Los Alamos National Laboratory

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Nuclear Fuels

IN THIS ISSUE

ABOUT THE COVER

New nuclear fuels are an important part of advanced fuel cycles. The image on the cover is of hydrated hafnium hydroxide beads, which are used as stand-ins for uranium in development tests of new tri-isotropic (TRISO) fuels. TRISO fuels will be used in high-temperature gas-cooled reactors (HTGRs), which are under development as an advanced reactor type. In addition to providing electric power, HTGRs could provide an important source of hydrogen for fuel-cell-powered vehicles, which were highlighted in President Bush's 2003 State of the Union Address. The hydrated hafnium hydroxide beads are formed from aqueous solution and are colorless; they were photographed against a blue background for this photo. The story begins on Page 7.

Excess plutonium disposition

Aqueous polishing of plutonium oxide for mixed-oxide fuel

This article was contributed by Randy Erickson of Nuclear Materials Technology Division (NMT-DO), Kent Abney of Actinide Process Chemistry (NMT-2), Kevin Ramsey of Pit Disassembly and Nuclear Fuels Technologies (NMT-15), and Deborah Dale of Actinide Analytical Chemistry (C-AAC).

As a result of the end of the Cold War and related reductions in the nuclear arsenals in the United States and Russia, many tons of plutonium are now surplus to national security requirements. In both countries, the principal planned method for the disposition of surplus plutonium is to use the plutonium in the form of mixed-oxide (MOX) fuel to generate electricity in existing commercial reactors. The responsibility for this program resides in the Office of Fissile Materials Disposition of the National Nuclear Security Administration (NA-26).

Los Alamos is involved in two key technical aspects of the U.S. plutonium disposition program: the demonstration of key technologies for the disassembly and conversion of plutonium weapon components (pits), and the polishing—or final purification—of plutonium oxide to provide material for fabrication of MOX lead test assemblies to support the fuel qualification and licensing process.

The demonstration of key technologies for the disassembly and conversion of plutonium weapon components has been accomplished with the Advanced Recovery and Integrated Extraction System (ARIES) installed in the TA-55 Plutonium Facility. The ARIES equipment is configured in several connected modules to perform the sequential steps involved in disassembly and conversion of parts to unclassified forms.

Two demonstration phases have been completed, establishing the requirements for the disassembly of each of the 32 pit types in the surplus stockpile. Currently, several modules are being upgraded to reduce operator exposure and make the equipment more suitable for a future production environment.

The culmination of this effort is the development of build-to-print drawings of the key process equipment to be installed in the Pit Disassembly and Conversion Facility, to be constructed at the Savannah River Site. Currently, this design activity is in the latter stages of Detailed (Title II) Design.

The plutonium from the ARIES processing is an unclassified form of plutonium oxide. The Office of Fissile Materials Disposition initiated a project in Fiscal Year 2002 to purify this type of material to specifications that would allow direct use in the fabrication of MOX-fuel lead assemblies. The initial project plan included an initial 5-kilogram demonstration of the polishing process to confirm that the required product purity could be achieved, followed by a production phase to generate a total of about 140 kilograms of high-purity plutonium oxide. To meet external schedule requirements, the material needs to be ready to ship by August 2004.

Aqueous Polishing

During the course of the 5-kilogram demonstration, several issues were encountered that resulted in additional demonstration activities. First, there were some chemical impurities in the product material that either exceeded specification levels or were not able to be measured to the required sensitivity. Second, some of the original equipment used for the demonstration was shown to be unreliable. Third, there were deficiencies in the quality-assurance practices that were identified.

There has been a concerted effort to resolve all of these issues over the past year. The final confirmation that Los Alamos can meet required product quality and production capacity is targeted for this July, which is an Appendix F milestone for the Laboratory.

Aqueous polishing process

The aqueous polishing process uses a conventional process-flow diagram of dissolution, ion exchange, oxalate precipitation, and calcination. Several batches of calcined plutonium oxide are blended to reduce the overall number of samples needing to be analyzed to confirm that the final product meets specification. Finally, the plutonium oxide is packaged in crimp-sealed convenience cans, placed inside welded containers, and stored until future shipment to a MOX fuel fabrication facility.

The primary equipment used for the polishing function is the Advanced Testing Line for Actinide Separations (ATLAS). This glovebox line was installed in the early 1990s to provide a flexible capability for full-scale separations research. The configuration used for this polishing project includes:

Process-flow diagram for aqueous polishing.
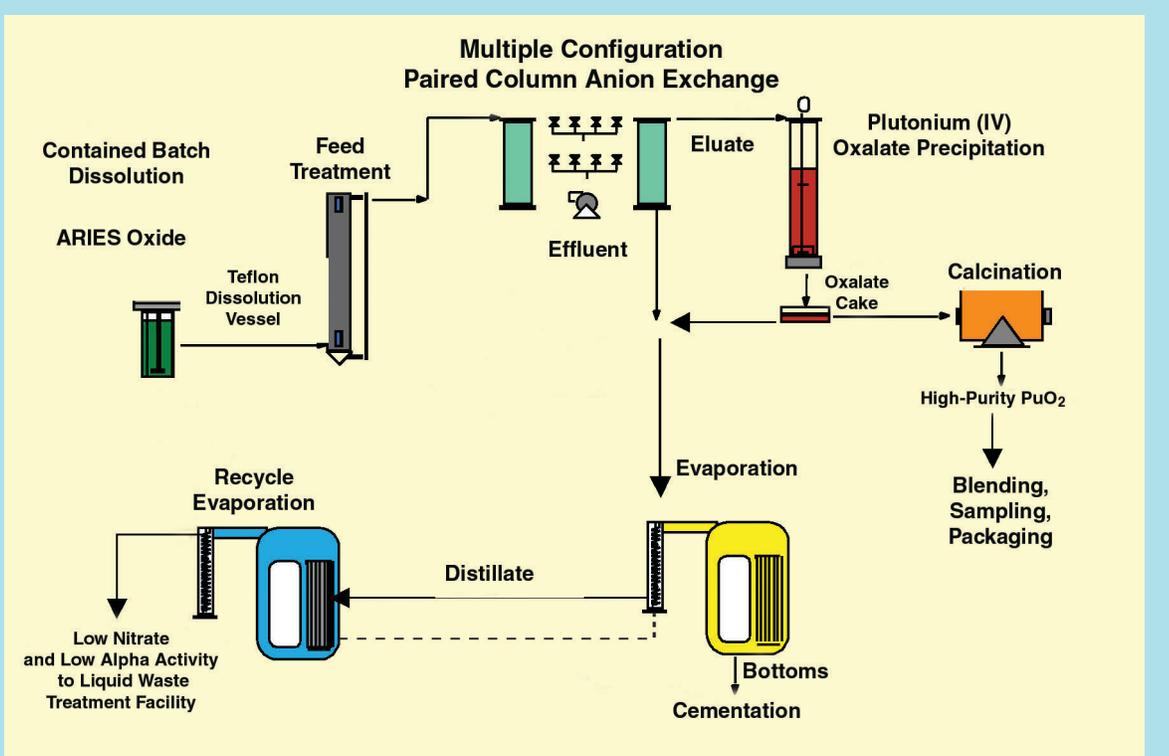
- nitric acid/hydrogen-fluoride dissolution in heated borosilicate glass vessels (or Teflon vessels recently installed to eliminate a potential source of boron contamination);
- ion exchange in two 3-foot columns using Reillex-HPQ™ resin with at least 400 liters of wash volume to maximize the final product purity;
- plutonium (IV) oxalate precipitation at approximately 65 degrees Celsius to help ensure proper oxide-powder morphology and to complete the purification efforts;
- calcination in a conventional muffle furnace using rigorously controlled heat, ramp, and soak temperature profiles;
- blending in a Turbula blender to allow larger master blends;
- trace and physical analyses of the blended product samples;
- packaging in a Cogema convenience can;
- welding into an ARIES inner can and decontaminating the welded can using the ARIES electrolytic decontamination module; and
- loading the welded cans into FS-47 shipping containers.

The aqueous and dry-powder operations are performed in Actinide Process Chemistry (NMT-2), the trace and isotopic analyses by Actinide Analytical Chemistry (C-AAC), the physical analyses and canning by Pit Disassembly and Nuclear Fuels Technologies (NMT-15), and packaging for shipment by Safeguards and Security (NMT-4).

Several analytical techniques are being used to evaluate the product plutonium oxide. Most of these techniques are well established and have been used to support a variety of programs at Los Alamos, the exception being the Coulter Counter for particle-size analysis. This apparatus was installed after the early demonstration activities showed that the laser-scattering instrument was providing insufficiently accurate results.

Results of the polishing demonstration

The 5-kilogram demonstration was intended to establish the baseline unit operations in the process-flow diagram and should have been confirmatory in nature. However, several issues—impurity, equipment problems, and quality control—were discovered during the course of the initial operations that required corrective actions.

At the onset of the demonstration, it was recognized that removing gallium from the plutonium feed materials would be challenging. To avoid any issues with the process to license the MOX fuel, an extremely low specification for gallium was established—comparable to levels that would be seen in conventional fuels over time as a result of fission-product buildup.
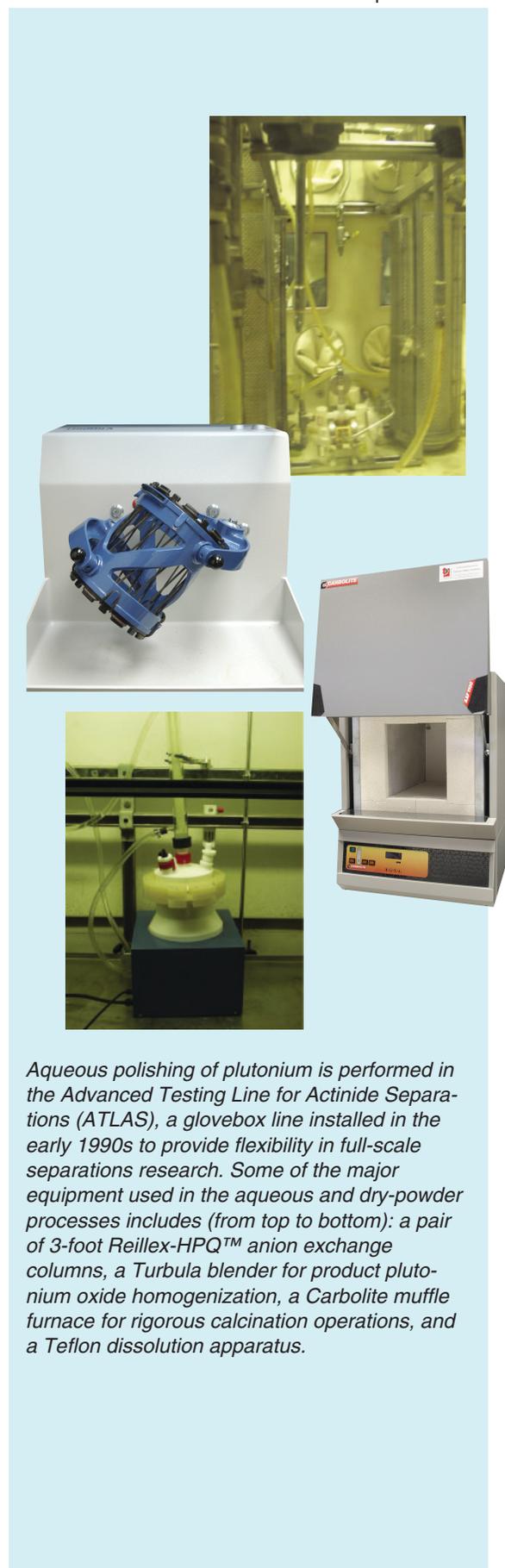
Aqueous polishing of plutonium is performed in the Advanced Testing Line for Actinide Separations (ATLAS), a glovebox line installed in the early 1990s to provide flexibility in full-scale separations research. Some of the major equipment used in the aqueous and dry-powder processes includes (from top to bottom): a pair of 3-foot Reillex-HPQ™ anion exchange columns, a Turbula blender for product plutonium oxide homogenization, a Carbolite muffle furnace for rigorous calcination operations, and a Teflon dissolution apparatus.

The early results showed the final product gallium concentrations declining from run to run, perhaps as the process equipment was decontaminated. However, detection limits were also above the specification. Negotiation with DOE (NA-26) resulted in a slight relaxation of the specification. In addition, feed materials have been limited to those that are already relatively low in gallium. Also, the measurement sensitivities have been improved so that the detection limit is now below the fuel specification.

