NMT Studies Fuel Fabrication Methods to Advance Efforts in Plutonium Disposition

The disposition of plutonium is an area of significant importance to the national security of the United States. In 1994 the Committee for International Security and Arms Control (CISAC), a standing committee of the National Academy of Sciences, conducted a study on the management and disposition of excess weapons plutonium. The committee concluded that the use of plutonium as fuel in existing or modified reactors with no subsequent reprocessing of the spent fuel is the leading contender for the long-term disposition of weapons plutonium. In addition to the inventory of excess weapons plutonium, the growing inventory of unseparated plutonium in spent nuclear fuel is a concern for maintaining peace and security on a global basis.

Using chemical separation methods, the plutonium in spent fuel can be recovered, as is done routinely in France and the United Kingdom, and subsequently used in nuclear reactors. Nuclear Materials Technology (NMT) Division personnel are studying two different processes for the eventual fabrication of plutonium fuel sources to power nuclear reactors and reduce the nation’s inventory of plutonium at the same time.

Approximately two years ago, the Department of Energy (DOE) formed the Office of Fissile Material Disposition (FMD), whose charter is to develop plans and technologies for the disposition of excess fissile material from the U.S. nuclear weapons program. The FMD is considering the option of converting weapons plutonium into mixed uranium-plutonium oxide (MOX) fuel for use in domestic or Canadian water reactors. Recently NMT Division did pioneer work on that option when they dissembled a pit from a nuclear weapon, separated the plutonium by the hydride-dehydride process, oxidized the plutonium, blended the PuO2 with UO2, and pressed and sintered a MOX fuel pellet.

Figure 1: On the left, plutonium dioxide produced from a dismantled weapon by the hydride/oxidation process. On the right, first MOX fuel pellet produced using the plutonium.
Another fuel under investigation is “nonfertile” fuel, which does not produce more fissile material than is consumed when it is burned in a reactor. Nonfertile fuel has the potential for allowing new or existing water reactors to become net consumers of plutonium instead of net breeders. The balance of this article discusses NMT’s study of nonfertile fuel.

At the request of the CISAC, Idaho National Engineering Laboratory (INEL) personnel investigated the feasibility of using a nonfertile fuel form for near-total destruction of weapons plutonium in existing or advanced light-water reactors. Neutronic performance results show the nonfertile fuel containing weapons plutonium to be a potential fuel for use in a pressurized-water reactor.

INEL evaluated the neutronic performance of a PuO$_2$-ZrO$_2$-CaO-Er$_2$O$_3$ fuel form suitable for use in a commercial boiling-water reactor. Plutonium oxide derived from weapons plutonium, calcia-stabilized zirconium oxide, and erbium oxide serve as the fuel, fuel diluent, and depletable neutron absorber, respectively. The results show this fuel form to be suitable for potential use in such a reactor.

**Los Alamos Studies**

The Los Alamos study of nonfertile fuel fabrication is supported with Laboratory Directed Research and Development Office funds. One goal of the study is to develop fuel fabrication methods that would allow weapons plutonium to be used as fuel in water reactors. Specifically, we have chosen the PuO$_2$-ZrO$_2$-CaO-Er$_2$O$_3$ evaluated by INEL as the fuel composition for our initial fuel fabrication study.

The first phase was the fabrication of a surrogate CeO$_2$-ZrO$_2$-CaO fuel. The purpose of the surrogate study was to 1) evaluate the feasibility of preparing the fuel by the solid-state reaction method using reagent-grade calcia (CaO), zirconia (ZrO$_2$), and ceria (CeO$_2$) as oxide precursors, 2) develop a powder comminution (pulverizing) method acceptable to glove box operations, 3) evaluate the behavior of PuO$_2$ in the fuel diluent using CeO$_2$ as the actinide surrogate, and 4) determine the specifications for a sintering furnace design and operation. The surrogate fuel enabled us to use a nonradioactive environment to study the effect of ball milling, green pellet formation, and sintering conditions on the microstructural development of a pellet of nonfertile fuel.

