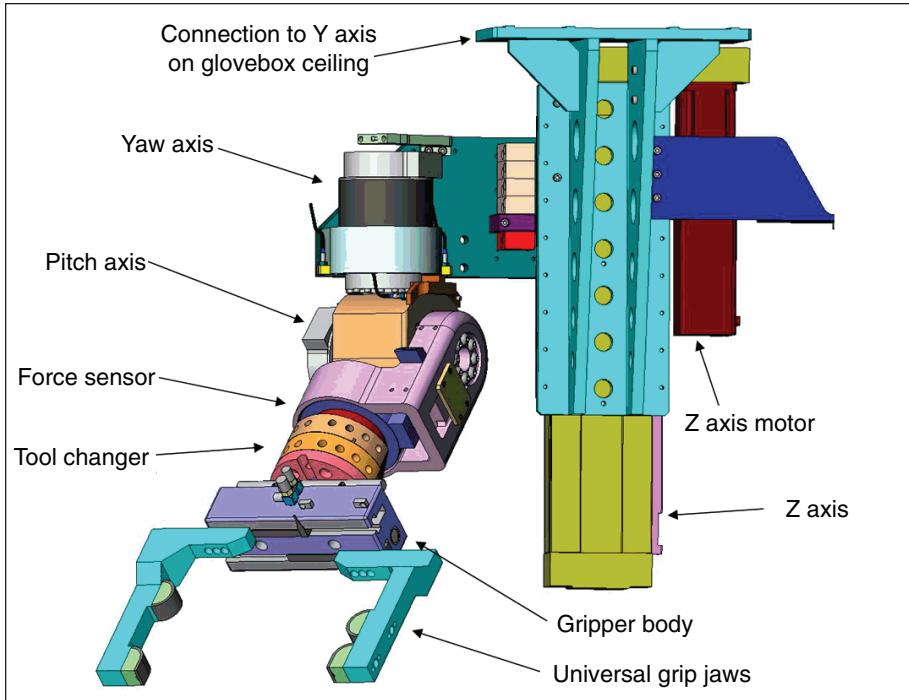
The NDA module, which is discussed in the article beginning on Page 28, is built around a three-degree-of-freedom (DOF) gantry robot. (A gantry robot is a stationary robot comprised of two or three orthogonal linear axes of motion.) The NDA robot uses commercial x and y axes, and a custom Los Alamos-designed telescoping z axis. In robotics parlance, an axis describes not only an angle of movement, but also the mechanical components that comprise the joint and drive or the robot. The robot allows fully autonomous 24-hour operation of the NDA system and uses four instruments to measure the fissile content of material containers without the need for operator oversight.

NDA was selected as the first module to be automated for several reasons. First, because it operates outside the glovebox environment, the robotic technology could be evaluated in a readily accessible and maintainable environment with minimal programmatic risk. Second, the tasks performed by the NDA module are highly repetitive and primarily involve the movement of well-defined objects between well-defined locations. Tasks of this type generally are easy to automate and NDA's success led to additional automation.

Pit disassembly module

The gantry robot used in the PDIS module is of similar design to that used in the NDA module. The PDIS robot consists of three commercial axes and a custom Los Alamos-designed two-DOF wrist. The PDIS system operates within a glovebox, and consequently additional consideration was given to maintenance needs and potential failure modes of the robot. Many of these potential events were explored through computer simulations before the design was completed.

These simulations also explored the workspace properties of the robot, which is of particular importance in this case because the PDIS robot shares its workspace with an automated Moore lathe and customized tool changer. The lathe, tool changer, and tooling add an additional five degrees of freedom to the system. All three automation systems share a common control system enabling coordinated motions involving several systems.



Los Alamos custom designed a two-degree-of-freedom wrist for the pit disassembly module robot, which enables the system to reorient items with its gripper during operations (left).

The pit disassembly module during system integration, showing the robot and grippers designed for the system (above).

Unlike the NDA robot, the PDIS robot is designed to function as a semi-autonomous system. It is intended to operate in collaboration with, and under the direction of, a human operator. While several common actions will be preprogrammed, many robot activities will be directed by the operator through manual controls. Currently, the PDIS module is being acceptance tested.