Analysis	Technique
Plutonium assay	Coulometric
Plutonium isotopics	Thermal Ionization Mass Spectrometry (TIMS)
Impurities (cations, lanthanides, actinides, and phosphorus)	Inductively Coupled Plasma-Mass Spectrometry/Atomic Emission Spectroscopy/Thermal Ionization Mass Spectrometry (IDMS)
Impurities (anions)	Ion chromotography
Moisture content	Loss-on-ignition
Particle size	Coulter Counter
Bulk and tap density	ASTM method
Surface area	Gas adsorption (BET method)
Carbon	Combustion IR

The net result is that the gallium levels in product materials are now routinely below the revised specification.

Over the course of the demonstration, it was also determined that there were similar issues with boron in the product material. The detection limits for boron were often at the specification limit, and routinely there would be an indication of impurity levels at or slightly above the detection limit. A potentially large source of boron impurity is the borosilicate glass dissolvers used in the dissolution process. The walls on these dissolvers are etched rapidly, requiring their replacement every five to six runs.

Teflon dissolvers have been installed to eliminate this source of boron. Several batches have been run through the Teflon dissolvers. However, analytical results for boron have not yet been completed. Even with the Teflon dissolvers, there is still an issue that detection limits are above the specification. Discussions are currently going on with NA-26 about relaxing the specification for boron as well, or using other DOE analytical laboratories that have lower detection limits.

During the initial demonstration, several of the major pieces of equipment were shown to have problems. The original calcination furnaces exhibited control problems, so new furnaces and controllers with computer-data logging were installed. The original V-blender was replaced with a Turbula blender to provide a more homogeneous product in a shorter time. The Turbula blender also allows for larger batch sizes to reduce the number of samples needed to characterize the final product material and to conform to standard industrial blending approaches.

As mentioned earlier, the laser-scattering particle-size analyzer was shown to provide inconsistent results. A Coulter Counter was installed to address this issue and to provide a capability that was directly comparable to previous industrial results.

The initial demonstration uncovered some significant quality-control issues. The primary methods for demonstrating product quality are the analytical measurements for product characteristics against the specification. These capabilities at Los Alamos have robust quality-management programs because of the recent efforts to re-establish pit manufacturing at Los Alamos.

There are other areas, however, that required an effort to establish an adequate quality program. For example, data traces for the

calcination-furnace temperature profile were not archived. Although operators confirmed that requirements were met in the data records, it is appropriate to archive available data to allow verification that the requirements were met. Also, there were incomplete and inconsistent entries in the data records.

Even though NA-26 had reviewed and approved the NMT Division quality program, it was clear that a specific project-level Quality Assurance Plan was needed to clearly identify the appropriate process control and documentation requirements for the day-to-day operations by personnel. Implementation of this quality plan is nearly complete, in anticipation of several internal and external QA audits over the next few months.

Future activities

Over the coming weeks, there will be a great deal of activity to demonstrate that all aspects of the product specification can be met, to demonstrate a complete implementation of the quality-assurance program, and to demonstrate that there is sufficient throughput to generate the required quantities by August 2004 even if there are unforeseen interruptions to the operations.

This article provides an overview of the significant work being performed by NMT-2, NMT-15, and C-AAC personnel to address the challenging endeavor of disposing of excess plutonium through aqueous polishing. Many aspects addressed briefly in this article will result in future articles that will describe in more detail the efforts that have been and will be accomplished to install and qualify equipment, improve measurement approaches, and establish quality operations.

Readers are encouraged to look for articles in future issues of ARQ from the following people: Fawn Coriz, Liz Bluhm, and Simon Balkey of NMT-2; Lawrence Drake of C-AAC; and Brian Bluhm and Jane Lloyd of NMT-15.

Larry Gibbons

In the top photo, Joe R. Martinez (left) and David Martinez, both with Actinide Process Chemistry (NMT-2), sample a stabilized plutonium oxalate batch (now oxide) for chemical trace analysis, particle morphology, and moisture content. The photo immediately above was taken through a glovebox window and shows plutonium oxalate cake in fused-silica roasting boats. The material is dried before it is placed in a stabilization furnace, where it is converted to plutonium dioxide. The actual color (which is skewed by the glovebox window reflection) is rusty orange; when converted to oxide it turns an "army" green.

Los Alamos delivers Qual-1 pit

With the successful, on-schedule delivery of a certifiable plutonium pit this past April, Los Alamos has re-established the United States' capability to manufacture nuclear weapons cores. That capability was lost when the Rocky Flats Plant in Colorado closed in 1989.

The effort to produce what is called the "Qual-1" pit took six years and more than 700 people, many of them from the Nuclear Materials Technology (NMT) Division. The newly manufactured pit is called Qual-1 because it was made with fully qualified processes. The pit is for the W88 warhead, which is carried on the Trident II D5 submarine-launched ballistic missile.

The Qual-1 pit is the first pit to be manufactured under a program to replace cores in the W88 warhead, which is carried aboard the Trident II submarine-launched ballistic missile.

"The Laboratory delivered on a major commitment to the Department of Energy's National Nuclear Security Administration, Congress, and the taxpayers," said Pete Nanos, Los Alamos' interim director, in making the announcement at Los Alamos' 60th anniversary celebration. "Our next challenge is to carry out the required experiments, analyses, and computer modeling so we can certify that this newly manufactured pit will perform reliably in the stockpile, without conducting underground nuclear tests."

The DOE selected Los Alamos in 1996 as the site to recapture the nation's ability to manufacture nuclear weapons pits in part because the Laboratory has the country's only full-capability plutonium facility and has made pits since the 1940s.

Los Alamos has made 18 pits under the current program, the Pit Manufacturing and Certification Integrated Project Plan. The first pit, called early Development Unit-1, was completed in February 1998. Last August the Laboratory made the first pit that used all 42 processes required for a certifiable pit, and in December of last year, the Lab completed the qualification of all 42 processes.

Los Alamos will make about half a dozen pits a year from now until 2007 with a goal to begin making 10 stockpile pits a year by 2007.

*National program will develop and implement
spent-fuel treatment and transmutation technologies*

Advanced Fuel Cycle Initiative: Closing the nuclear fuel cycle

Two full years of oil imports—the untapped energy in the spent nuclear fuel currently stored in the United States—is the potential-energy equivalent of a staggering six-billion barrels of oil. Such is the bounty—and the challenge—facing team members of the Advanced Fuel Cycle Initiative (AFCI).

AFCI, a national program directed by the DOE Office of Nuclear Energy, Science and Technology, teams Los Alamos; Idaho National Engineering and Environmental Laboratory; Savannah River Technology Center; and Oak Ridge, Argonne, and Lawrence Livermore National Laboratories.

AFCI's stated goal is to enable the future of nuclear power by developing and implementing spent-fuel treatment and transmutation technologies to enhance the performance of the proposed high-level waste repository and reduce the cost of geologic disposal for the United States.

The Los Alamos team is currently centered in AFC-PO (Advanced Fuel Cycle Project Office), with Michael Cappiello as the Los Alamos program manager, Kemal Pasamehmetoglu the project's national director for fuels, and Cappiello its national director for transmutation technology.

The Los Alamos AFCI work uses the skills of personnel from many divisions, including Nuclear Materials Technology (NMT), Physics (P), Chemistry (C), Decision Applications (D), Los Alamos Neutron Science Center (LANSCE), Materials Science and Technology (MST), Applied Physics (X), Theoretical (T), Engineering Sciences and Applications (ESA), and Nonproliferation and International Security (NIS).

Spent nuclear fuel comprises the "waste" or byproduct of typical light-water reactors. It consists largely of uranium dioxide (about 96 percent) and a hodgepodge of elements produced by the fission and neutron absorption processes within the fuel, including plutonium, neptunium, and other higher actinides (americium, curium, etc.), as well as fission products such as the lanthanides and lighter elements like strontium, krypton, and cesium, among others.

The geologic repository referred to in AFCI's goal is the proposed Yucca Mountain site in Nevada. At the current 2,000-metric-ton annual rate of spent fuel production by electricity-generating nuclear power plants, its statutory capacity will be reached in 2015. A decision for a second repository will be made in the 2007 to 2010 time frame.

Commercial light-water reactors (LWRs) account for one-fifth of the electricity production in the United States, one-sixth globally. In addition to the obvious implication for independence from fossil-fuel electricity generation, their great advantage lies in their minimal generation
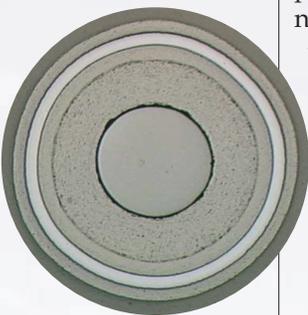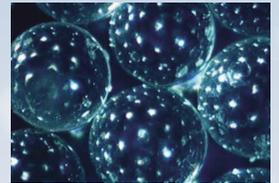
of the greenhouse gas, carbon dioxide, and therefore, their positive impact on mitigating global climate change.

Moreover, in the face of dwindling oil supplies and an overdependence on the oil of volatile Middle Eastern nations, nuclear power offers the possibility of greatly assisting a hydrogen-based domestic economy, for example, by providing hydrogen for automotive fuel

Advantages of Advanced Fuel Treatment

cells without the release of additional carbon into the atmosphere. Nonetheless, many scientists believe that light-water reactors can remain a viable alternative to greenhouse-gas-producing (largely coal-fired) electric-power plants, only if researchers can ultimately close the nuclear fuel cycle.

In general, this means treating spent nuclear fuel to reduce its volume, its radiotoxicity, and its decay-heat load, thereby greatly altering what remains for geologic storage. In the process, additional energy is extracted. This is akin to burning trash, in the sense that it changes the form of matter in the waste

(molecular rearrangement in trash burning, elemental transmutation in the case of the closed fuel cycle) and liberates energy.

However, there are consequences in each case: Trash burning produces undesirable carbon dioxide, and a closed fuel cycle using today's technology yields separated streams of fission products and recycled plutonium. The minor actinides are customarily sent to waste (an outcome that would be altered if they could be reintroduced into a transmuter).
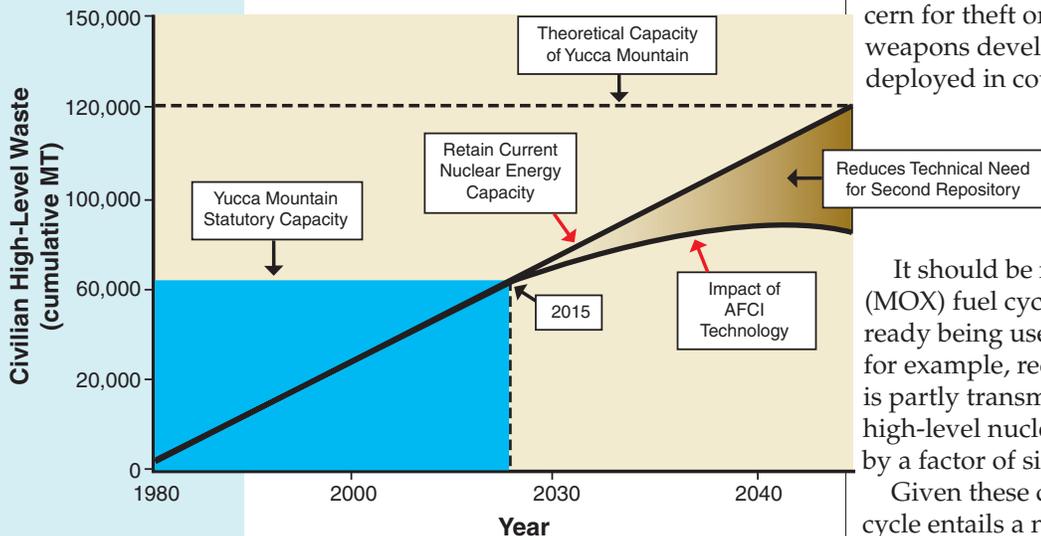
The recycled plutonium is potentially a concern for theft or diversion to clandestine weapons development if the technology were deployed in countries that did not already possess nuclear weapons. The minor actinides greatly increase the radiotoxicity (i.e., cancer risk) of waste requiring geologic disposal.

It should be noted that the mixed-oxide (MOX) fuel cycle (see ARQ Spring 1996) is already being used in other countries. In France, for example, recycled plutonium in MOX fuel is partly transmuted in power reactors and the high-level nuclear waste is reduced in volume by a factor of six.

Given these complex issues, closing the fuel cycle entails a number of complementary approaches. In general, AFCI project activities encompass improved nuclear fuels, actinide separations, separate management of fission product elements, advanced safeguards, and transmutation capability—the ability to consume transuranic actinides in the spent nuclear fuel, such that a smaller quantity of lower-radiotoxicity, reduced-decay-heat waste products will result.

The actinide transmutation activity parallels the age-old alchemist's goal of "lead into gold." In this instance, the operant descriptor might be "waste into dollars," given that, overall, the project could result in a net savings of between $35 billion and $50 billion, a

combination of revenue from rescued-fuel energy production and savings from the delay or elimination of the need for a second geologic repository beyond Yucca Mountain.

