



The Actinide Research Quarterly

of the Nuclear Materials Technology Division

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NMT's Contributions to the Cassini Saturn Mission Follow Division's Space Exploration Tradition

Some of NMT Division's handiwork will be soaring across the solar system on its way to Saturn in the near future. Many NMT members, primarily in Actinide Ceramics and Fabrication (NMT-9), have produced special heat and energy sources to help keep things running aboard the Cassini orbiter and Huygens probe, which will be launched in October of 1997 toward the Saturnian system to explore the gas giant, its mysterious rings, and some of its frigid moons.

Because Saturn is so far away from the sun, solar rays there are only a small fraction as strong as they are on earth. To gain enough electrical energy and warmth to operate, Cassini would need solar panels so large that it would be nearly impossible to launch and maneuver. Hence, the orbiter and probe must carry their own thermal and electrical generators to run the scientific experiments and other equipment aboard. A conventional battery cannot be used because of the mission's long duration and the prohibitive weight of such an energy source.

Long-lasting energy without excess weight can be provided by the natural radioactive decay of plutonium-238. With a half-life of approximately 87 years, this material will reliably produce a constant flow of energy for at least 25 years and for even

longer thereafter at reduced levels. For the Cassini mission, radioisotope thermoelectric generators (RTGs) will provide the electrical energy needed, while lightweight radioisotope heating units (LWRHUs, or RHUs for short), will keep equipment warm enough to function.

Cassini will use three RTGs, which convert thermal energy from plutonium decay into electrical energy. Each RTG contains 72 small pellets of plutonium-238 dioxide, each about the size of a marshmallow and weighing 150 grams. Each pellet is encased in many layers of protective materials, and the complete unit is called a general purpose heat source (GPHS) module. All together these 72 heat sources put out 4400 watts of thermal energy at an operating temperature of 1200°C to 1300°C. Using a set of thermocouples, the RTG converts this heat energy to about 285 watts of electricity.

To maintain operating temperatures, the Cassini orbiter and Huygens probe will use 157 RHUs distributed in various locations.

Each unit contains a 2.7-gram pellet of PuO₂ encased in a platinum-rhodium alloy, which, in turn, is protected inside a graphite shell. This multilayer casing, as well as that for the GPHSs, is designed to keep the PuO₂ safely contained when subjected to

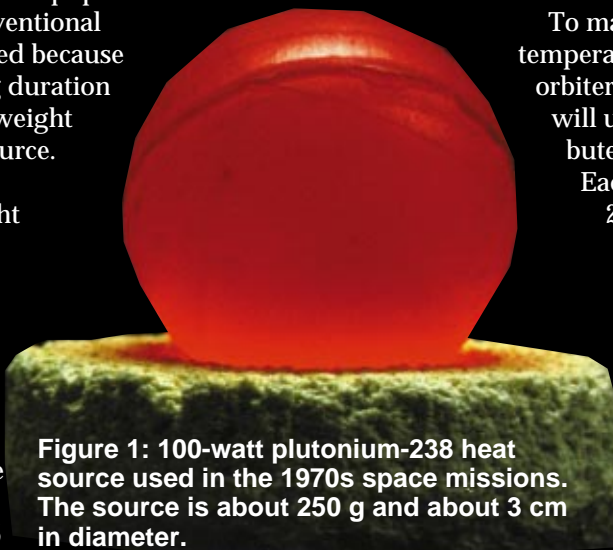


Figure 1: 100-watt plutonium-238 heat source used in the 1970s space missions. The source is about 250 g and about 3 cm in diameter.

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Guest Editorial

Managing and Minimizing Radioactive Waste Continue to Challenge the DOE Complex



K.K.S. Pillay,
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Over the past five decades, the Congress of the United States has promulgated numerous environmental statutes applicable to all sectors of the U.S. economy. In 1969 the National Environmental Policy Act (NEPA) initiated revolutionary changes to reduce waste generation and prevent environmental pollution. A series of laws and regulations following NEPA has increased pressure on both industry and the government to be responsible managers of the environment. After extensive public debate resulted in the Nuclear Waste Policy Act of 1982, there have been many positive developments directed at long-term management of radioactive wastes in the U.S. Although several amendments to 1982's landmark legislation have changed the scope of the efforts, the ultimate goal continues to be isolation from the biosphere of long-lived radioactive nuclides in wastes. Waste management of plutonium (and other actinide) processing is a subset of these and other regulatory requirements. The specific topic addressed in this editorial is the management of primary waste generated from plutonium processing.

