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Los Alamos National Laboratory • A U.S. Department of Energy Laboratory

The Actinide Research Quarterly

of the Nuclear Materials Technology Division

In This Issue

Introducing
The Actinide Research Quarterly

My first year as Division Director has been a challenging yet rewarding experience. Although the Division has faced many challenges, I am gratified by the "can do" attitude our personnel continually exhibit. The foundation of NMT Division's science and technology excellence is our capabilities of actinide process chemistry, plutonium metallurgy, surface and separation sciences, actinide ceramics, actinide characterization and analysis, and manufacturing technologies. This new periodical is aimed at communicating NMT's technical progress to TA-55 workers, to peers at Los Alamos, and to customers.

Our latest NMT Division Science and Technology Assessment, just completed in October, is proof of the enormous scientific and technological resources within our Division. Several examples from the Division Review are highlighted in this issue.

K.C. Kim, NMT's chief scientist, has agreed to coordinate and edit *The Actinide Research Quarterly*. I encourage all NMT employees to contribute news items, recent publication titles, and any other materials that will keep our publication informative and interesting.

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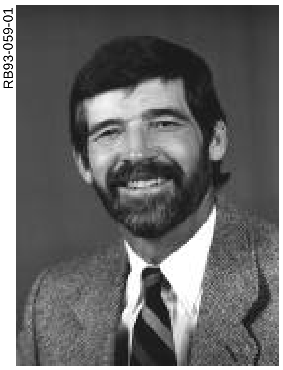

Bruce Matthews, Director of NMT Division

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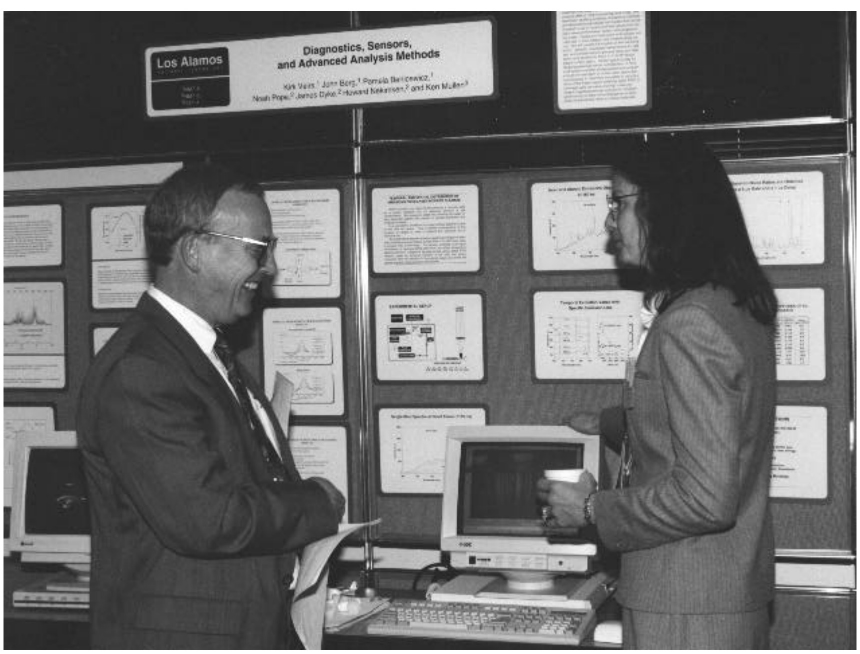


Professor Darryl DesMarteau, an External Review Committee member from Clemson University, discusses a poster presentation with Pam Benicewicz.

NMT Develops Acid Recycling Process

By recycling nitric and hydrochloric acid before discharge, NMT hopes to meet or exceed EPA environmental standards, reduce the radioactivity of effluents discharged from the site, and minimize waste by reusing acids.

NMT is implementing projects that will ultimately help meet the goal of eliminating TA-55 hazardous and radioactive waste discharge, which now contains higher levels of nitrates, chlorides, and radionuclides than the Environmental Protection Agency mandates. By recycling nitric and hydrochloric acid before discharge from the site, NMT hopes to meet or exceed EPA environmental standards, reduce the radioactivity of effluents discharged from the site, and minimize waste by reusing acids.