The latter goal is readily attainable, given the projected reduction in waste volume to about one percent of what is currently targeted for geologic storage. "Hopefully, if we're successful, we'll get by with one repository forever," commented AFCI national director for separations, James Laidler of Argonne.

Another benefit of closed fuel cycles is what can be characterized as the fissionable resource advantage. In the face of long-term dwindling global supplies of uranium, the ability to separate and recycle fissionable materials from spent fuel is viewed as an important plus.

A long-term initiative in two parallel phases

Currently in the first five-year period, during which the focus is the development of proliferation-resistant fuels and of separations processes for current light-water reactor spent fuels, the research is geared toward evaluating technologies for the deployment of a spent-fuel treatment facility in 2015.

In addition, researchers must identify candidate transmutation systems, whose deployment will likely not occur until at least 2022. This sort of timeline illustrates the program's complexly interdigitated activity matrix.

Termed "Series One" and "Series Two" to designate intermediate and long-term project phases, respectively, the project timeline currently extends to 2040. This is not an unexpected duration, because if the Secretary of Energy's 2010 Initiative is successful, the United States will have operational nuclear power plants at least through 2070, and advanced reactors beyond that date.

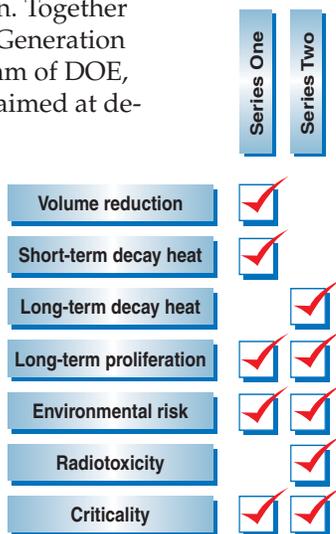Series One and Two activities are essentially executed in parallel, with Series One research
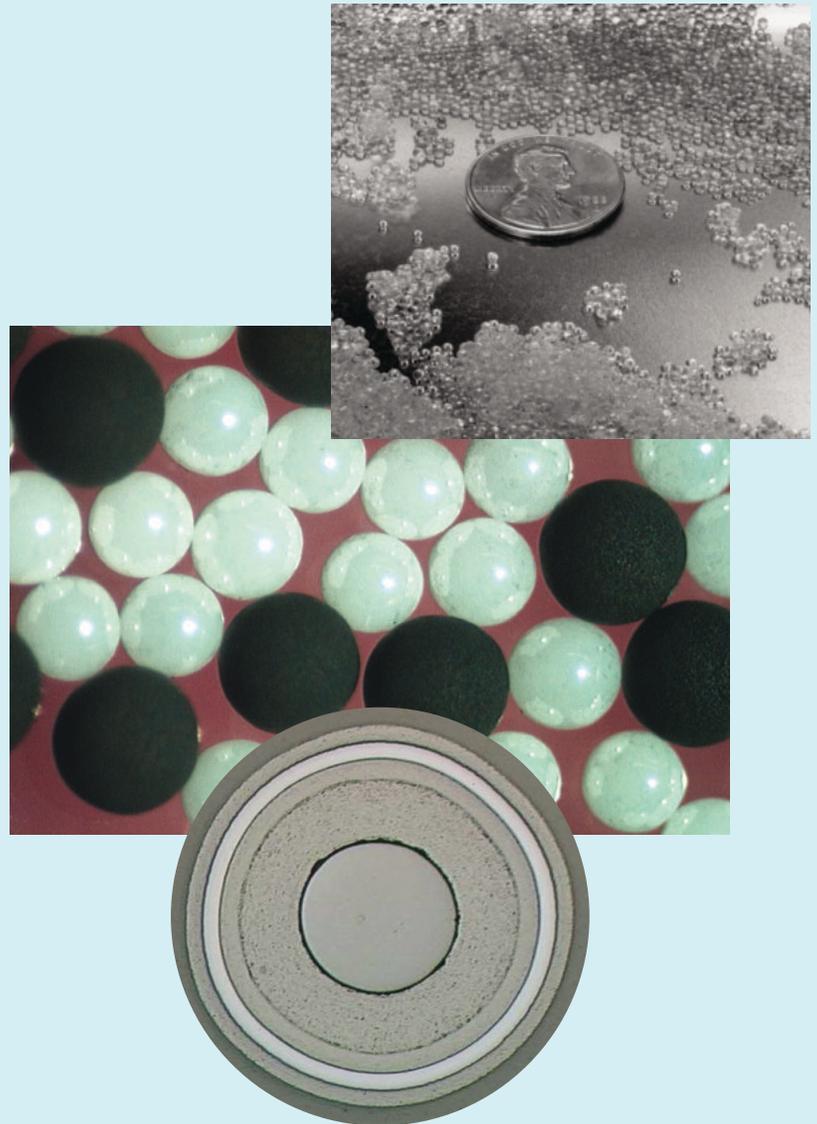
These images show various stages in the preparation of small beads that form the core of tri-isotropic (TRISO) fuels for high-temperature gas-cooled reactors. TRISO fuels are kernels of uranium oxide or plutonium oxide that are coated to prevent the escape of fission products. Hafnium and zirconium dioxide beads are being used as stand-ins for uranium in development tests at Oak Ridge National Laboratory. The hafnium beads are about 500 micrometers in diameter before being dried and heated to produce the final oxide. In the top photo, the small size of the hafnium beads is illustrated next to a penny. The oxide beads are then coated with multiple layers of carbon and a layer of silicon carbide. The middle photo shows both uncoated (white) and coated (black) zirconia beads. In the photo immediately above, the multiple layers of the coating are visible in the microscope image of a coated zirconium oxide bead that has been cut open to examine the thickness and uniformity of the various layers.

focusing on the current generation of reactors and their near-term successors. Series Two research and development is directed toward the development of fuel and chemical processing technologies needed to support a sustainable nuclear energy system in this country.

The potential growth requirements for the U.S. nuclear energy system, needed to curtail greenhouse gas emissions and to provide the means for hydrogen recovery from water, demand a closed fuel cycle for the maximum efficient resource utilization. Together with the Generation IV program of DOE, which is aimed at de-

	Series One	Series Two
Volume reduction	✓	
Short-term decay heat	✓	
Long-term decay heat		✓
Long-term proliferation	✓	✓
Environmental risk	✓	✓
Radiotoxicity		✓
Criticality	✓	✓

Series One and Two Benefit Comparison

veloping advanced reactor technologies, the AFCI program strives to accomplish this turn-around in the production of energy for homes, industry, and transportation.

Deployed in 2015, the Series One spent-fuel treatment facility will process spent commercial reactor fuel to recover plutonium and neptunium for incorporation in proliferation-resistant fuels for burning in current Light-Water Reactors (LWRs) or intermediate-term Generation IV reactors. The minor actinides and certain heat-generating fission products

will also be extracted from the commercial spent fuel and stored, either for eventual disposal (in the case of fission products) or for minor actinide transmutation in fast reactors or in subcritical accelerator-driven systems

The intermediate-term prototype Generation IV reactor is slated for deployment in 2015, and will be focused on efficient hydrogen production and plutonium destruction. The longer-term Generation IV reactor will be deployed after 2030 and will be focused on efficient uranium usage and waste minimization.

The Accelerator Driven System provides an efficient option for destruction of the Series Two plutonium and minor actinides without the creation of additional actinide waste.

The fast-neutron versions of the Generation IV reactors as well as Accelerator Driven Systems will be capable of fissioning the minor actinides (primarily americium and curium) down to low levels, something that is not possible to accomplish efficiently in LWRs.

Burning of these highly radiotoxic elements is crucial to decreasing both the radiotoxicity and long-term decay-heat burden of spent nuclear fuel. Many scientists believe that the demonstration of the feasibility of this transmutation system is the key to transitioning to a nuclear-energy economy.

Series One project activities in the next several years can be viewed as precursors to crucial milestones that occur in 2006. In that year, separations and fuels technologies must be selected, and those selections will directly impact aspects of the design of the spent fuel treatment facility.

In the following year, a major decision point arrives when the government is slated to make a decision on whether or not to commit to deployment of the advanced fuel cycle system. If the decision is positive, it could lead to the initiation of construction of the large spent-fuel

treatment facility in 2010. Based on the opinion of some experts, it appears that the long duration of AFCI may be entirely consonant with other constraints, particularly economic ones.

For example, Ernest Moniz of the Massachusetts Institute of Technology opined in a recent talk here that on economic and nonproliferation grounds, open-fuel-cycle reactors will remain the choice through midcentury if we are to use nuclear power to help in meeting the increased global demand for electricity over the next 50 years. (See story on Page 12.)

"Open fuel cycles will dominate for a long time," predicts Moniz, citing economic and proliferation constraints while at the same time admitting that "Yucca Mountain isn't the answer for growth scenarios," and that tapping the "unlimited uranium resource" in seawater would be "expensive."

Moniz claims that the spent fuel from the open fuel cycle could be stored for 50 to 100 years before disposal in a repository. This would allow time for development and evaluation of advanced fuel cycles for which this spent fuel becomes a resource.

Moniz does raise the interesting question of whether "reducing that long actinide tail . . . matters"—i.e., whether there has been enough discussion on the advantage of transmuting the long-radiotoxicity actinides in spent fuel. For those researchers intimately involved in the rigorous and demanding activities of AFCI, the answer would appear to be a resounding "yes."

—Vin LoPresti
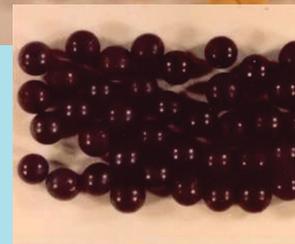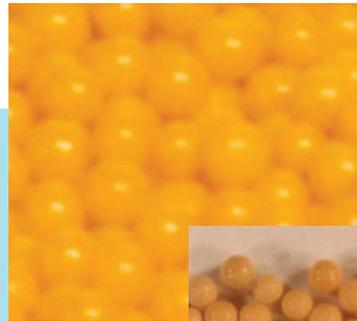
Uranium dixoide beads shrink in diameter during drying and sintering. These images, from top to bottom, show 960-micron wet beads, 610-micron dry beads, and 300-micron sintered beads. After sintering, layers of carbon and silicon carbide are added to the uranium dioxide beads. Many of the beads containing the enriched uranium fuel are packed together into spherical or cylindrical fuel elements to be used in the core of high-temperature gas-cooled reactors.

ERNEST MONIZ

Suppliers must overcome safety, economic, nonproliferation, and waste concerns

Soaring energy demand requires 400-percent growth in nuclear power over the next 50 years

P hysicist Ernest Moniz, a former undersecretary of the DOE, sees nuclear power production as having great potential for meeting the soaring world demand for electricity while preserving an acceptably clean environment. Environmentally, nuclear power has one great advantage: It does not produce carbon emissions.
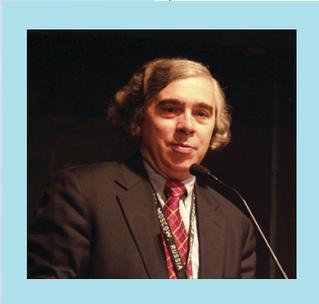
About one-sixth of the electrical power in the world today is generated in nuclear power plants—a fraction comparable to the amount of hydropower. Most of the rest is generated by burning fossil fuels: coal, oil, and gas. In the United States, only about one-fifth of electricity is nuclear-generated—but that 20 percent takes the place of fossil-fuel energy production that would mean another 175 metric tons of carbon equivalent per year streaming into the atmosphere.

Nevertheless, despite the environmental advantages of nuclear power, Moniz said in a recent talk at Los Alamos, "If nuclear power does not grow by roughly a factor of 4" in the next 50 years, then nuclear energy will turn out to be "too much pain for very little gain." Furthermore, to achieve that 400-percent growth, nuclear power suppliers must find ways to overcome potentially show-stopping concerns related to safety, economics, nonproliferation, and waste.

Moniz, a Massachusetts Institute of Technology (MIT) professor, presented a Director's Colloquium on nuclear power Jan. 14. He is participating in an 18-month study funded by MIT and the Alfred P. Sloan Foundation that will release a report this year on the near-term, mid-term, and long-term prospects for nuclear energy.

The study group includes several MIT faculty members (in fields spanning science, nuclear engineering, economics, and political science) and a faculty member from Harvard. An external advisory group includes members with backgrounds as diverse as venture capital and politics. Two members of the group are former chiefs of staff to the president. The group will develop recommendations on nuclear power research and development, nonproliferation, and relevant collaborations involving the United States and Russia.

For the past year, Moniz has also been spending about a quarter of his time on a sabbatical at Los Alamos, working with collaborators here that are researching aspects of advanced nuclear energy systems (see story on Page 15).