**Figure 2:** Scanning electron micrographs of ball milled powder: particle agglomerates and surface of agglomerate.
The equivalent spherical diameter of the precursor powders was determined using laser diffraction analysis. The particle size and morphology of the precursor powders was characterized using scanning electron microscopy (SEM). Sintered pellets were ground in an agate mortar and subsequently analyzed for crystalline phase content using x-ray diffractometry (XRD). Pellets were formed as follows: Reagent-grade ZrO$_2$ (87.19 wt%), CaO (10.12 wt%), CeO$_2$ (2.69 wt%), stearic acid (1 wt%), and polyethylene glycol (1 wt%) were dry ball milled for 24 hours. As shown in Figure 2, large (greater than 500 µm) agglomerates were formed as a result of ball milling the ZrO$_2$, CaO, and CeO$_2$, precursor powders for 24 hours. The scanning electron micrographs show a broad particle-size distribution. Submicron particles are visible on the surface of the agglomerates. The equivalent spherical diameter of the ball milled powder was determined to be 87.3 µm. The milled powder was uniaxially pressed into pellets at 310 MPa. The green pellets were sintered for 5 hours at 1200 °C, 1400 °C, and 1700 °C in an atmosphere consisting of 80% N$_2$ and 20% O$_2$. The bulk density and volume percent of open porosity were determined using the immersion density technique. Grain and pore structure including average size and size distribution were determined using optical microscopy and SEM analysis. As shown in Figure 3, significant increases in the bulk density of the surrogate fuel pellet occurred between the sintering temperatures of 1200 °C and 1400 °C. A significant decrease in open porosity (vol%) of the surrogate fuel pellet occurred between the sintering temperatures of 1400 °C and 1700 °C. Formation of the calcia-stabilized zirconia occurred as a result of sintering the fuel pellets at 1200 °C, complete solid solution formation between the surrogate (CeO$_2$) and the stabilized zirconia occurred as a result of sintering the fuel pellets at 1700 °C, and significant grain growth occurred as a result of increasing the sintering temperature from 1400 °C to 1700 °C.

Summary and Conclusions

Dry ball milling of the precursor powders did not produce a highly reactive powder for pellet fabrication (i.e., pressing and sintering). Future work will examine the feasibility of using vibratory and/or attrition milling methods to produce reactive precursor powders for the solid-state reaction synthesis. A significant increase in the bulk density of the surrogate fuel pellet occurred between the sintering temperatures of 1200 °C and 1400 °C. A significant decrease in open porosity (vol%) of the surrogate fuel pellet occurred between the sintering temperatures of 1400 °C and 1700 °C. Formation of the calcia-stabilized zirconia occurred as a result of sintering the fuel pellets at 1200 °C, complete solid solution formation between the surrogate (CeO$_2$) and the stabilized zirconia occurred as a result of sintering the fuel pellets at 1700 °C, and significant grain growth occurred as a result of increasing the sintering temperature from 1400 °C to 1700 °C.

The principal developers of this project are Kevin B. Ramsey and H. Thomas Blair of NMT-9, Actinide Ceramics and Fabrication.
Neutron Source Recovery Reduces the Nuclear Danger, Responds to National Need

In the past when radioactive neutron sources were manufactured and used extensively, the mechanisms for future disposal of those sources were not well-thought-out. Although their manufacture continues today (albeit on a reduced scale), there are still no federal or commercial programs to recover or store excess or unwanted sources. In addition, unwanted sources cannot currently be disposed of as waste because federal and state restrictions prohibit such disposal, and no disposal facilities for these sources exist in the United States.

Within the last several years, concerns have been raised about the potential risks to the public health and safety from aging neutron sources held by private companies, universities, and government entities. The aging of these sources, coupled with the increasing complexity of the licensing of nuclear materials, has made neutron source ownership more burdensome and costly. Defense downsizing and the economic downturn in the oil and gas industry have made many neutron sources unnecessary; however, source owners who want to get rid of their excess or unwanted sources have no options for doing so. The consequences are both economic and legal. Alternatives such as improper storage or illegal disposal will lead to public health and safety risks as well. Los Alamos National Laboratory, through the Neutron Source Recovery Project, is attempting to alleviate this situation by providing source owners a safe and legal option for disposal of their nuclear material.