The acid recycling is a two-step process. First, the waste stream is evaporated to separate volatiles (components of the waste stream that readily evaporate—mostly water and acid in this instance) from dissolved radioactive salts and other inorganic salts. These nonvolatile salts are then mixed with cement for disposal. Second, the evaporated acid-water mixture is separated into a low-acid (mostly water) stream that may be safely discharged and a concentrated acid stream that is suitable for reuse.

Nitric acid is removed from the waste stream by fractional distillation, a method that separates the components in a mixture of volatiles by distilling each component at its respective boiling point. NMT demonstrated the feasibility of removing nitric acid by evaporating a simulated waste stream in the ATLAS line at TA-55. Results of this demonstration were encouraging: 82% of the nitric acid was recovered during the first pass through the ATLAS line. In the second and third passes of clean distillate through the evaporator, 43% and 21%, respectively, of the original nitric acid feed was concentrated to 10.7 *M* HNO_3 and 12 *M* HNO_3. The distillation column to be built will enable continuous production of ≥12 *M* HNO_3.

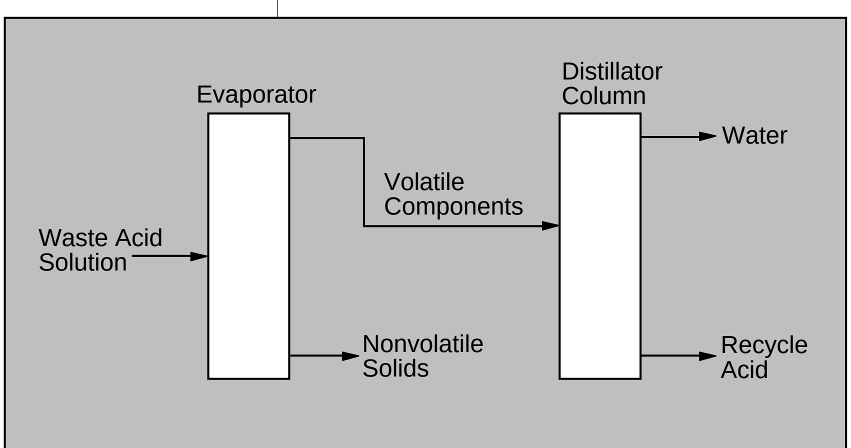
NMT designed a 10-inch-diameter, 13-foot-tall stainless steel column with a reboiler and condenser for distillation of nitric acid. Once built, the column will operate in room 434 (the old evaporator room) inside the Plutonium Facility and will concentrate 500-liter batches of waste solution with a nitric acid concentration of 2 to 6 *M* to a concentration of 12 *M* HNO_3. Automated controls will enable the system to sense process changes (such as a change in acid concentration in the incoming waste stream) and to change operating conditions so that final acid concentrations remain constant.

The recycling of hydrochloric acid requires a slightly different approach. Because a simple distillation of water and hydrochloric acid limits its maximum acid composition to about 6 *M* HCl, concentrating hydrochloric acid beyond this level from the waste stream is somewhat more difficult than concentrating nitric acid. NMT will employ a vapor-phase membrane separator to solve this problem. The separator will employ tubes of Nafion, a DuPont product that permits water in the stream to pass through while trapping hydrochloric acid.

As national environmental regulations become increasingly stringent, improved waste treatment methods such as NMT's acid recycling at TA-55 become critically important to the welfare of both the Laboratory and the nation.

Project contributors include **Tom Mills, Elaine Ortiz, Noah Pope, Wayne Punjak, Steve Schreiber, Louis Schulte, Brad Smith, Coleman Smith, Wayne Smith, and Steve Yarbro.♦**

Figure 1. Acid recycle and recovery system.



Milliwatt Surveillance Program Ensures RTG Safety and Reliability

The U.S. military's need for a rugged, reliable power source with a long life was met with the radioisotope thermoelectric generator, or RTG. The battery-like RTGs provide power for permissive-action links, a family of devices that reduce the possibility of obtaining detonation from a nuclear warhead unless a controlled numerical code is used.