Ernest Moniz
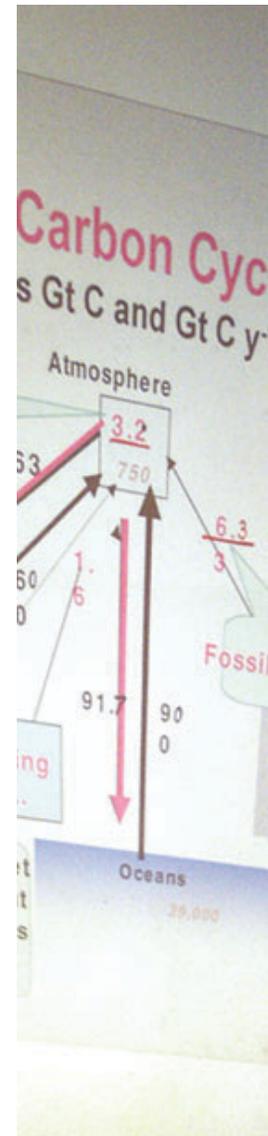
Moniz noted in his talk that the DOE's Energy Information Administration projects a global increase of 2.7 percent per year (compounded) in electricity usage. Some developing countries are experiencing a huge growth in the demand for electricity. China, for example, had an "enormous rate of growth" in electricity use last year—10.5 percent.

The United Nations Human Development Index, a measure of human well being, says countries will need the capacity to generate about 4,000 kilowatt hours (kwh) of electrical power per person per year to provide the "good life" for their citizens in 2050.

Among the 25 highest-population countries in the world, only four are likely to have 4,000 kwh per person per year by midcentury, Moniz said. Developed, affluent countries such as the United States have achieved or surpassed the goal.

In China, because of the relatively stable population, electricity supply growth of only 3.2 percent per year will make it possible to reach the benchmark. Countries like India could get close if they exercised exceptionally good management. Many African nations "don't have a prayer" without major international assistance.

But there's another consideration: the issue of climate change. Today, roughly 6 gigatons of carbon pour into Earth's atmosphere each year from fossil-fuel use. Half remains in the atmosphere, adding to the existing 750-gigaton carbon load in the atmosphere.

The bottom line is, Moniz said, that in 50 years, carbon emissions must be at today's level or lower for the safety of the planet. "We can't really be emitting much more carbon than we are today" despite the greatly increased demand for electricity, he said. Unless the world is willing to spend a lot more money on carbon research, he said, it is obvious that we must consider nuclear energy as an option.

He outlined a "2050 scenario" that would provide the necessary factor-of-4 increase to make nuclear energy viable. It would require use of nuclear energy to produce approximately 1,000 gigawatts electric (GWE) in the United States, Europe, and developed East Asia; approximately 100 GWE in the Former Soviet Union; and approximately 400 GWE in the developing world. The United States alone would need to produce 400 to 500 GWE through the use of nuclear reactors.

The world couldn't look to Japan for a major increase in nuclear energy production because its population is expected to drop by 18 percent in the next 50 years, and its energy demands will be limited accordingly. France already generates the bulk of its electricity with nuclear power plants and has a stable population.

Moniz outlined three possible time periods on the "technical pathway" to nuclear power success. The "incremental period"—which he also called the "survival period"—would run from the present to the year 2015 and would be an "enabling period" of little change. The "growth period"—from 2015 to midcentury—would see four to five times more deployment of nuclear power.

There would be "environmental drivers" in the industrialized world. Economic considerations would be important. The general fuel-cycle architecture and the deployed infrastructure would be similar to what we see today, but some new techniques would be possible. He mentioned high-temperature gas-cooled reactors (HTGRs), high-burnup fuels, the use of boreholes for disposal of waste, and

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hydrogen production for transportation fuel as possible areas where applied research could yield results in the next 10 to 20 years.

In general, however, there would be little change in technology. The bloom of new technology would come at midcentury, when new approaches in architecture and infrastructure would mature and closed fuel cycles could become important.

Commenting on details on the technical pathway, Moniz said he believes that the open fuel cycles of the present—an approach in which nuclear fuel goes through the reactor just once and then is disposed of directly in a repository after a suitable period of "cooling" (10 to 50 years)—will dominate the field for the next 50 years.

The rise of closed-fuel-cycle systems—with facilities that use special techniques to separate and reuse radioactive materials—won't come until the second half of the century. However, he noted the critical Advanced Fuel Cycle Initiative work done by people at Los Alamos that is contributing to developing this new technology (see story on Page 7).

For nuclear power advocates to succeed in achieving a major gain in its use, Moniz indicated, they must deal successfully with several major issues. Nuclear power researchers must find ways to cut costs, reducing capital costs by at least 25 percent and promoting government incentives if they are to have hope that reactor-produced power can compete. They must overcome the public concern about nuclear power plants—a concern that was greatly increased by the Three Mile Island and Chernobyl accidents.

They also must strengthen the international norms and security safeguards regarding radioactive materials—especially separated plutonium—to reduce proliferation concerns, and minimize transportation risks. And while advocates need to enable the robust development of open-fuel-cycle technology for several decades, they also need to nurture research and development on closed fuel cycles.

Researchers must explore new ideas in waste disposal. One of Moniz' favorite ideas is disposal through boreholes dug deep into stable crystalline rocks. He projected a map done by Grant Heiken of Environmental Geology and Risk Analysis (EES-9) that indicated that most places likely to have expanding nuclear power production would have access to such crystalline formations.

Researchers also must collaborate with the Russians who are now "quite prepared," Moniz said, to work with the United States to stop further plutonium proliferation, minimize waste, achieve safety, and evaluate storage options including geological isolation.

And, finally, they must re-evaluate the Atoms for Peace paradigm and find new approaches to nuclear leadership. Moniz mentioned the possibility of using U.S.-Russia collaboration as a bilateral "seed vehicle," perhaps within the framework of the "Group of Eight" (G8) nations, an informal organization of eight countries (including the United States) that meet each year to discuss broad economic and foreign policies.

—Charmian Schaller

Los Alamos collaborates with MIT

Researchers assess nuclear fuel recycling options for an advanced fuel cycle

A Massachusetts Institute of Technology (MIT) study on the viability of nuclear power is expected to produce final recommendations in August. In the meantime, it has spun off a Los Alamos study on a key issue: assessment of nuclear fuel reprocessing options for an advanced fuel cycle.

Scientists at Los Alamos began work last June assessing the performance of current and proposed nuclear materials separation technologies. Once their work is completed, they hope to have methodologies defined that will compare nuclear-fuel-reprocessing options to

Once-Through

Uranium mining, milling, conversion

Enrichment fuel production

Reactor fuel utilization

Used fuel storage

Disposal

quantify them in terms of environmental performance, proliferation resistance, economic considerations, and safety criteria.

Ed Arthur of the Strategic Research Directorate (ADSR) is the principal investigator on the project, and nine other researchers are participating—Scott DeMuth, Richard Farman, Drew Kornreich, Andrew Koehler, and Faris Badwan, all of Decision Applications (D) Division; Web Keogh of the Chemistry Division Office (C-DO); Gordon Jarvinen of the Nuclear Materials Technology Division Office (NMT-DO); Kory Budlong-Sylvester of Safeguards Systems (NIS-7); and John Miller of Nuclear Criticality Safety (HSR-6).

For the last year, Ernest Moniz, cochairman of the MIT nuclear power study, has spent part of his time on a sabbatical at Los Alamos. He and Arthur's team members have discussed many aspects of nuclear power production extensively.

Arthur said Moniz believes that 50 years from now, there could be a major need for the increased production of nuclear power (see story on Page 12). Moniz also believes, Arthur said, that this increase in nuclear-energy production must be large to have significant impact on future needs and problems. Otherwise, in Moniz's view, it is not worth continuing development of nuclear power because of the significant challenges associated with the effort.

Arthur added, however, that in five decades the world probably will also still be struggling to maintain an acceptable atmosphere by minimizing carbon-dioxide emissions. "The only developed (energy) technology that can meet both of those requirements is nuclear," Arthur said. "The picture at midcentury could be *lots* of nuclear power."

At present, the nuclear power plants in the United States use an open fuel cycle. In such a cycle, uranium is mined, milled, and converted; enrichment is carried out; the resulting reactor fuel is used just once; the used fuel is stored; and, finally, the used fuel is disposed of as waste in a geologic repository.

As an alternative, a closed fuel cycle could exist in which nuclear power plants continue to breed new fuel material (plutonium and smaller amounts of other transuranic elements); cooling of discharged fuel occurs; material recycling allows most of the resulting

material to be fed back into the reactor and used again; and a greatly reduced amount of material is ultimately disposed of as waste.

Between these two extremes lies the concept of a "modified once-through (recycle, actinide-burning)" power plant in which uranium is enriched to produce fuel that is used in the reactor; the used fuel is stored and then reprocessed to recover transuranic actinides (plutonium, neptunium, americium, and curium); these actinides are then recycled into special burner reactors; and

Modified Once-Through

Uranium mining, milling, conversion

Enrichment fuel production

Reactor fuel utilization

Used fuel storage

Reprocessing fuel production

Disposal

the small amount of material remaining is disposed of as waste.

Clearly, reprocessing is an essential component in both the "modified once-through" and "closed-fuel-cycle" approaches. It is therefore logical to assess proposed reprocessing approaches on the basis of economics, proliferation resistance, safeguardability, safety, and waste criteria.

Los Alamos scientists discussed reprocessing issues with Moniz and with John Deutch, an MIT chemistry professor, and became familiar with Deutch's informal paper, "MIT Nuclear Energy Study Group, Notes on Reprocessing."

The paper noted that dozens of separation schemes have been proposed over the years, but the most widely used reprocessing technology has been plutonium-uranium extraction (PUREX) process, which relies on liquid-liquid extraction separations.

In PUREX, uranium and plutonium are extracted from a liquid phase of aqueous nitric acid into an immiscible liquid phase containing a kerosene-like solvent and tributyl phosphate. The fission products generally remain in the aqueous stream. The plutonium and uranium are purified in subsequent steps and transformed into uranium oxide and plutonium oxide products. The process uses extensive quantities of flammable, organic solvent.

Recently, Argonne National Laboratory has advanced a technique called pyroprocessing that relies on electrorefining for the crucial actinide separations.

Under a DOE program, Argonne demonstrated pyroprocessing by treating about 25 tons of experimental breeder reactor II fuel produced in Idaho between 1963 and 1994. This fuel is 63 percent uranium-235 at burnup. The Argonne fast-reactor project proposes to use pyroprocessing as the central separation method for treating high-burnup fuel produced from fast reactors operated in an integrated-fuel-cycle operation.

How does an objective scientist tell which process—PUREX or pyroprocessing—best meets society's concerns about safety, cost, proliferation resistance, and waste production?
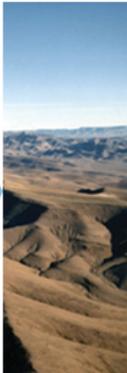
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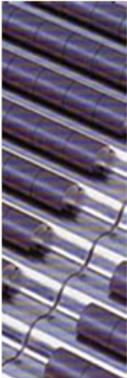
Reactor material breeding
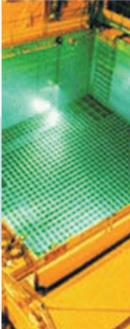
Cooling

Reprocessing fuel production

Disposal

"In order to reach an informed conclusion," the MIT paper says, "there should be a documented model that permits the systematic, quantitative comparison of alternatives from the viewpoint of these four objectives….The proposed evaluation model should be sufficiently flexible to deal with a range of spent fuel scenarios…." It is that model that Los Alamos scientists are trying to develop, using a comparison of PUREX and pyroprocessing to test their methodology.

Although the study is in its early stages, a conceptual methodology is developing, and Arthur said the Los Alamos researchers have reached some preliminary conclusions in their first comparison:

■ The total cost of a reprocessing system is primarily the result of front-end (fuel handling) and back-end (waste management) costs. A preliminary assessment of PUREX has been completed, and it shows, based on actual plant data, that separations operations account for less than 20 percent of total plant cost.

■ There is a large disparity between the real costs of PUREX and the estimated costs for pyroprocessing. For example, pyroprocessing-plant capital costs have been estimated (on paper) to be 10 times less than for a PUREX plant. However, there are no large-scale, pyroprocessing-based reprocessing plant data upon which to draw.

■ The two processes appear comparable on total waste generation.

■ In terms of safety, a preliminary assessment of millirem levels of radioactivity at the site boundaries indicates that PUREX could offer some advantage over pyroprocessing under certain circumstances.

The issues of proliferation resistance and safeguardability are difficult to evaluate and will require much more work. Arthur said more detailed modeling of facilities and operations must be done to produce credible and meaningful comparisons on these issues—and they are vital.