During the past 55 years, the plutonium inventory in the U.S. increased from half a microgram to nearly 400 M tons. The U.S. defense sector produced an estimated 111.4 M tons of plutonium between 1944 and 1988, and about 4% of that now exists in numerous waste forms. This estimate does not include the plutonium that was released to the environment during nuclear explosions and is now part of the natural background worldwide. It is estimated that the 100,000 m³ of transuranic wastes now stored at six DOE sites contain 3.4 M tons of plutonium. Approximately 2 M tons of plutonium and other actinides are present in high-level liquid wastes currently stored at three DOE sites. A significant positive inventory difference of nuclear materials within the DOE complex includes an additional 2.8 M tons of plutonium. At least some of this plutonium is likely to end up in transuranic (TRU) waste streams. The majority of TRU wastes is expected to be transferred to the Waste Isolation Pilot Plant (WIPP), and the

high-level wastes at two sites (Savannah River and West Valley) are being vitrified for eventual geologic disposal. Ongoing environmental restoration, decontamination, and decommissioning of DOE facilities coupled with future operations within the weapons complex, even though they will be limited, are expected to generate another 78,000 m³ of TRU wastes during the next two decades. Current plans are to place all TRU wastes, both contact-handled and remote-handled, in the WIPP facility in Carlsbad, New Mexico, when the facility becomes available. It is now scheduled to open in April 1998, almost 19 years after WIPP construction was authorized by the U.S. Congress. The opening of WIPP is contingent upon DOE receiving EPA's certificate of compliance and a favorable Resource Conservation and Recovery Act-related decision by the State of New Mexico.

Although many programs specifically address the legacy wastes at the DOE sites, facilities that routinely generate wastes from continued operation have been placed in an awkward position. In conformance with all applicable mandates from the U.S. Congress and the President, all federal facilities are required to reduce waste generation by 50% by 1999, using baselines established several years ago. However, corrective actions mandated by the Defense Nuclear Facilities Safety Board for safety issues, as well as insufficient financial resources, have limited their ability to comply with the above laws and regulations in the near term.

An unexpected new source of TRU wastes headed for WIPP will result if one of the DOE sites declares a large portion of its actinide residue inventory as waste. The alternatives proposed for managing the plutonium residues at Rocky Flats have a common feature: packaging and shipping from 3.1 M tons to 6.3 M tons of plutonium from Colorado to New Mexico in the most expeditious manner. This approach, if it takes place, will merely transfer the problems of material chemistry, confinement, and storage from the present location to another location outside Colorado. It seems that the only criterion used

in developing the strategies is a reduction in the mortgage for on-site storage. The relative abundance of plutonium in these residues would also pose serious nuclear material safeguards concerns. The alternatives presented by Rocky Flats have the potential to increase the plutonium content of WIPP wastes by at least 100% (possibly 200%) of the previous estimates. This dramatic change in the role of WIPP to that of a dump for excess plutonium has the potential to create additional problems and delays for opening WIPP.

Although there are large allocations of resources to manage legacy wastes, the management of newly generated wastes has been relatively neglected in DOE's planning. The present and future operating plans for plutonium facilities within the DOE complex require compliance with all existing environmental regulations and the special regulations that are applicable only to federal facilities. Following the passage of the 1990 Pollution Prevention Act, both the EPA and DOE have proposed a variety of waste minimization and pollution prevention goals. The U.S. industry has demonstrated significant progress in

pollution prevention through major process changes, resource recovery and recycling, and investments in new technologies. Because the DOE was late in adopting the mission of environmental management, a new strategy is necessary to hasten progress in waste minimization within the DOE weapons complex.

According to recent reports by the General Accounting Office and the National Research Council, the ambitious plan now pursued by DOE for waste minimization and pollution prevention is eclipsed by internal organizational problems and unrealistic expectations. An operating facility such as the Plutonium Processing Facility at Los Alamos must comply with all environmental regulations and other mandates. However, the Defense Program Office of DOE, which uses the facility, has invested very little in new technologies needed to meet these mandates. Governments may regulate, environmentalists may agitate, but it is only the technology experts who can innovate to solve the problems of waste generation. Let us hope that they will be listened to and their advice taken.

“Governments may regulate, environmentalists may agitate, but it is only the technology experts who can innovate to solve the problems of waste generation.”

Division Entices Students with Overview of Actinide Research

On August 28, 1996, NMT Division opened its doors to all Laboratory postdoctoral researchers and graduate research assistants for its first one-day seminar on “Careers in Actinide Sciences.” Thirty-five current Laboratory “postdocs” and students participated in exciting, unclassified discussions of plutonium-related programs at TA-55. This event, supported by the Laboratory upper management, was intended to inform attendees of our work and interest them in careers in actinide sciences.