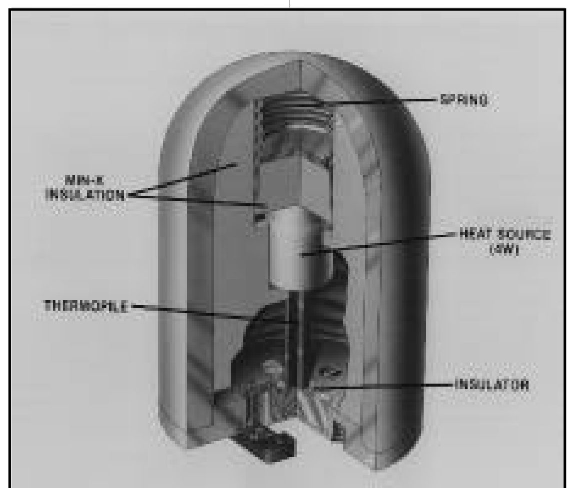
Often referred to as "nuclear batteries" by the media, RTGs convert heat to electricity by harnessing the thermal energy produced by decay of radioactive isotopes—usually plutonium-238. A silicon-germanium thermopile inside the RTG converts the heat energy into electricity.

Because the milliwatt RTG operates from a nuclear heat source rather than from chemical action as in regular batteries, the power sources provide a minimum of 25—30 years of service life for tens of milliwatts of electrical power without needing replacement. Theoretically, the devices are capable of producing electrical power much longer, but helium, a byproduct of isotope decay, can build up and eventually rupture the casing.

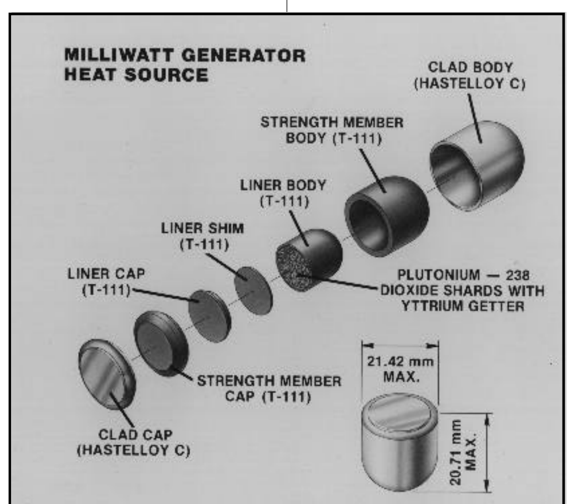
These RTGs are relatively compact—4.9 cm in diameter and 8.8 cm long for the 4.5-watt model and 6.8 cm by 9.3 cm for the 4.0-watt version.

The General Electric Neutron Devices Department in Largo, Florida, manufactured RTGs from 1975 to 1990, when all production of the devices ended. NMT-9 began its involvement with the Milliwatt RTG Program in 1980, when the Group began manufacturing heat sources for RTGs. Today NMT-9 performs a broad range of RTG activities, including surveillance testing of shelf-life and stockpile returns, shelf-life heat source inventory, and destruction of excess RTGs and heat sources. All RTG testing and plutonium reclamation activities are done by NMT-9 except vibration and mechanical shock testing, which is performed by ESA-MT.

The heat sources for milliwatt RTGs are identical except for the amount of plutonium oxide granules contained in the 4.0-watt and 4.5-watt models. EG&G Mound Applied Technologies qualified initially all lots of the alloy used to encase heat sources with pressure-burst tests. Production lot testing of T-111 has been consistent with the desired probability of failure being less than 0.005 for the conditions of a 25-year-old, 4-watt heat source exposed to a 1010-degree-Centigrade fire for two hours. Plutonium heat sources are enclosed first in a 0.51-mm-thick liner of alloy T-111 (90% Ta, 8% W, and 2% Hf), then in a 1.02-mm-thick strength member of T-111, and finally in a 0.51-mm-thick clad of oxidation-resistant Hastelloy C (55% Ni, 17% Mo, 16% Cr, 5% Fe, 4% W).



NMT-9 performs electrical tests on all shelf-life and stockpile-return RTGs. These tests include thermopile impedance, RTG open circuit voltage, RTG loaded (130 ohm) voltage, and RTG base isolation resistance. A thermopile tester in PF-3 can also test eight characteristics of thermopile operation. Some RTGs are also selected for vibration and mechanical shock testing by ESA-MT personnel. RTG output voltages before, during, and after the tests can then be compared.



Trent Latimer is Project Leader of the Milliwatt Surveillance Program.♦

The low-temperature sublimation separation (LTSS) process is an excellent method for separation of radioactive materials and will likely lead to applications throughout the nuclear industry.