—Charmian Schaller

Code Validation

Benchmark experiments in the VENUS critical facility
Advances in code validation for mixed-oxide fuel use in light-water reactors

A typical commercial thermal reactor produces around 250 kilograms of plutonium per giga watt-year electric output. The majority of the spent fuel discharged from the world's 430 or so commercial nuclear reactors has been designated for interim storage and eventual direct disposal. Those utilities that have opted for the so-called "once-through"—or open—fuel cycle have chosen this option after considering the particular circumstances (political, economic, strategic, logistic, historic, etc.) that apply locally. With different local circumstances, other utilities have chosen to reprocess their spent fuel.

Current commercial reprocessing plants are all designed to separate the plutonium remaining at discharge for reuse. Historically, the rationale was to be able to recover sufficient plutonium to enable a buildup of fast reactors, which were expected to be deployed as uranium reserves became scarce and prices rose. For a variety of reasons, but principally the low price of uranium ore, fast reactors have not yet been deployed commercially and projected timescales for doing so have been put back everywhere.

There are several technical options for plutonium management involving reuse in existing light-water reactors (LWRs). Partial core loading of mixed-oxide (MOX) fuel assemblies in pressurized water reactors (PWRs) is already well established on a commercial basis, with thirty-seven reactors in Europe (two in Belgium, 22 in France, 10 in Germany, and three in Switzerland) currently operating with part-MOX loading. There is less experience in MOX usage in boiling-water reactors (BWRs), with just two BWRs in Germany currently using MOX fuel.

Partial MOX loading in a PWR involves the substitution of a fraction of the uranium dioxide (UO_2) assemblies with MOX assemblies of the same mechanical design. This avoids issues of thermal-hydraulic and mechanical-handling incompatibility. The plutonium concentration in the MOX assemblies is usually adjusted so that the reactivity lifetime of the MOX fuel coincides with that of the UO_2 fuel, in which case the average discharge burnups of the two fuel types will be the same.

The presence of MOX fuel batches in a LWR affects the nuclear design characteristics of the core in a complicated fashion. Consequently, a need was seen in the late 1980s to develop or to improve the in-core management codes and to benchmark and validate them against experimental data. An insufficient validation might induce a dramatic increase of the uncertainty factor with a possible reduction of the reactor power required to keep reactor operations within very stringent safety margins.

Pierre D'hondt

Based on experience accumulated during 25 years of collaboration, the Belgian Nuclear Research Center (SCK•CEN), together with Belgonucléaire, implemented a series of benchmark experiments in the VENUS critical facility at SCK•CEN in Mol, Belgium. The experiments were designed to provide organizations concerned with MOX fuel the ability to calibrate and improve their neutronic calculation tools.

The VENUS critical facility

The VENUS critical facility is a water-moderated zero-power reactor. (VENUS is short for Vulcain Experimental NUclear Study; Vulcain was a projected marine reactor.)

VENUS consists of an open (nonpressurized) stainless-steel cylindrical vessel and a set of grids that maintain fuel rods in a vertical position. After a fuel configuration has been loaded, criticality is reached by raising the water level within the vessel. Because the neutron flux is very low, no water circulation is needed to keep the fuel rods at low (room) temperature. The reactor shutdown is induced by emptying the vessel.

A view of the VENUS critical facility showing the top of the nonpressurized vessel and the grid that holds the fuel rods in a vertical position.

VENUS capabilities

The VENUS critical facility is characterized by a high experimental flexibility:
- direct access and manual handling of individual pins,
- easy loading of the reactor,
- special (dismountable) pins and removable grid-parts,
- easy water density simulation (down to 50 percent of density at room temperature),
- reactor control through water level variation or absorber rod deployment, and
- boron poisoning (up to 2,000 parts per million).

The following parameters can be measured with high accuracy in VENUS:
- critical water level,
- reactivity coefficient of the water level,
- axial fission rate distribution,
- horizontal fission rate distribution,
- spectrum indices,
- fission rate distribution inside a fuel rod,
- detector response, and
- delayed neutron fraction.

Experimental programs in VENUS

Because of its experimental flexibility, a series of experiments related to plutonium use in LWRs has been run in VENUS since 1990.

An early experiment investigated plutonium recycling in LWRs. The program, called VIP (VENUS International Programme), used fuel with high plutonium and gadolinium content. The aim of VIP was to validate reactor codes with respect to MOX fuel for both PWRs and BWRs. It focused on the criticality and fission-rate distribution calculation.

VIP was divided into two stages: VIP-BWR and VIP-PWR. VIP-BWR considered three mockups: an all-UO_2 8x8 subassembly, an all-MOX 8x8 subassembly, and an island MOX 8x8 subassembly. VIP-PWR considered two mockups: an all-MOX 17x17 subassembly and a similar assembly containing burnable gadolinium absorber rods. This program showed the validity of reactor codes such as DORT, TORT, GOG, TWOTRAN, and LWRWIMS for calculating criticality and fission-rate distributions in present-day fuel assemblies.

To cope with future developments in the nuclear fuel cycle and the tendency of going to higher burnups, there was a need to investigate a possible positive void coefficient at high plutonium enrichments.

Therefore, an international program called VIPO (Void coefficient measurement In Plutonium mixed Oxide lattice) was established to determine the influence of a void bubble in a LWR reactor using high plutonium enrichment (i.e. from 10 percent to 15 percent) and the validation of related computer codes.

A special experimental device, the so-called void box, was developed and constructed to simulate a void in the reactor core.

The VIPO program was successful in providing accurate benchmarks for validating reactor codes. It was shown that nowadays reactor codes can calculate fission-rate distributions with and without voids with an accuracy better than 2.5 percent.

After VIPO came VIPEX-PWR. This program, which was an extension of VIP-PWR, was aimed at determining core physics parameters of MOX assemblies that are of interest for reactor operations, such as the delayed neutron fraction (β_{eff}), the americium-241 effect, the moderator density effect, the control rod worth, and flux tilt within MOX rods.

The fraction of delayed neutrons is very important in reactor control; for plutonium-239 it is about three times as small as for uranium-235. So in principle, plutonium becomes prompt critical much faster than uranium. Although in a mixed plutonium-uranium configuration this effect is less pronounced, it is still important to have good quantification of the β_{eff}.

The aging of plutonium by the decay of plutonium-241 to americium-241 has a significant influence on the reactivity of a MOX assembly. By reloading pin by pin the same configuration used in VIP, the americium-241 effect was measurable after four years.
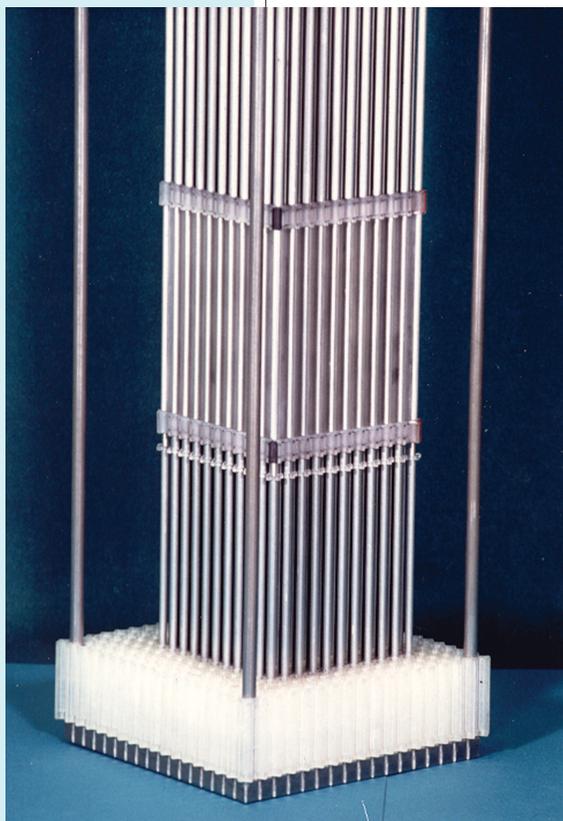
This experimental device, called a void box, simulates a void in a reactor core. This void box is traversed by highly enriched plutonium fuel.

The present-day pitch—the distance between the fuel rods—of LWR fuel assemblies (12.6 millimeters) has been optimized for the use of uranium. This pitch results in a very undermoderated configuration for MOX fuel.

Investigation of moderator density effects on the reactivity can validate reactor codes for searches for the optimized pitch for MOX and for differences in hot and cold reactor conditions.

In VENUS, water-density reduction is simulated by inserting aluminum rods between fuel pins. To measure the effect of moderator density, the rods are pulled out from the configuration in several steps.

Because the neutron spectrum in a MOX assembly is harder—has higher energy—than in a UO_2 assembly and the neutron-absorbing capacities of control rods are mainly based on the absorption of lower-energy thermal neutrons, the effectiveness of control rods—the control-rod worth—is less in a MOX fuel assembly. The control-rod worth can easily be measured in VENUS by inserting stepwise control rods at several positions and by determining the resulting difference in critical water level.

The reason for measuring the flux tilt is that the peripheral rods of the MOX assembly are subject to a large shift of the neutron spectrum because the neutrons from the UO_2 assembly have a lower average energy than those from the MOX assembly.

The fission cross section for low energy is higher, resulting in a higher reaction rate for these neutrons. This is partially compensated by the higher neutron flux in the MOX assembly, but large power differences inside the fuel rod are still expected. Such flux tilt in VENUS is measured by inserting activation foils in between fuel pellets in a demountable rod or by direct measurements on the fuel pellet.

After VIPEX-PWR, we investigated recent BWR configurations in the BWR-NBN program, which was very similar to VIP-BWR, but for 9x9 BWR configuration.

We also investigated the use of weapons-grade plutonium in LWRs. This was possible through the availability of plutonium fuel with plutonium vectors close to weapons-grade plutonium. This program, called IMP (Investigation of Military Plutonium), was an internal SCK•CEN program.

Recently, a new international program—REBUS—was implemented in VENUS. The program aims to establish an experimental benchmark for validation of reactor physics

Some useful definitions

Delayed neutron fraction (β_{eff}): delayed neutrons appear in a fission event after an amount of time, as opposed to prompt neutrons, which appear almost immediately. While the fraction of delayed neutrons is small (less than 1 percent), accurate prediction of the effective delayed neutron fraction (the beta effect—or β_{eff}) is vital for controlling the fission chain reaction in a nuclear reactor.

Americium-241 effect: The decay of plutonium-241 into americium-241 over time (plutonium-241 has a half-life of approximately 14.1 years).

Moderator density effect: The change in reactivity of the system when the density decreases from an increase in temperature or when the density increases from a decrease in temperature.

Control-rod worth: The effect on reactivity of inserting the control rods into the system.
Flux tilt: A comparison of the flux in a MOX-fueled assembly (in particular, rods on the periphery vs.rods in the center) to that in surrounding uranium dioxide-fueled assemblies.

View of the REBUS container that will contain the spent fuel bundle.

codes for the calculation of the loss of reactivity due to burnup for PWR fuel, both for UO_2 and MOX fuel bundles.

The rationale for REBUS lies in the fact that present criticality safety calculations of irradiated fuel often have to model the fuel as fresh fuel because no precise experimental confirmation exists of the decrease of reactivity due to accumulated fission products.

In other situations only actinide depletion is allowed to be taken into account and the influence of fission products has to be disregarded.

The fact that this so-called "burnup credit" cannot (completely) be taken into account has serious economic implications for the transport, storage, and reprocessing of irradiated fuel. For long-term geological storage it is almost imperative to apply burnup credit.

In VENUS, the measurement of this burnup credit is based on comparative reactivity measurements between a fresh fuel bundle and an identical (initial composition) irradiated fuel bundle. Loading irradiated fuel bundles in a critical facility is rather new and for VENUS the necessary infrastructure for loading spent fuel in the REBUS container had to be installed.

Currently, an experimental program for three fresh fuel configurations has been executed and a dummy test for fine-tuning the loading procedure of irradiated fuel bundles is under way. The experimental program for the spent MOX fuel and for the spent UO_2 bundles will be executed this spring.

Conclusions

The experiments carried out in the VENUS critical facility have demonstrated that VENUS is a very flexible and easy-to-use tool for the investigation of neutronic data as well as for the study of licensing, safety, and operation aspects.

Such data allow validation of the reactor physics codes for MOX use in LWRs. This validation has made it possible to safely operate thirty-seven PWRs and two BWRs with partial MOX-core loadings.

While MOX currently is not being used in a VVER—the Russian version of a PWR—a program to use weapons-grade plutonium in VVERs is under way.

Based on the knowledge and experience resulting from VENUS' years of successful experimentation, researchers believe that the future use of MOX in a VVER is a viable option posing low technical risk.

Status of research and development
after the IMF8 Workshop

Inert matrix fuel has the potential to produce electricity while burning up more plutonium

Claude A. Degueldre, a scientific project leader at the Paul Scherrer Institute (PSI) in Switzerland, briefed a Los Alamos National Laboratory audience March 20 on the status of research and development on inert matrix fuel (IMF)—an approach that has the potential to greatly reduce the amount of reactor-produced plutonium that should be placed in geological disposal.