Allen Hartford discussed the Laboratory postdoctoral and student recruiting program and the opportunities that NMT presents to the new work force. Nuclear Materials and Stockpile Management Program Director Paul Cunningham talked about national and international nuclear materials issues and the program's long- and short-term goals. NMT Division Director Bruce Matthews presented his vision for the disposition of nuclear materials, the division's mission, and our scientific and technological opportunities to help reduce the nuclear danger. Group Leaders of NMT-2, NMT-5, NMT-6, and NMT-9 discussed their individual groups' specific programs, activities, and potential growth areas. Anyone interested in any of NMT's programmatic activities are encouraged to contact NMT group leaders or project leaders about their specific areas.

Wayne H. Smith, and Douglas E. Wedman of NMT-6, are the principal researchers.

Electrodialysis Reduces Waste in the Treatment of Pyrochemical Salt Residues

Many of the processes used to treat residues from plutonium recovery and purification operations require the addition of large quantities of reagents. The result is substantial volumes of waste solutions that require additional treatment before they can be disposed of. Examples of such processes include aqueous treatment methods for pyrochemical salt residues. Typically, the chloride-based salts and other residues are dissolved in concentrated hydrochloric acid, additional reagents are added to adjust the plutonium oxidation state, then the plutonium is separated from the remainder of the matrix via ion exchange and/or solvent extraction. While the plutonium is collected in concentrated form, the original dissolver solution, rinse solution, eluant solution, and/or back-extraction solution remain to be treated before disposal.

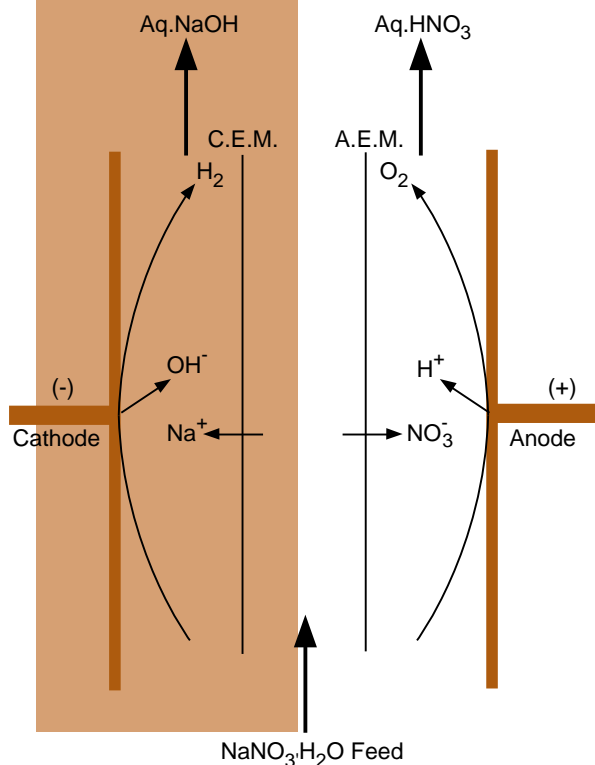


Figure 2. Electrodialysis of a sodium nitrate solution. The positively charged sodium ions in the feed solution migrate through the cation exchange membrane toward the negatively charged cathode. The electrochemical reaction taking place at the cathode is the reduction of water to form hydrogen gas and hydroxide ions. The net result is formation of sodium hydroxide in the catholyte strip solution. The negatively charged nitrate ions in the feed solution migrate through the anion exchange membrane toward the positively charged anode. The electrochemical reaction taking place at the anode is oxidation of water to form oxygen and free protons. The net result is formation of nitric acid in the anolyte strip solution.

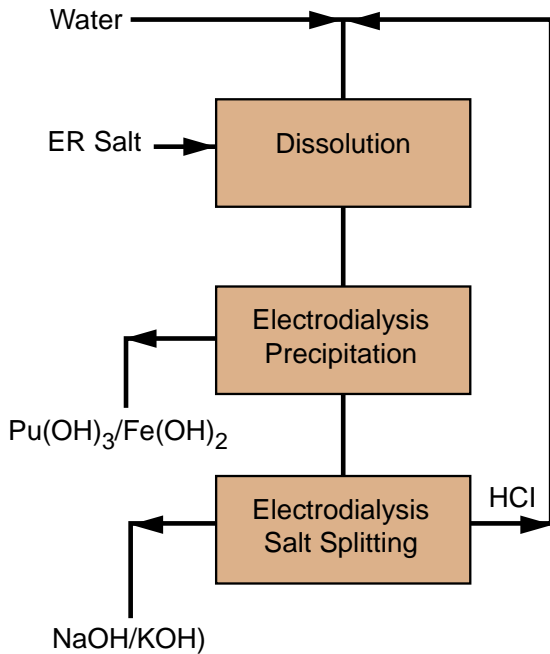
As part of a research effort to find more efficient processes that generate less waste, we have been investigating the use of electrodialysis to treat process residues. Electrodialysis uses an applied electrostatic field coupled with ion exchange membranes to split salts into their respective acid and base components. This process is shown schematically in Figure 2. The electrodialysis process has a very high efficiency and can be run until essentially all of the ions have been stripped from the feed solution. Its major industrial application is to create potable water from sea water by lowering the salt concentration in the feed solution down to the parts-per-million level.