Low-Temperature Sublimation Separation

Many methods exist for removing radioactive materials from liquids, slurries, and sludges. Some methods use elevated temperatures to drive off the solvents and some use filtration or membrane separation to remove suspended particulates. Still others use sophisticated ion-specific exchange columns to separate solutes and solvents. Very little is known, however, about decontamination of radioactive liquid waste at low temperatures and pressures. Because freeze drying technology uses low temperatures and pressures to remove a solvent or volatile component from a frozen solution by sublimation (drying), it promises to show an intrinsically high separation possibility. NMT investigated the process to determine its effectiveness in decontamination of radioactive liquid waste. The resulting low temperature sublimation separation (LTSS) process proved to be an excellent method for separation of radioactive materials and will likely lead to applications throughout the nuclear industry. The process features

- high decontamination / separation factors;
- low temperature and pressure operation;
- no use of additives, which require large volume reductions;
- small solid residue volume;
- low maintenance cost (no moving parts);
- low energy input;
- simple, safe, unattended operation; and
- well-established technology with wide-ranging applications.

The vacuum sublimator shown in Figure 4 illustrates the LTSS process. Waste solution is introduced into the sublimation chamber, which contains the heat exchangers. Ice forms around the heat exchanger cold surfaces. Later, heat is applied to the heat exchanger to facilitate sublimation. The product chamber, which contains a condenser, collects sublimed (nonradioactive) water vapor and stores it as ice. Because the sublimation operates at very low pressures, the water molecules (and other volatile solvents) sublime continuously from the sample chamber ice surface and are transported to the condenser side and refrozen at the colder condenser surfaces.

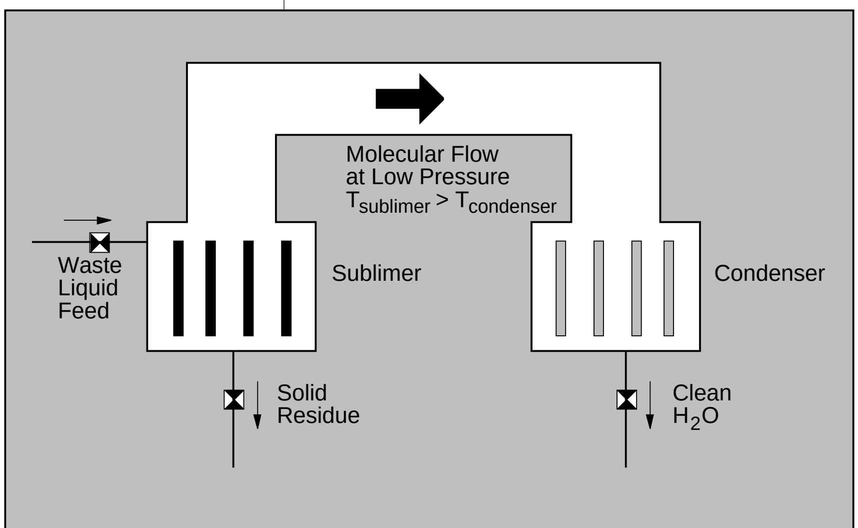


Figure 4. LTSS vacuum sublimator.

As heat is supplied to the frozen material for sublimation, the difference in temperature between the two chambers drives mass transport of the nonradioactive solvent from the sublimation chamber to the condenser. Steady state transport conditions are achieved by continuously supplying the heat of sublimation to the frozen solution in the sublimation chamber and continuously removing it at the condenser. This process of mass transport of condensable vapors between two condensing surfaces maintained at two different temperatures is best accomplished in the absence of noncondensing gases, such as air in this case. These noncondensing gases are initially pumped away from the chamber before starting the sublimation separation.

LTSS is the first application of the freeze drying process to consider the condensate itself the product, rather than the solid residue. This new twist has drawn the attention of several commercial manufacturers and developers to this work.

Experiments performed recently on uranium containing reactor water showed that decontamination factors of 6×10^7 were achievable. NMT work on depleted uranium containing 200 g of uranyl nitrate per liter yielded a condensate with uranium concentrations below the detection limits of sensitive analytical methods. Work is planned for other radioactive materials, including plutonium.