Degueldre's host at the Seaborg Institute Seminar was David Clark, institute director, who noted that Degueldre, a Doctor ès Sciences from the University of Liege, Belgium, and currently a professor at the University of Geneva, Switzerland, also gave a Seaborg Institute lecture two years ago.

Degueldre, who has expertise in the analytical chemistry of actinides as well as IMF, said that Switzerland has five light-water reactors. As a result, its stockpile of plutonium—a product of light-water reactors fueled by uranium—is increasing by approximately 750 kilograms per year. The Swiss population, he said, would like to get rid of this plutonium. "My work for the last 10 years was to find a scientific solution to that problem," he added.

Countries including Japan and France are facing the same problem—one that is also important in the United States, a nation with much more "open space," where the current plan is to bury long-lived, high-level waste in Yucca Mountain near Las Vegas, Nev., and in similar future geologic depositories where it will have to remain for more than 10,000 years. Plutonium, a radiological hazard, has a half-life of 24,000 years, and the world's nuclear reactors are now producing about 70,000 kilograms of it per year.

Research indicates that IMF might provide some valuable answers to the problem, making it possible to produce electricity while burning up more plutonium and minor actinides in a way that is proliferation-resistant, economical, and ecological, while also being safe and sustainable.

Since 1995, Degueldre has been the primary organizer of a series of world conferences on IMF. The most recent, "IMF8," was held in Japan. Degueldre said in his Los Alamos presentation that the number of scientists working on IMF is growing. He said these scientists, from many different countries and various research fields, have become "a group of friends working together," evaluating a variety of strategies, exchanging samples and data, and comparing results at each annual meeting.

Claude Degueldre

This article was contributed by Claude Degueldre of the Paul Scherrer Institute, David L. Clark of the Seaborg Institute, and Charmian Schaller of Communication Arts and Services (IM-1).

The process of producing plutonium isotopes in a nuclear reactor is shown here. Fertile ^{238}U captures a neutron to produce ^{239}U. The ^{239}U is unstable with a half-life of 23.5 minutes and decays via loss of a beta particle to produce ^{239}Np, which is also unstable toward beta decay, and generates ^{239}Pu. ^{239}Pu can either under fission to produce more energy, or it can capture neutrons to successively produce ^{240}Pu, ^{241}Pu, etc.
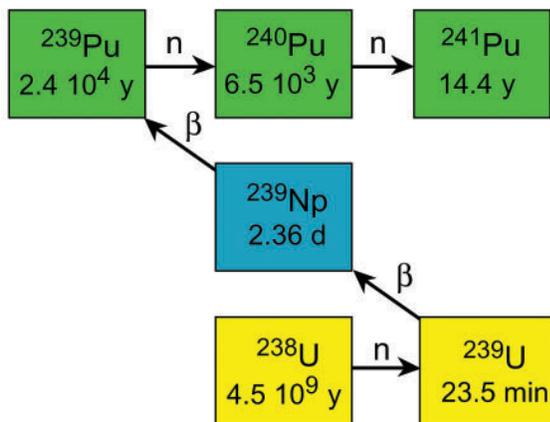
In a recent paper—"Inert Matrix Fuel Strategies in the Nuclear Fuel Cycle: the Status of the Initiative Efforts at the 8th Inert Matrix Fuel Workshop"—Degueldre and a coauthor, T. Yamashita of the Japan Atomic Energy Research Institute (JAERI) in Tokai, Japan, explained the motivation for their studies and summarized the work in progress.

"The 'raison d'être' of the Initiative for Inert Matrix Fuel," the authors said, "is to contribute to research and development studies on inert matrix fuels that could be used to utilize, reduce, and dispose both weapon- and light-water-reactor-grade plutonium excesses."

They said IMF, a promising once-through strategy, could be used in the existing commercial light-water reactors in Europe, Japan,

Russia, and the United States; in the Canadian pressurized heavy-water reactors; or in future transmutation units where actinides could be bombarded by neutrons and converted to different nuclides with shorter half lives.

"This option has the advantage of reducing the plutonium amounts and potentially minor actinide contents prior to geological disposal," Degueldre and Yamashita said in their paper.

On the other hand, IMF could be used in a "multirecycling strategy"—in other words, the actinides encapsulated in inert matrices could be run through a reactor or a transmutation device more than once; each time through, more would be consumed. After a last cycle, the final spent IMF would be disposed of in a geologic repository.

Plutonium production in UOX vs. plutonium consumption in IMF

In conventional nuclear reactor fuel, fissile ^{235}U is enriched to approximately 3 percent in a matrix of ^{238}U, usually in the form of a UO_2 (or UOX) fuel pellet.

While not directly fissionable, the ^{238}U is fertile in the sense that it can capture a neutron to generate ^{239}Pu, which is fissile, and the ^{239}Pu can capture neutrons to generate ^{240}Pu, ^{241}Pu, again fissile, etc. In this way, fertile ^{238}U is transmuted into isotopes of plutonium (and other actinides) in the reactor neutron spectrum.

During the lifetime of the fuel in the reactor, some of this plutonium is also burned up in fission reactions, but a significant amount remains in the spent nuclear fuel, generating concern about the potential proliferation of plutonium.

Spent fuel can be reprocessed to separate out the plutonium for burning in a reactor in the form of a mixed-oxide (MOX) fuel. While the process of burning separated plutonium in the form of MOX has a long history, the pace

of plutonium removal is slow or unachievable because while MOX burns some plutonium, the neutron irradiation of the fertile $^{238}UO_2$ matrix continues to produce more plutonium.

Ideally, one would like to replace the fertile $^{238}UO_2$ matrix with a nonfertile, "inert matrix" to avoid the production of plutonium in nuclear reactors and to efficiently achieve its consumption. This is the concept of an inert matrix fuel.

IMF composition and loading

Degueldre described how the material selection for the inert matrix is guided by the neutronic properties of the elements and/or isotopes based on their transparency for neutrons in the reactor—an essential requirement for the term "inert matrix."

The desired thermodynamic properties of the materials include high melting point (approximately 3,000 Kelvin), good thermal conductivity, chemical compatibility with cladding, low chemical solubility in hot water, and high density.

Detailed studies around the world have produced a number of candidate materials that include stabilized ceramics such as $Ca_xZr_{1-x}O_{2-x}$, $Y_yZr_{1-y}O_{2-y/2}$, or other ceramics such as $ZrSiO_4$, $Y_3Al_5O_{12}$ (yttrium aluminum garnet), MgO or $MgAl_2O_4$ (spinel), or even nitrides or carbides. Degueldre has been a long-standing proponent of the use of yttria-stabilized cubic zirconia for IMF.

In some cases, a burnable poison (such as erbium, gadolinium, holmium, or boron) or a small amount of fertile additive (such as thorium or uranium) is introduced to improve the neutronic characteristics of the fuel—i.e. by maintaining the reactivity constant over the in-pile irradiation time. Other additives may also be required to stabilize the inert material in the presence of plutonium.
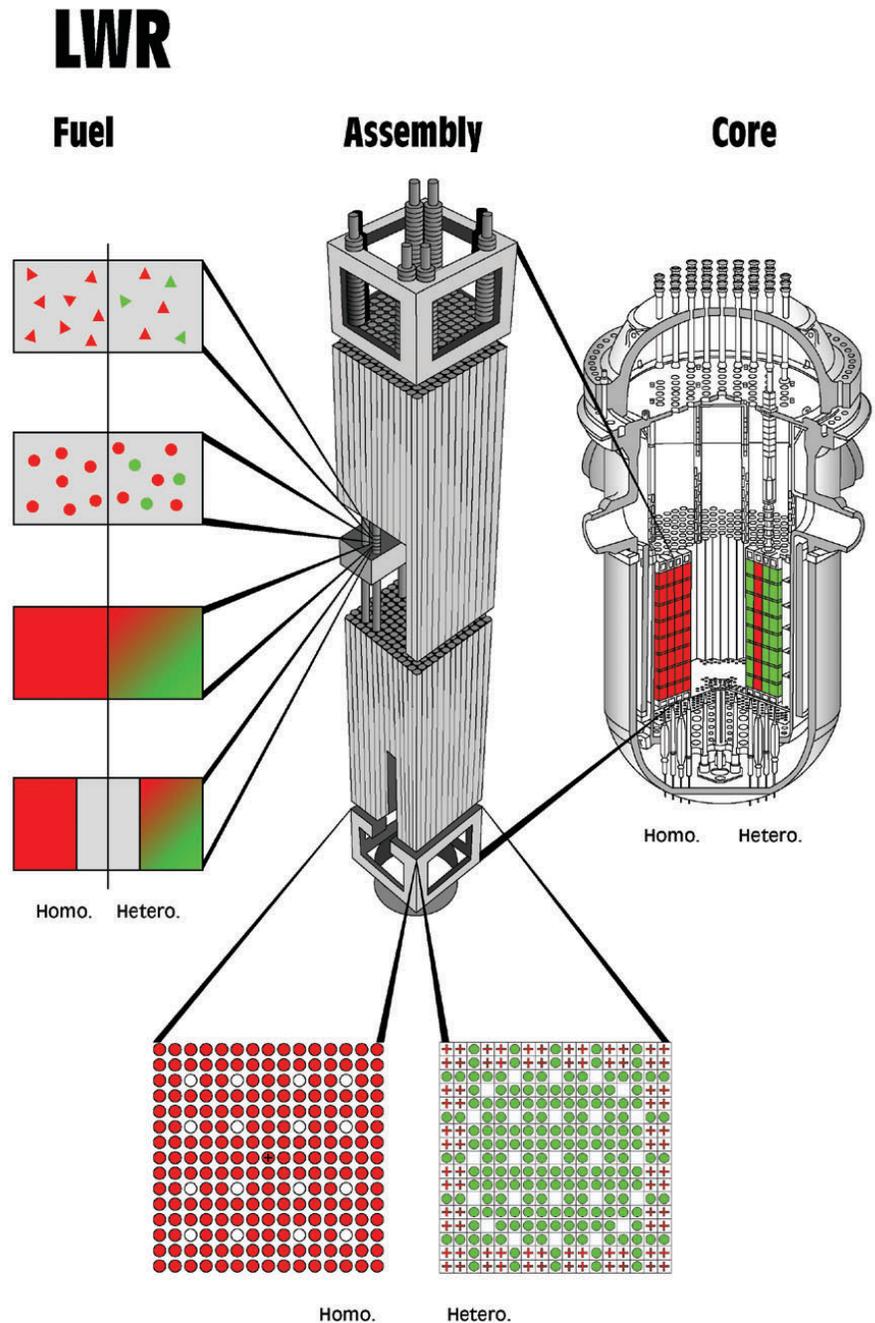
This diagram shows the three levels for IMF use in light-water reactors considering homogeneous vs. heterogeneous system or loading concept at the fuel, assembly, and core levels. The fuel is either a solid solution ceramic homogeneously doped with plutonium (red) or heterogeneously doped with some uranium (green), or is a composite material with particulates or microspheres (again plutonium-doped red, uranium-doped green) imbedded in inert matrix material. The fuel assemblies themselves may be homogeneous (all fuel rods in a given assembly contain IMF, red) or heterogeneous (red IMF rods distributed among green UO_2 fuel—e.g. the French Advanced Plutonium Assembly (APA) concept). The reactor core may also be loaded homogeneously (with red IMF assemblies), or the UO_2 core may be partially loaded with some IMF assemblies forming a heterogeneous core loading.

IMF pellet fabrication, modeling, and in-pile irradiation tests and studies have been conducted around the world in Canadian, Dutch, French, Japanese, Organization for Economic Cooperation and Development (OECD), Russian, and U.S. research reactors.

The IMF can be loaded into a reactor in a number of ways. At the fuel-pellet level, the fuel can be either homogeneous (100 percent IMF) or heterogeneous (IMF doped with some UO_2). The fuel assemblies themselves may be homogeneous—i.e., all fuel rods in a given assembly contain IMF—or heterogeneous, with the IMF rods distributed among the UO_2 fuel rods.

The reactor core may also be loaded homogeneously with IMF assemblies, or the UO_2 core may be partially loaded with some IMF assemblies forming a heterogeneous core loading as it is in practice for MOX.

The introduction of the IMF rods into a UO_2 fuel assembly is quite complicated because of the large differences in the neutron spectra of the cell types and their interaction with one another. Many detailed modeling and reactor irradiation studies have been carried out to examine the optimum arrangement of IMF fuel assemblies relative to the UO_2 assemblies in the core.

From such studies it seems clear that additional research on IMF will continue to make a major contribution to the development of nuclear power as a safe and reliable source of energy. The research on IMF has great relevance to the broader area of nonproliferation and nuclear disarmament because it could mean the burning of excess plutonium from power plants.