Emergency Responses

Over the past two years, the nation has called upon Los Alamos personnel to assist in the removal of abandoned or damaged neutron sources from a variety of locations. In all of these cases, the sources were determined to be a potential threat to public health and safety. Six sources were retrieved from a site in Oklahoma, where they were abandoned.
by a oil well logging company that had gone out of business. Three abandoned sources were picked up from a derelict vehicle parked for nine months in a vacant lot in Illinois. A damaged source, leaking radioactive material, was retrieved from a storage shed located in a residential neighborhood in large town in Texas. The source was breached in an oil well logging incident that resulted in contamination of the well site, a logging truck, and a storage shed.

Shipping
A significant part of this project is the coordination of shipments and the receipt of nuclear materials. Project staff provide detailed procedural information to the owners of the surplus neutron sources about packaging, monitoring, and shipping. Because of the stringent requirements for shipping nuclear materials, Los Alamos personnel work closely with the shippers to ensure compliance with all regulations. Our highly trained shipping and receiving personnel then efficiently unpack and batch the sources to the reprocessing area.

Dismantlement and Processing
Neutron sources are typically made from long-lived, radioactive materials mixed with a low-atomic-weight, nonradioactive material and doubly encased in small metal containers. These stainless steel (and sometimes tantalum) capsules must be removed to facilitate the recovery process. A remotely operated decladding cutter, using the principles of a conventional pipe cutter to reduce or eliminate metal turnings, is used to remove the capsules from the neutron source material. The source material is then dissolved in acid to separate out the radioactive elements and reduce the neutron emissions to background levels. It is estimated that the separated source materials require 1/700 of the storage space of the original source.

Remote Material Processing Capability at TA-55
A computer-controlled system allows remote handling of the decladding and dissolution operations. The process also eliminates the need for interim storage, thus reducing operator exposures by approximately 300 percent. The equipment has been used reliably in the harsh glove box atmosphere to process more than 100 neutron sources.

Engineers are currently reconfiguring the neutron source processing operations to further reduce operator exposures. The new processing line concept employs the use of remote manipulators similar to those used in hot-cell operations. Remote handling capabilities coupled with traditional glove box flexibility should add a new dimension to the existing capabilities at the Plutonium Facility. We will be able to handle highly radioactive materials of all types with greatly reduced exposure to the operator. Other programs that handle large amounts of americium or other highly radioactive materials should be able to utilize this unique processing capability to reduce operator exposures significantly.

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Division Director Discusses Plutonium Future Part I

“And they shall beat their swords into plowshares, and their spears into pruning hooks: nation shall not lift up sword against nation, neither shall they learn war anymore.” Isaiah 2:4

Conversion of weapons plutonium into mixed uranium-plutonium oxide fuel for energy production in civilian power reactors could make the old prophet’s vision come true. In fact, technologists at Technical Area-55 made the first step in that direction last year when they disassembled a pit, separated the plutonium by the hydride-dehydride process, oxidized the plutonium, blended the PuO₂ with UO₂, and pressed and sintered a MOX fuel pellet. It was a simply symbolic but truly significant piece of technology demonstration.

The use of nuclear materials has always had two sides: peaceful energy for civil electrical power—and weapons of mass destruction. This dichotomy has always been carefully separated in the minds of policy makers, grudgingly accepted by scientists and engineers, but closely connected in the minds of the public.

Today, fission energy produces about 20% of the world’s electrical power, but less than 1% of the energy value of the uranium fuel material is extracted. On the other hand, the United States and the Former Soviet Union produced tens of thousands of nuclear warheads, numbers that are substantially beyond any reasonable requirement for defense or any conceivable act of aggression. Why military development of fission energy has been carried beyond rational need, while peaceful development has been suppressed in this country is beyond understanding. But indeed that has been the political position; current U.S. policy, established by the Carter Administration and executed by the Clinton Administration, discourages the use of plutonium for civil purposes. European countries, Japan, India, and Russia are increasing or planning to start the recycling of plutonium in light-water and breeder reactors, while in this country, reprocessing, recycling, and breeding are nearly extinct technologies. While the rest of the world is developing greater reliance on nuclear power, the U.S. is on a path to foreclose on that option, which many feel will be essential for electrical power, economic growth, and protection of the environment in the next century. Nevertheless, the U.S. is getting about 7% of its electricity from plutonium created by in situ fission in the cores of light-water reactors.
More plutonium has been synthesized than any other man-made element, and because of its high energy content and radioactive properties, plutonium is both attractive and hazardous at the same time.