The LTSS process can be enhanced by other low-temperature methods such as freeze concentration and fractional precipitation, two concepts that are very closely related. Both involve separation of a solvent and solutes by lowering the solution temperature. Exactly which process occurs depends on the initial concentration of the solution. Freeze concentration occurs when a solution of relatively low concentration is cooled to its freezing point. As a solution cools, pure ice precipitates while the solution becomes more concentrated. Fractional precipitation refers to the same process when more than one solute is present in solution at high concentrations. In that case, if one solute is radioactive then it is possible to segregate the solutes.

Project contributors include **Tom Blair, Nick Coppa, Eloy Cordova, Bobby Eustler, Rudy Fernandez, John Franklin, Ubaldo Gallegos, Ed Martinez, Jim McFarlan, Jim McHale, and Wilfred Romero.**♦

LTSS is the first application of the freeze drying process to consider the condensate itself the product, rather than the sublimed solid. This new twist has drawn the attention of several commercial developers to this work.

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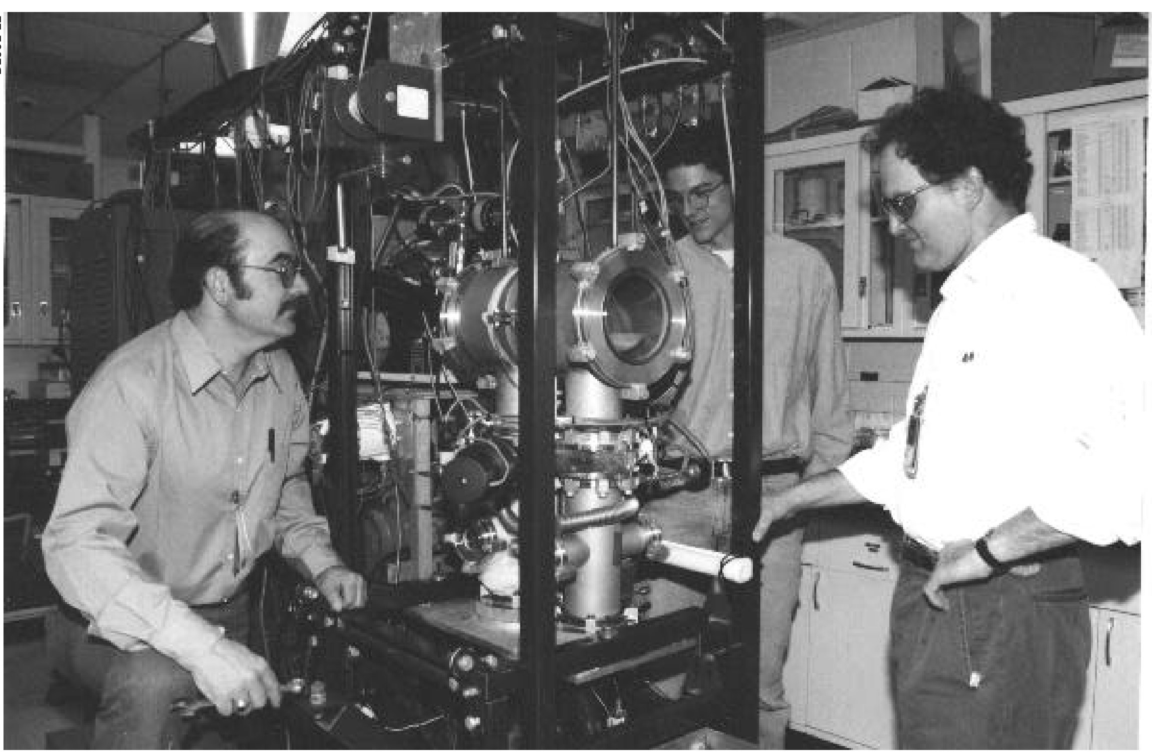
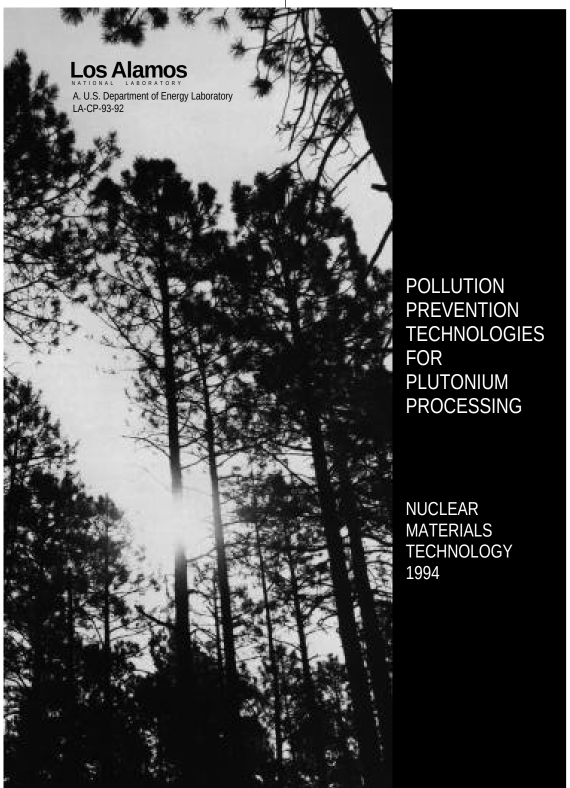