In the future, standard UO_2 fuel assemblies could be placed together with plutonium IMF assemblies in a reactor such that no net plutonium would be generated. Fuel rods could be configured such that the amount of plutonium generated from the fertile [238]U in the standard fuel is equal to the amount burned in the inert matrix.

Specific studies presented at IMF8 and completed in early 2003

With the study, "Thermal conductivity of zirconia based inert matrix fuel: Use and abuse of the formal models for testing new experimental data," Degueldre not only presented and recommended high-quality data, but also emphasized the purpose of understanding safety-relevant data trends such as ZrO_2-IMF thermal conductivity as a function of temperature or dopant fraction, for example.

The experimental thermal conductivity of an inert matrix fuel material based on yttria-stabilized cubic zirconia—$Er_xY_yPu_zZr_{1-x-y-z}O_{2-(x+y)/2}$ ($x+y = 0.15$, z: [0.05-0.15])—has been measured and intensively studied.

The hyperbolic thermal conductivity trend with temperature known for pure zirconia, ZrO_2 (similar to that known for urania, UO_2), is reduced by the presence of isotopes, impurities, dopants, and oxygen vacancies, which act as scattering centers. They contribute to conductivity reduction to a flat plot with temperature for stabilized zirconia.

It has also been experimentally observed that the thermal conductivity of $Er_xY_yM_zZr_{1-x-y-z}O_{2-(x+y)/2}$ (with M = Ce or Pu, $z = 0$ or ~0.1 and $x+y = 0.15$) derived from laser flash measurements is rather constant as a function of temperature in the range 300 to 1,000 K. The thermal conductivity was observed to depend on the concentration of dopants such as $YO_{1.5}$ and/or $ErO_{1.5}$, or PuO_2.

For example, the bulk material conductivity of $Er_{0.05}Y_{0.10}Pu_{0.10}Zr_{0.75}O_{1.925}$ is about 2 W m^{-1} K^{-1}. In this study, the thermal conductivity data of both monoclinic and stabilized cubic zirconia-based IMF are tested with model calculations to understand the experimental data in a semiquantitative way. The derived conductivity models were applied for zirconia, accounting for the effects of isotopes, impurities, and dopants.

The model may be only used in a semi-quantitative way and empirical adjusting corrections are needed.

The experimentally observed thermal conductivity, which is rather constant as a function of temperature, is justified theoretically and verified semi-quantitatively when applying the model. The thermal conductivity was experimentally observed and modeled to depend on the concentration of dopants such as $YO_{1.5}$ and/or $ErO_{1.5}$, CeO_2 (analogous to PuO_2) or PuO_2. The thermal conductivity of stabilized cubic zirconia-based IMF may be improved by using a minimum of trivalent dopants (Er, Y), which are plutonium-loading connected, and by producing material with very low porosity. These data allow the calculation of the IMF pellet central temperature in-pile during irradiation.

Degueldre also reported on the fabrication and irradiation of plutonium-containing inert matrix fuels for the "Once Though Then Out" (OTTO) experiment, a JAERI, Nuclear Research and consultancy Group (NRG), and PSI project. This irradiation test, in the High Flux Reactor at NRG, Petten, Netherlands,
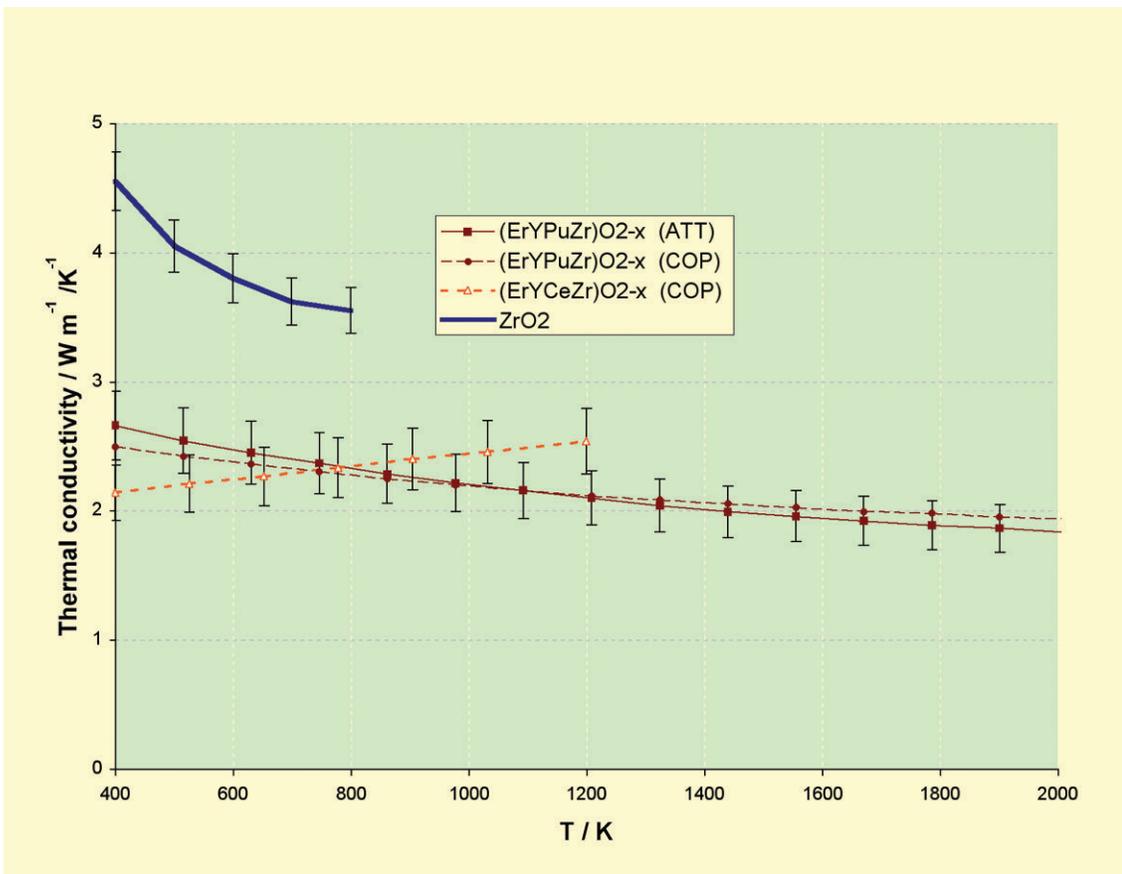
A comparison of bulk thermal conductivities for ZrO_2 (similar data for UO_2) and $Er_{0.04}Y_{0.14}Pu_{0.09}Zr_{0.73}O_{1.91}$, $Er_{0.04}Y_{0.14}Pu_{0.08}Zr_{0.74}O_{1.91}$ as IMF and $Er_{0.07}Y_{0.10}Ce_{0.15}Zr_{0.68}O_{1.93}$ surrogate IMF (with Ce as an analog for plutonium). Note: These data are used to calculate the IMF pellet temperature during in-pile irradiation.

started on Oct. 27, 2000, and was completed in late December 2002.

Seven plutonium-containing fuel segments were prepared for an irradiation experiment in which both zirconia-based and spinel-based targets were irradiated up to a plutonium burnup of about 200 GW d m^{-3}, corresponding to a plutonium depletion of 50 to 60 percent.

For the OTTO experiments, two basic types of plutonium-containing pellets were fabricated: composite pellets and homogeneous pellets. The composite pellets contained spinel as an inert matrix; the homogeneous pellets were based on a zirconia matrix. For the composite spinel fuels, both macro- (250 mm inclusions) and micro-dispersed (25 mm inclusions) fuels were fabricated.

Two IMF pellets (solid solution and macro-dispersed) are shown at the beginning of life before irradiation in the frame of the OTTO project. The pellet on the left is a representative pellet made of $Er_xY_yPu_zZr_{1-x-y-z}O_{2-(x+y)/2}$ material. The pellet on the right is a composite material pellet. Visible microspheres made of $Er_xY_yPu_zZr_{1-x-y-z}O_{2-(x+y)/2}$ are partially popping out of the pellet's white surface, which is made of $MgAl_2O_4$ spinel.

Each fuel contained either uranium or erbium dopant and resulted in four spinel fuels and two zirconia-based fuels. A MOX sample was fabricated for reference. In total seven segments were prepared and irradiated for the OTTO project.
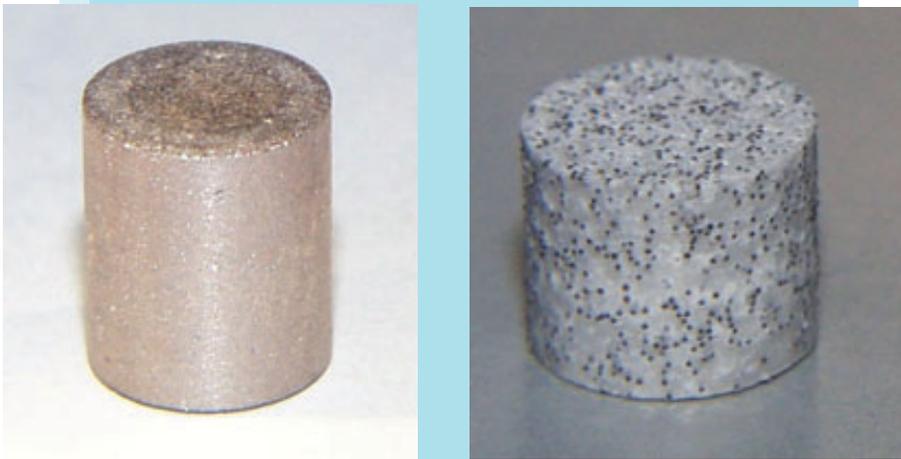
The results of the dimensional measurements, density measurements, ceramographies, and x-ray images of the samples at the beginning of life were given. Some preliminary results of the irradiation were also presented. The on-line analysis of the thermocouples showed good agreement with design calculations derived from thermal conductivity data of the tested IMFs.

The neutrographies of the segments made during the irradiation showed limited axial swelling (less than 2 percent) of the fuel stacks, except for the microdispersed material. The segment, which contained a microdispersed spinel-based sample, appeared to be damaged. Extensive postirradiation examinations were performed in early 2003 and are still in progress.
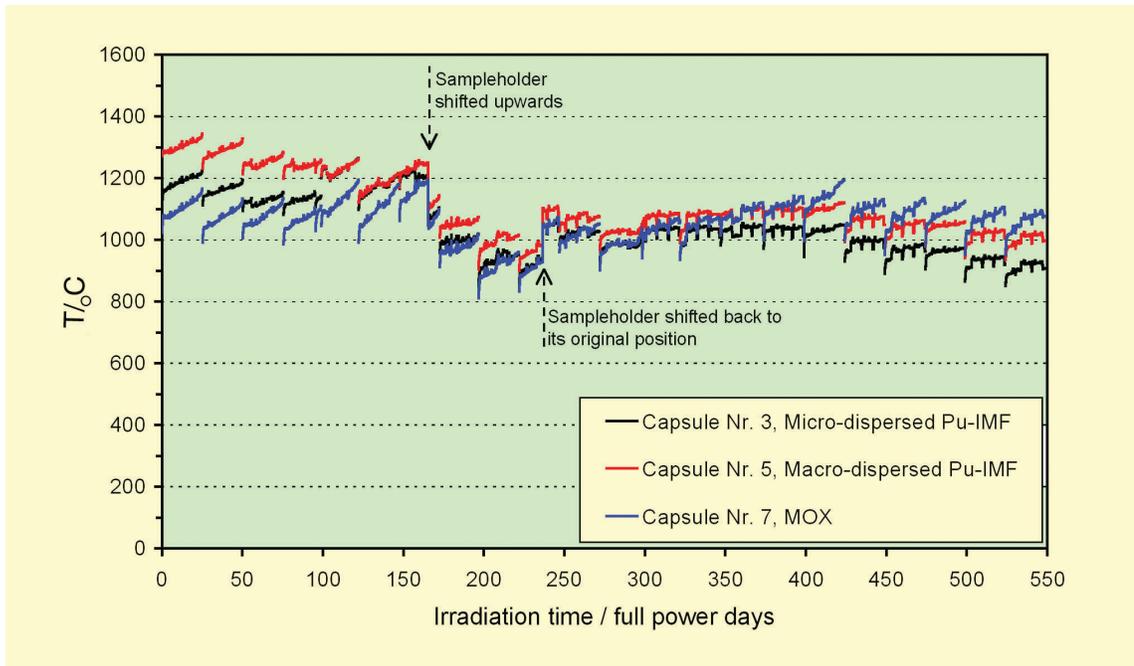
Worldwide interest

The Degueldre-Yamashita paper shows the range and depth of current IMF research. It says that from 1995 through 2002, seven workshops on the topic were held—three in Switzerland, one in Italy, one in France, one "within a European Community organization," and one in the Netherlands.