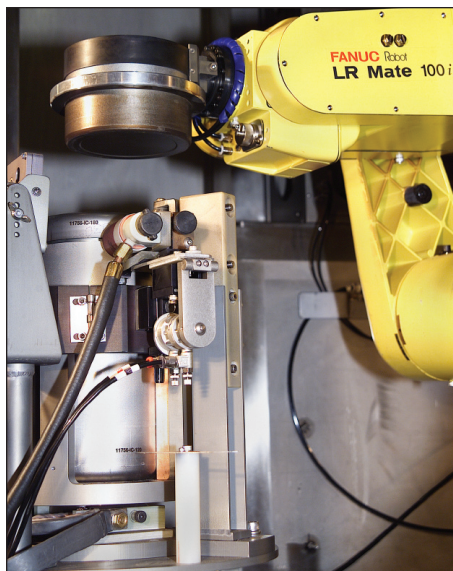
Robotic integrated packaging system module

The ICAN module is more complex than either the PDIS or NDA modules. Like PDIS, ICAN is contained within a glovebox, which is divided into three separate compartments: the hot side, the cold side, and the fluid-processing side. Both the hot and cold sides of ICAN have their own five-DOF Fanuc LR Mate 100i robots. These robots were the first industrial robots used to automate an ARIES glovebox. These two robots are integrated with an automatic welding system, an electrolytic decontamination system, two separate leak-testing systems, and a radiation survey system. These additional systems add another 10 degrees of freedom to the system, bringing ICAN's total to 20 degrees of freedom of automation. While ICAN has twice the automation complexity as PDIS and nearly seven times that of NDA, it has the smallest footprint (40 square feet) of the automated ARIES modules. In fact, ICAN fits inside the NDA or PDIS systems.

ICAN is designed for fully automated (but attended) operation. The system receives a crimped convenience can of fissile material in the hot-side chamber of the glovebox. The robot then places the convenience can in a stainless steel DOE



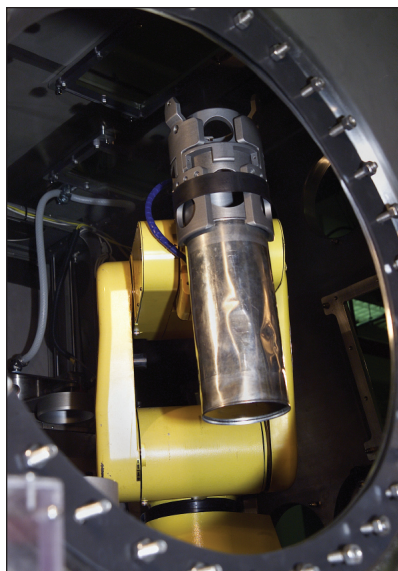
The hot-side chamber of the robotic integrated packaging system (RIPS) glovebox. RIPS is also known as the robotic packaging (ICAN) module.



The hot-side FANUC LR Mate 100i robot moves a material container during acceptance testing (above left).

The hot-side robot prepares an inner can for welding (above right).

While suspended from the glovebox ceiling, the cold-side robot works in cramped quarters to survey a freshly decontaminated inner can (above).



3013 inner can. The inner can assembly is placed in the automatic welding system, which welds the can under an inert helium atmosphere. The welded inner can is transferred to the hot leak-check system, which verifies that the weld hermetically seals the can. Then the inner can is transferred to the electrodecon chamber where the can is decontaminated electrolytically. The fluid-processing chamber of the glovebox contains the necessary equipment to support the decontamination operations.

The electrodecon chamber also serves as an airlock between the helium atmosphere of the hot-side chamber and the air atmosphere of the cold-side chamber of

the ICAN glovebox. Following decontamination, the cold-side robot processes the welded inner can through a series of radiation surveys to determine the presence, amount, and location of any removable or fixed contamination on the can's exterior.

If the can passes the radiation survey, the system performs a second helium leak check to verify that the weld is intact. Once this cold leak check is complete, the can is released from ICAN.

The radiation survey is independently verified by a second survey performed by a radiological control technician (RCT), but the automated survey process may eventually eliminate the need for an independent RCT survey. If a process failure occurs, ICAN is capable of autonomous, semi-autonomous, and manual fault recovery in collaboration with an operator.

Simulation was extensively used to design ICAN. Simulations studied the workspaces of the robots, the maintenance needs of the system, and alternative design configurations. The robots used in ICAN are commercial grade, unlike the customized systems used elsewhere in ARIES.

The commercial robots are designed with expected lifetimes in excess of 20,000 hours. If replacement is necessary, the ICAN glovebox is designed to allow for either robot to be removed and replaced. These activities have been demonstrated both in simulation and during acceptance testing. It is even conceivable that these robots could be reused in other glovebox applications. Because of the customized nature of their designs, it is unlikely that either the NDA or PDIS systems will be reused. The ICAN system has completed acceptance testing and is awaiting approval for operation.

Future automation systems

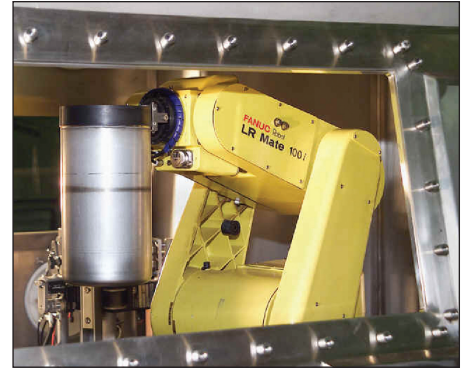
Future automation systems will surpass the capabilities of NDA, PDIS, and ICAN. Through the DOE University Research Program in Robotics, The University of Texas at Austin in conjunction with Los Alamos has demonstrated several new automation capabilities. One promising avenue of research is modular robotic systems whose kinematics can be reconfigured for different tasks within gloveboxes and could be maintained with far greater ease than current systems. Other promising research includes compliant manipulators that can safely share workspaces with human operators and advanced simulation and programming capabilities that will speed the integration of future automation systems.