Ed Martinez (left), John Franklin (center), and Nick Coppa (right) with the freeze drying experimental apparatus.

NMT Division Science and Technology Assessment a Success

The University of California's contract with DOE requires that the Laboratory perform an annual science and technology assessment. In fiscal year 1994, each technical division and program office was required to perform such an assessment. NMT Division performed its self-assessment in the three-month period between April and June of this year. The Division prepared a report of this work and submitted it to the External Review Committee (which reports to the Laboratory Director) to assist their assessment. This report and the Committee's report will be integrated into the final Laboratory report submitted to the University of California.

The NMT Division Science and Technology Assessment, formerly called a Division Review, was conducted October 24—26, 1994, at the J. Robert Oppenheimer Study Center. The first day of the assessment was devoted to ERC orientation and a tour of the Plutonium Facility. The second and third days were devoted to technical presentations and poster sessions. This year's review theme was "Pollution Prevention Technologies for Plutonium Processing." A dozen technical presentations and approximately two dozen poster presentations highlighted a broad range of the Division's significant scientific and technical activities during the past year to two years.



Deputy Laboratory Director Jim Jackson reports, "I wanted to take a few minutes to congratulate you and your division for doing an excellent job with your Science and Technology Assessment. The External Review Committee was very complimentary of your presentations. They thought the quality of your technical work was excellent." The proceedings of this year's review presentations have been published in two documents (LACP-94-213 and LALP-93-92).

For further information, contact K. K. S. Pillay at (505) 667-5428.

B1883-2

Staff Member Tim Nelson (left) leads one of the tours of the Plutonium Facility for members of the External Review Committee.

B1883-6

Bruce Matthews leads the division update of the NMT Science and Technology Assessment held at the Study Center on October 25.

Recent Publications and Reports

The following reports and publications were processed by the Division Office during this quarter:

K. M. Axler, P. C. Lopez, and J. A. McNeese; "Los Alamos Site Update," 14th Annual Pyrochemical Workshop; Boulder, Colorado, October 31–November 3, 1994.

K. M. Axler and P. C. Lopez; "Process Optimization, and Equipment / Material Substitution for Waste Minimization," 1995 Los Alamos National Laboratory Pollution Prevention Showcase, Santa Fe, New Mexico; January 16–18, 1995.

R. A. Bibeau, A. Sandoval, N. Pope, and T. Hayes; "A Computer-Based Supervisory Control and Data Acquisition System for Plutonium Waste Recovery," (Informal Distribution), October, 1994.

B. Cort, J. Ward, F. Vigil, and R. G. Haire; "Resistivity Studies of Cubic Americium Hydrides from 20 to 300 K," (submitted to *Journal of Alloys and Compounds),* November, 1994.

V. R. Dole, E. Garcia, J. A. McNeese, and W. J. Griego; "Distillation Separation of Chloride Salts from Plutonium," 14th Annual Pyrochemical Workshop, Boulder, Colorado, October 31–November 3, 1994.

J. Johnston, P. Trupp, Y. Rivera, and J. Lugo; "Glovebox Safety Training," Shaping the Environment Session of the 11th Symposium on Training of Nuclear Facility Personnel, San Antonio, Texas; April 9–13, 1994.

S. M. Long, "Design and Fabrication of NDA Standards," 1994 Plutonium / Uranium Recovery Operations Conference, Knoxville, Tennessee; October 17–20, 1994.

S. M. Long, "Design and Fabrication of NDA Standards," 17th Symposium on Safeguards and Nuclear Material Management, Cologne, Germany; May 9–11, 1995.