A total of 350 participants were involved. They came from 17 countries: Australia, Belgium, Canada, the Czech Republic, France, Germany, India, Israel, Italy, Japan, the Netherlands, Russia, the Republic of Korea, Sweden, Switzerland, the United Kingdom, and the United States. Three international organizations participated: the OECD, the Commission of the European Communities, and the

T/°C (y-axis)

Sampleholder shifted upwards

Sampleholder shifted back to its original position

— Capsule Nr. 3, Micro-dispersed Pu-IMF
— Capsule Nr. 5, Macro-dispersed Pu-IMF
— Capsule Nr. 7, MOX

Irradiation time / full power days (x-axis)

The central temperature of the IMF during the OTTO irradiation in the High Flux Reactor, Petten. Comparing the micro- and macro-dispersed IMF composite with the MOX reference.

Neutron radiography of two IMF segments during the OTTO experiment. IMF composition: $(Er,Y,Pu,Zr)O_{2-x}$ and $(Y,Pu,U,)O_{2-x}$. Pellet diameter 8.00 mm, stack length 67.0 and 67.7 mm, density of plutonium fissile at beginning of life: 0.37 and 0.34 g cm^{-3}, density 5.80 and 6.02 g cm^{-3} respectively. This image was obtained after one cycle.

International Atomic Energy Agency. Fourteen universities from all over the world (including the University of New Mexico), 17 national laboratories (including Los Alamos and Oak Ridge), and five industrial firms were represented.
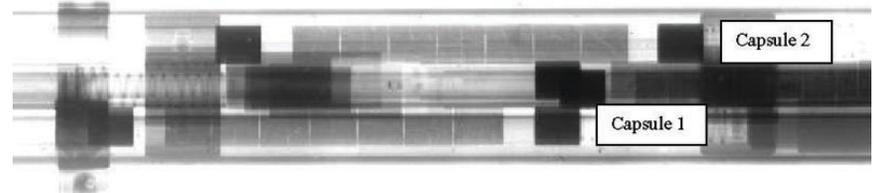
In the last eight years, 86 papers on IMF have been published in the *Journal of Nuclear Materials* and in *Progress in Nuclear Energy*. There have been an additional 88 communications published in five internal reports. Samples and data have also been exchanged, and there have been cooperative activities.

IMF8 was the first Initiative for Inert Matrix Fuel workshop held outside Europe. The Degueldre-Yamashita paper said it "was the consequence of the intensive activity of Japan in the initiative." Sixty participants attended IMF8; they gave numerous presentations during ten sessions. The proceedings of the workshop are published in the *Journal of Nuclear Materials*.

For more information on the upcoming IMF9 workshop to be held Sept. 10-12, 2003, at Sellafield, U.K., see http://www.bnfl.com/website_sellafield.nsf/conference_intro.htm.

Degueldre urged members of his audience to get involved in IMF, a new area of science. "The door is still open for IMF," he said. "You can still join us for collaborative research work."


Capsule 2

Capsule 1

Four NMT photographers win awards

A photograph featured on the cover of "Actinide Research Quarterly" has won first place in the Scientific/ Technical category at the 44th annual Imaging Professionals of the Southwest (IPSW) Conference. Mick Greenbank received the award for his color-enhanced shot of an intermetallic crystal formed by the flux-growth technique (right). The photograph accompanied a story in ARQ 3rd/ 4th Quarter 2002 about researchers discovering unexpected superconductivity in a plutonium compound.
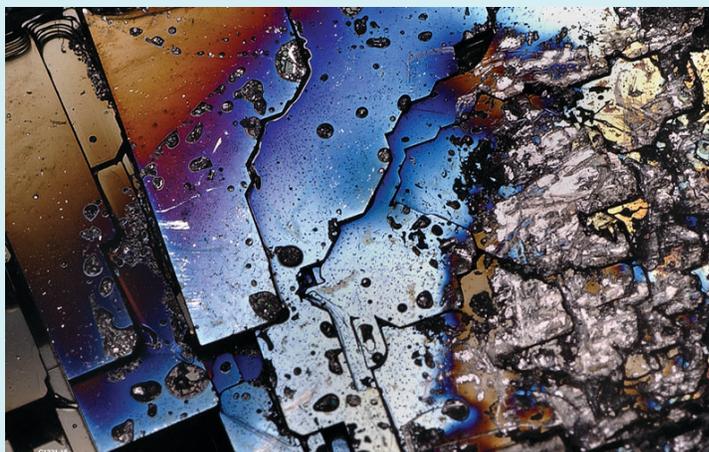
Three other photographers in Nuclear Materials Technology (NMT) Division also received awards at the IPSW competition, which was held in Albuquerque in May. Joe Riedel garnered first place in the Illustrative

Mick Greenbank

black-and-white category for a photo of the interior of the Salt Center in Salt Lake City, Utah. Michelle Stump won first place in Portraiture for her photo of a young girl. Dixon Wolf received both third place and honorable mention in the Scientific/Technical category for his workplace images "Blue Lab" (left) and "Yellow Lab" (below left), respectively. Wolf also won a third-place award in Portraiture for his photo called "Dog Coming and Going."

Greenbank, Riedel, and Wolf are members of Nuclear Materials Information Management (NMT-3); Stump is with Nuclear Materials Science (NMT-16).


Dixon Wolf

ARQ wins Award of Excellence

"Actinide Research Quarterly" has won an Award of Excellence in Technical Communication for 2002 from the Society for Technical Communications. The award was presented by STC's Southwest Regional Chapter to K.C. Kim, chief scientist for Nuclear Materials Technology Division, who has since retired; editor Meredith Coonley, Communication Arts and Services (IM-1); and designer Susan Carlson, also IM-1. The three issues of ARQ submitted for the award in the Technical Publications category were 4th Quarter 2001, 1st Quarter 2002 and 2nd Quarter 2002. Carlson also received an Award of Merit in Technical Art for a poster for "Plutonium Futures—The Science Conference 2003."
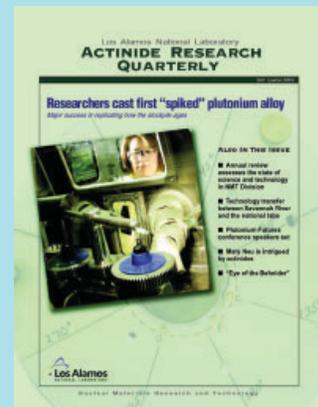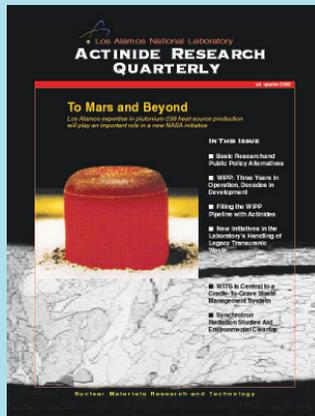

ATOMICS presented as a best safety practice

Nuclear Materials Technology (NMT) Division's ATOMICS process was presented as a best safety practice at the 2002 Executive Safety Summit held in December in Chantilly, Va. Former Los Alamos Director John Browne and Scott Gibbs, deputy associate director for Operations, attended the summit and presented the poster showcasing the ATOMICS behavior-based safety process.

ATOMICS, which stands for Allowing Timely Observations Measures Increased Commitment to Safety, was first introduced to the Laboratory in 1996. The purpose of ATOMICS is to develop and implement an employee-driven, behavior-based safety process for NMT Division. Workers are trained to observe and collect data on safe and at-risk on-the-job behaviors. The data is used to measure safety and address safety problems.

The Executive Safety Summit served to define the path forward for implementation of safety management initiatives and applying an Integrated Management approach to DOE site missions and activities in 2003.
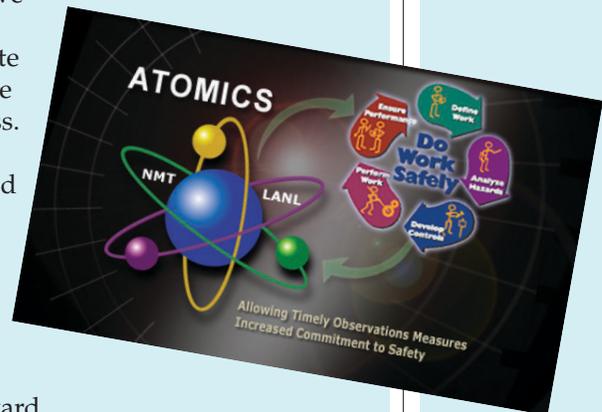
Plutonium Futures Conference next month in Albuquerque

More than 170 papers will be presented at next month's "Plutonium Futures—The Science Conference" in Albuquerque. About four dozen papers will be presented as oral sessions; the rest will be featured during poster sessions.
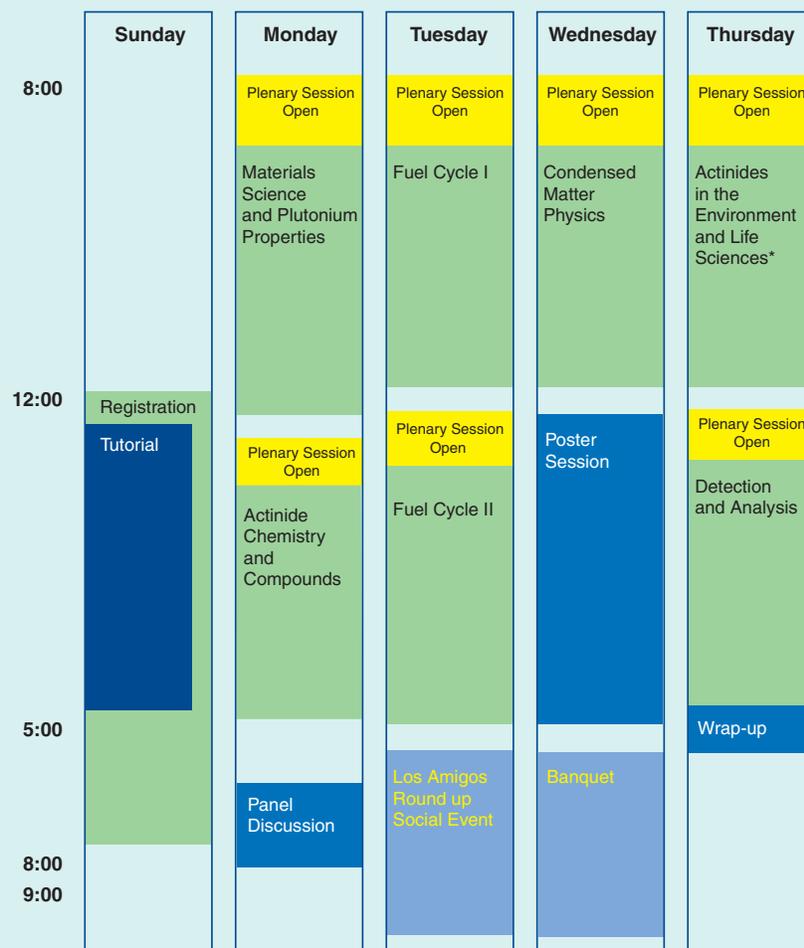
The conference this year will have a slightly different format than in the past. On Sunday, July 6, the Seaborg Institute will host a Plutonium Primer and tutorial. The conference sessions begin Monday, July 7, and continue through Thursday, July 10. Each session will be a half day with a plenary speaker or two at the beginning of each session. Plenary speakers include Vic Reis, former DOE assistant secretary for defense programs; Pierre D'hondt of SCK•CEN; Helen Caldicott, founder of Physicians for Social Responsibility; and many notable researchers in the actinide sciences.

Charles Loeber, author of "Building of the Bombs: A History of the Nuclear Weapons Complex," will be the banquet speaker Wednesday evening, July 9. Loeber worked as a nuclear weapons engineer for more than 37 years for the Department of the Army, the DOE, and Sandia National Laboratories. He has been giving presentations on the history of the nuclear weapons complex for more than 20 years.

The conference will be held at the Albuquerque Marriott Hotel. Late registration will be accepted until opening day of the conference. For more information, check the Web site, www.lanl.gov/pu2003.

JULY 6–10, 2003
ALBUQUERQUE
NEW MEXICO, USA

PLUTONIUM FUTURES
—THE SCIENCE CONFERENCE

Los Alamos
NATIONAL LABORATORY

Plutonium Futures
—The Science Conference
July 6–10, 2003

	Sunday	Monday	Tuesday	Wednesday	Thursday
8:00		Plenary Session Open	Plenary Session Open	Plenary Session Open	Plenary Session Open
		Materials Science and Plutonium Properties	Fuel Cycle I	Condensed Matter Physics	Actinides in the Environment and Life Sciences*
12:00	Registration				
	Tutorial	Plenary Session Open	Plenary Session Open	Poster Session	Plenary Session Open
		Actinide Chemistry and Compounds	Fuel Cycle II		Detection and Analysis
5:00					Wrap-up
		Panel Discussion	Los Amigos Round up Social Event	Banquet	
8:00					
9:00					