Technologies are also emerging that will increase the sensor density of automation systems, thereby enabling increased system intelligence and supporting condition-based maintenance. Condition-based maintenance would significantly improve confidence in the ability of automation systems to operate reliably in hazardous environments by supporting a maintenance-on-demand paradigm. In addition, the fundamental design of robotic systems is becoming increasingly influenced by biological systems (*see sidebar*).

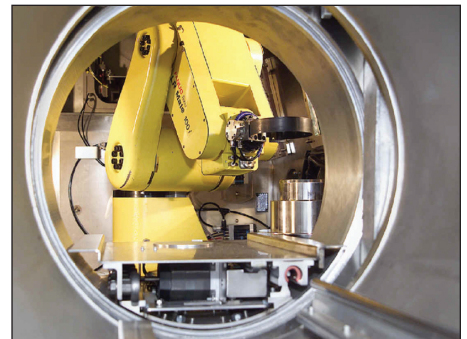
New developments in robotics, such as the Stanford Stickybot, may provide a platform reduce or eliminate certain glovebox tasks, and future manipulators may be designed to mimic human arms and installed in unused gloveports where they can function just like a human operator, only without the limitations of gloves. Such systems will be able to safely share workspaces and work independently or in direct collaboration with humans. These technologies almost certainly will be implemented in the next generation of glovebox automation projects.

In the next couple of decades, automation systems developed, based on the experience gained in automating the ARIES pilot line, will be installed in many applications throughout the DOE nuclear complex. The Pit Disassembly and Conversion Facility (PDCF), for which ARIES is the prototype, will incorporate extensive automation systems throughout the facility.

Future systems will embody even greater complexity than those currently deployed, will be more intelligent, will be able to monitor their own maintenance needs, and will increasingly resemble biological systems. Modularity will become commonplace, easing maintenance tasks and allowing systems to be configured, deployed, and programmed for different tasks with ease. Sensor, electronics, and processing power have finally allowed engineers to successfully design and deploy robots as critical components in nuclear material handling operations. Robots will soon be appearing in a glovebox near you.



The hot-side robot moves a welded inner container during acceptance testing.



The hot-side robot poses for a photo through the glovebox airlock during testing.

BIO-INSPIRED ROBOTICS FOR NUCLEAR APPLICATIONS



Stickybot climbs a window at Stanford University.

This article was contributed by Paul Day and Cameron Turner, Plutonium Manufacturing and Technology Division.

Biologically inspired robots such as the Stanford Stickybot mimic the abilities of biological organisms. Stickybot gets its name from the directional dry adhesives used for its feet. These adhesives, inspired by the natural mechanisms seen in geckos, leave no residue on surfaces and are capable of detaching with zero pull-off force. This capability is achieved through directionality of adhesion or the ability to resist a shear load in one direction while shear loading in the opposite direction causes the material to detach from the surface. Robots using feet made with directional dry adhesives show promise for a variety of applications in nuclear, chemical, and biological fields.

The Stanford Biomimetics and Dexterous Manipulation Laboratory conducts research into dry adhesive technology, including the duplication of hierarchical compliance seen in geckos. The gecko's adhesive pads rely on three levels of compliant microstructures with feature sizes as small as a few hundred nanometers, or billionths of a meter. These structures can achieve intimate contact with surface imperfections and generate adhesion through Van der Waals forces. Researchers at Stanford are currently attempting to reproduce these structures synthetically.

The current generation of synthetic directional dry adhesives uses directional polymer stalks. These specially shaped stalks are made of elastic polymer with feature sizes of a few hundred microns. Their special angled shape produces their directional

adhesive capabilities. After initially touching a surface, the stalks pull themselves further into contact as they are loaded along the contact surface and peel off smoothly when loaded in the opposite direction. Stickybot uses pads composed of hundreds of these stalks to adhere to and climb smooth surfaces such as stone, metal, and glass.

At Los Alamos, bio-inspired robots could be used to climb on the inside of gloveboxes and the associated systems (trolley, ventilation, etc), where they could navigate glovebox interiors, probing for contamination, leaks, or other conditions requiring human attention. Bio-inspired robots developed for hazardous nuclear and chemical environments could perform many of the typical maintenance, surveying, decontamination, and housekeeping activities. Someday, humans may conduct actinide research alongside and assisted by bio-inspired robots.

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