**Plutonium: the most toxic substance known to man.**
**Plutonium: the enabler of world peace.**
**Plutonium: enough to kill everyone in the world many times over.**
**Plutonium: enough energy to power the world's economy for centuries.**

The extremes are as varied as the people with opinions. So where is the truth and what should we do with the plutonium? The fact is that there is a glut of plutonium in the world today. First isolated and identified in 1941 by Glenn Seaborg, plutonium has proliferated from micrograms to hundreds of tons. In the U.S. alone there are 99.5 metric tons left over from the nuclear weapons buildup during the Cold War. An equal or greater amount exists in Russia, the United Kingdom, France, and China, the other declared nuclear weapons states. A recent DOE study has identified 26 metric tons of plutonium wastes in various forms of solids, compounds, residues, and solutions that are unsuitable for long-term storage. This "legacy plutonium," left over from the production campaigns during the Cold War nuclear weapons buildup, is currently stored at various sites in the weapons complex. Los Alamos has 2.6 tons of plutonium; about 95% of this plutonium resides here at TA-55.

Tens of thousands of nuclear warheads containing plutonium pits were produced during the Cold War. Fortunately, because of worldwide political events and activities initiated by the Bush Administration and continued by the Clinton Administration, these worldwide nuclear stockpiles are being reduced, as planned by presidential directives and international treaties. In 1991 the United States and the Soviet Union signed the Treaty on the Reduction and Limitation of Strategic Offensive Arms (START I), and both countries are dismantling nuclear weapons, reducing stockpiles to 8,000 to 9,000 each. The START II treaty, which is still under negotiation, could reduce strategic warheads to less than 3,500 each. In addition, the legacy wastes in the U.S. are being stabilized and prepared for long-term storage.

These international issues are defining the future of TA-55 technologies. The goal of the Stockpile Surveillance Program is to ensure the reliability and safety of the enduring stockpile, the Advanced Recovery and Integrated Extraction System will demonstrate an automated process for dismantling plutonium pits, the Pit Rebuild Program will maintain the de minimis capability for manufacturing plutonium components, the Enhanced Surveillance Program will determine the effects of plutonium property changes on the reliability of aging weapons, and the 94-1 Residue Reduction Projects are aimed at stabilizing, separating, and storing legacy plutonium wastes.

The reduction of nuclear weapons and the stabilization of nuclear wastes is the correct thing to do; however, the activities have raised new concerns over safety, security, and final disposition of the plutonium. In the next Actinide Research Quarterly, I will discuss this approach and its implications for the nation and TA-55.

Bruce Matthews
NMT Designs and Fabricates Standards for Nuclear Material Assay

How good is a nondestructive assay (NDA) measurement? A measurement done by an NDA instrument is only as good as the standard that is used to calibrate the instrument. Standards are needed to calibrate various NDA instruments such as neutron coincidence counters, gamma-ray counters, and calorimeters. These instruments measure a variety of nuclear materials being produced in the DOE nuclear community. The measurements help alleviate problems associated with shipper/receiver differences and the measurement and storage of residues and waste.

Los Alamos National Laboratory has taken a lead role in the fabrication of uranium and plutonium standards, along with other actinides such as neptunium and americium. These standards have been fabricated for several laboratories within the DOE complex.

Planning the fabrication of standards requires very precise detailing. Designs encompass components such as precise weighing, destructive analysis of samples, specialized containers, diluents, and the use of post-fabrication NDA measurements to confirm that the standards meet all preliminary expectations before they are used in instrument calibration.