S. M. Long, "Update on Fabrication of 12% PU-240 Calorimetry Standards," Second International Workshop on Calorimetric Assay, Santa Fe, New Mexico; November 14–16, 1994.

B1883-30

Nick Coppa (right) presents his research on new approaches to the immobilization of nuclear materials to interested attendees.

K. K. S. (Sam) Pillay, "Disposition Scenarios and Safeguardability of Fissile Materials Under START Treaties," 1993 ANS Winter Meeting, San Francisco, California.

M. A. Reimus, B. T. Martinez, J. R. Stevens, and T. E. Boyd; "Pilot-Scale Ash Dissolution and Dicesium Hexachloroplutonate (DCHP) Precipitation," Los Alamos National Laboratory report LA-12 890-MS (UCNI).

N. A. Rink, Editor, "Quarterly Status Report of the Nuclear Materials Packaging and Repackaging Project and Vault Work-Off Project, Fourth Quarter of FY 1994," September, 1994.

P. L. Wallace, L. D. Calvert, M. H. Mueller, T. C. Huang, J. N. Dann, and S. Weissmann; "A New Materials Data Compilation Can Help in Actinide Phase-Diagram Research," in *Actinide Processing, Methods and Materials* (B. Mishra and W. A. Averill, eds.), pp. 5–20; The Minerals, Metals, and Materials (TMS) Society, Warrendale, Pennsylvania, 1994.

L. A. Worl, F. C. Prenger, T. L. Tolt, A. R. Schake, D. D. Padilla, D. R. Romero, and D. D. Hill; "Magnetic Separation CRADA 1994 Phase Two Report," Milestone Report for CRADA to DOE / EM-50, October, 1994.

C. Zawodzinski, W. H. Smith, and K. R. Martinez;"Cobalt (III) - Catalyzed Electrochemical Oxidation of Selected Organics," Eighth International Forum on Electrolysis in the Chemical Industry, Lake Buena Vista, FL; November 13–17, 1994.

Newsmakers

NMT Wins LDRD Funding

This year, NMT won funding for three Laboratory-directed research and development (LDRD) projects that began in FY95. The LDRD program, managed by the LDRD office in the STB Program Directorate, is designed to fund innovative research and development ideas. The LDRD program is divided into three areas: individual projects for fundamental research; program development, which identifies significant program development opportunities; and competency development, which strengthens Laboratory core competencies.

Two NMT proposals were recommended for funding in the program development category. **Ken Chidester** of NMT-9 is the principal investigator for the proposed work "Disposition of Weapons Plutonium as Non-Fertile Fuel for Light Water Reactors." Ken is also project leader of the newly established Plutonium Disposition Project and this LDRD funding will augment the fundamental research component of Ken's work. **Nicholas Coppa** of NMT-6 is the principal investigator for "Decontamination of Radioactive Liquids by Freeze Concentration and Fractional Precipitation" (see related article on page 4).

Barbara Cort of NMT-5 submitted a successful proposal, "Structural and Magnetic Characterization of Actinide Materials," that was recommended for competency development funding in the nuclear and advanced materials core competency area.

Mark Williamson of NMT-6 is expected to receive funding for half of his time in the Accelerator-Based Conversion Program. Also, **K. C. Kim**, Division LDRD point-of-contact, is working to secure additional funds from the LDRD reserve to support two to three small innovative research ideas.

Congratulations to all Division employees who worked on these successful proposals for LDRD funding!

Keith Axler

Keith Axler, a staff member in NMT-6, has finished the thesis for his Ph.D. degree from the Colorado School of Mines. The title of Keith's thesis is *Engineered Materials for Application in Severe Metallurgical Environments.*

NMT Division Submits its Top Three Technical Accomplishments for 1994

In response to a Newsbulletin request for top technical accomplishments during the year, NMT submitted a list of three ongoing projects: High Gradient Magnetic Separation, Open Architecture Machine Control System, and Low-Temperature and -Pressure Separation and Stablization of Radioactive Materials. These accomplishments will be included in the Newsbulletin's 1994 year-in-review article, scheduled for publication on January 6, 1995.

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The Actinide Research Quarterly is published quarterly to highlight recent achievements and ongoing programs of the Nuclear Materials Technology Division. We welcome your suggestions and contributions.

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