Each NDA instrument varies in the amount of material it is able to measure. Segmented gamma scanners (SGSs) and neutron counters are able to generate precise measurements from 0 grams to 250 grams. Calorimeters, depending on chamber size, are able to measure from 0 grams to 5 kilograms. With these limits in mind, various amounts of nuclear materials are needed to accommodate the calibration ranges of the instruments. The standards also need to demonstrate linearity in the entire range of NDA measurements. For a given set of standards, the instrument reading should be linearly dependent on the quantity of the materials being measured.

Standards should be fabricated of material similar to that of the actinide being measured. We do not calibrate an instrument with pure plutonium metal if we are measuring oxides or waste. Measurements on the SGS are done on residues or low-density materials. Neutron instruments measure high-density materials such as metals, piping, residues not exhibiting high alpha-neutron emission, glass, and leaded gloves.

Every standard fabricated is created from a highly pure actinide material. The material is roasted, sieved, blended, and sampled. The analysis of the sample consists of isotopic compositions (enrichment), actinide percent purity, and levels of impurities associated with the material, such as iron or lead. A variety of the standards have been subjected to multiple assays by several analytical laboratories. The multilaboratory analysis is used to minimize the bias of any one laboratory assay and to ensure the homogeneity of the blended batch. Multiple samples also give results that help to develop good statistical comparisons among samples and laboratories.
Another important element in the fabrication of calibration standards is the containers that hold the standards. The container shape/size configuration can affect the radiation and thereby the NDA measurements. Each standard-fabrication assignment is carefully analyzed during the planning stage for the construction of a container that is easily handled in the glove box and compatible with the instrument it was designed for. SGS can standards are specialized cans eleven inches in height and four inches in diameter (Figure 6). These are ideal dimensions to help alleviate problems in end-effects and for gamma-ray transmission through the standard. SGS standards consist of oxide diluted with diatomaceous earth, which is used to homogenize and transfer the oxide uniformly throughout the container.

SGS drum standards consist of twenty-four-liter polyethylene bottles, each containing a known amount of oxide diluted in diatomaceous earth. These are stacked in a 55-gallon drum to simulate a homogeneous drum standard. New standards for neutron shuffler drums consist of oxide diluted with diatomaceous earth in one-inch diameter zirconium vials (Figure 7). The vials are stacked in a 55-gallon drum and can be varied for instrument calibration simply by adding or deleting vials to the drum. New calorimeter standards are now being fabricated with 12% plutonium-240 oxide. These will be distributed throughout the complex and are measured throughout the year to compare calorimetry measurements among labs. They also will provide us with a higher-wattage standard to complement the 6% plutonium-240 wattage standards that already exist. These standards consist of two mechanically sealed, “food-packed” cans containing two kilograms of high-burnup oxide.

With the problems associated with shipping materials today, details on dimensions of the standards need to be compatible with the shipping containers. Containerization must meet all requirements for shipping; therefore, certification of packaging demands double- or triple-encapsulated containers depending on what Department of Transportation drums are used.

Designs are greatly affected by these restrictions. It is therefore imperative that research be done before standards are fabricated. This will alleviate future problems in supplying a compatible standard used in measuring the various types and amounts of actinide materials and complying with all shipping requirements.

There has been and always will be a continuous demand for calibration standards throughout the NDA community. With new and better technology, Los Alamos will be the forerunner in producing these needed standards for the DOE complex and for other actinide measurement users.

Figure 7: Zirconium tubing for shuffler standards. Oxide is diluted with diatomaceous earth in these one-inch diameter vials.

The main designers of the standards work are T. Hsue, S. M. Simmonds, J. K. Sprinkle, and P. M. Rinard, all of NIS-5, and V. L. Longmire and S. M. Long of NMT-4.
Neutron Source Recovery Reduces the Nuclear Danger, Responds to National Need (continued from p.5)

International Request
Los Alamos is currently responding to an international request to reprocess 316 Pu-239/beryllium neutron sources currently owned by the German government. These sources were originally part of East Germany’s nuclear materials inventory. Most of them are relatively small and should be accommodated easily in existing TA-55 capacities. The German request expands our response to sealed-source disposal problems into the international arena.

Responding to an Expanded National Need
The neutron source program will be expanded to recover additional types of neutron sources. This effort, the “Radioactive Source Recovery Program,” will be sponsored by the U.S. DOE, Environmental Management Program Office. The Department has noted the experience, personnel expertise, and unique facilities that exist at Los Alamos as well as our excellent track record of delivering on emergency requests.

The scope of this program will include the routine recovery of Am-241 and Pu-238 neutron sources at both TA-55 and the CMR Building. Use of the CMR Wing 9 Hot Cells will enable us to handle larger Am-241 sources as well as the recovery of Pu-238 neutron sources. While the starting date for receiving sources is presently uncertain, the initial planning phases of the project are well underway. An environmental assessment has been completed for the expanded capability at the CMR Building with the finding of “no significant impact.” Strong support has been received from people across the nation and state who have a stake in our business, and press reports have stated the recovery of neutron sources is valuable for reducing radioactive risks to health and safety nationwide.

Advisory Committee Rates NMT as “Outstanding/Excellent”

The results of the 1995 Science and Technology Assessment for NMT Division were announced in the NMT Division Review Committee’s (DRC’s) final report delivered to Laboratory Director Sig Hecker in late January. The charter of each DRC, appointed by the Laboratory Director, is to assess a given division’s science and technology and to advise the Director and Division Director on important issues. The overall Laboratory Science and Technology Assessment based on the various DRC reports is then presented to the Science and Technology Panel of the University of California President’s Council on the National Laboratories. The NMT Division’s accomplishments were highly praised during the review period, July 1, 1994, to June 30, 1995. The Committee noted dramatic improvements on all fronts with an “outstanding/excellent” rating in the division’s overall performance. The next Division Review is slated for March 1997. The main themes will be “Stockpile Stewardship” and the “Space Mission.”
Publications, Presentations, and Reports (December 1995–March 1996)

Journal Publications


Conference Presentations

The following papers were presented at the American Chemical Society Meeting in New Orleans, LA, during March 24-29, 1996:


The following papers were presented at the 4th International Conference on Nuclear Engineering in New Orleans, LA, during March 10-14, 1996:


The following papers were presented at the 3rd International Policy Forum, Management & Disposition of Nuclear Weapons Materials, Lansdowne Executive Conference Center, Lansdowne, Virginia, March 19-22, 1996:

S. M. Dinehart, “Plutonium Stabilization Research and Development.”


Reports


The following NMT members have been appointed to the Laboratory "core competency" teams: Larry Avens in Complex Experimentation and Measurement, Brett Kniss in Nuclear Weapons Science and Technology, and Walt Stark and Steve Yarbro in Nuclear and Advanced Materials. The teams operate under the guidance of the Science and Technology Base (STB) Program Office and the Core Competency Senior Advisory Committee, composed of a number of the Laboratory directors. The teams provide a link with the Laboratory technical staff and report quarterly to Technical Working Group of the Laboratory Leadership Council (LLC) on the "status of the science and technology base."

In addition, Tim Nelson was appointed to the Science and Engineering Advisory Council, which reports directly to the director of STB Programs and provides input to the LLC working groups. Other NMT members continue to contribute to a number of advisory groups. Looking at all the recent appointments, it appears that these days the Laboratory listens when the NMT Division talks.

NMT Earmarks Funds for Awards

The Los Alamos Award Program (LAAP) enables Lab managers to recognize the exceptional contributions and noteworthy achievements of their employees in a timely manner. NMT will earmark a sizable portion of its LAAP funds to recognize and reward the accomplishments of individuals and teams for their science and technology efforts.

The cash awards will recognize excellence in publications, patents, technology transfer, major program developments, science education, and other technological innovations. A team headed by Chief Scientist K. C. Kim will review nominations and make recommendations to Division Director Bruce Matthews. Nominations for single individuals or teams may be made at any time, but nominations for this year's awards are due to Kim by May 31. Self-nominations are welcome. The nomination should describe specific accomplishments within the period June 1, 1995, through September 30, 1996. If you have any questions or suggestions about NMT's part in the LAAP, contact Kim at 7-7753 or via e-mail: kck@lanl.gov.

Publications, Presentations, and Reports (continued)