Electrorefining (ER) salts, because of their relatively uncomplicated chemical composition, were the first materials chosen to demonstrate the applicability of electrodialysis to the treatment of plutonium process residues. These salts are predominantly a 1-to-1 mixture of sodium and potassium chloride salts containing small quantities of plutonium in the form of metal and salts, and lesser amounts of americium and other heavy metals salts.

A proposed flow sheet for treating ER salts by electrodialysis is shown in Figure 3. The salt is first dissolved in a minimum amount of approximately 1 M hydrochloric acid. The resulting solution is then passed through a three-compartment electrolysis cell. As the feed solution flows through the cathode compartment of this cell, the acidic solution is slowly neutralized as hydroxide is generated at the cathode. As the pH increases, the heavy metals, including plutonium, undergo a homogeneous precipitation and can be collected on a filter configured into the recycle loop. At this point the relatively small amounts of heavy metals and actinides will have been separated from the large residue matrix and can be further treated in concentrated form for disposal or recovery of the actinides.

The feed solution, which now contains only trace quantities of actinide elements, is then sent to the central compartment of a conventional electrodialysis cell for a salt-splitting operation. Here hydrochloric acid is generated in one strip solution while a mixture of potassium

Figure 3. Proposed Electrodialysis/ER Salt Flow Sheet.



and sodium hydroxides is generated in another. The hydrochloric acid stream can be recycled to the head-end process for dissolution of the next ER salt to be processed and/or to be used for other chloride processes. Before it is finally disposed of, the electrogenerated sodium/potassium hydroxide solution can be used as a final polishing agent for both chloride- and nitrate-based waste solutions for further removal of actinides.

In the electrodesalination flow sheet the only reagent added is water; dissolution of the salt is accomplished by hydrochloric acid recycled from the electrodesalination operation. Separation of plutonium from the salt solution is carried out by homogeneous precipitation using electrogenerated hydroxide ions. No additional oxidizing or reducing agents, washing, or eluent solutions are required. The net result is a tremendous decrease in the quantity of reagents used and a corresponding decrease in the quantity of waste generated. Also since a majority of the solutions generated are recycled back into the system, there will be a greater probability for removal of the actinides from the process solutions with each successive cycle.

The proposed electrodesalination flow sheet was demonstrated in a “cold” experiment using neodymium as a surrogate for plutonium, and iron as a representative for all heavy metals that might be present in an actual ER salt residue. In the first electrodesalination stage greater than 99.99% of the neodymium and iron were stripped from the feed solution and collected on the filter. In the second electrodesalination stage the salts were stripped from the feed solution down to the parts-per-million level, and the final compositions of the strip solutions were approximately 6 M mixed sodium/potassium hydroxide and 3 M hydrochloric acid. The concentration of the alkaline solution is sufficient to use “as is” in the final polishing step of the waste stream solution. The acid solution is more concentrated than necessary to recycle to the head end of the process to dissolve the next ER salt, but it could be diluted with a quantity of the deionized feed solution. Other aqueous-chloride-based processes use much more concentrated hydrochloric acid solutions, requiring a preconcentration of the strip solution before use. A separate process to accomplish this task is currently under development.

Future work planned for this project calls for performing “hot” tests on actual ER salt residues. Once the electrodesalination technology has been demonstrated on this chemically noncomplex system, more chemically complex residues will be investigated. Of particular interest are the calcium-based direct oxide reduction salts, which contain many more heavy metal impurities and in greater concentrations than in the ER salts. However, a greater challenge than removing the actinides may reside in developing a flow sheet that can handle the calcium ion, which has only limited solubility in alkaline media.

In summary, preliminary investigations have demonstrated the applicability of electrodesalination to the treatment of selected plutonium process residues. The net benefits in the use of this technology are a tremendous decrease in the quantity of reagents used and corresponding decrease in volumes of waste generated compared to the aqueous recovery processes currently in use.

“The net result (of using the electrodesalination process) is a tremendous decrease in the quantity of reagents used and a corresponding decrease in the quantity of waste generated.”

Jerry Foropoulos, Jr., NMT-6, is the principal researcher for this project.

Volatile Halocarbons Get the Treatment in a One-Step Process

Halogenated hydrocarbons or "halocarbons" are alien to our natural environment. They have varying effects on plant and animal metabolism, and little is known of their ultimate effect on biological life in general. This lack of knowledge is partly because halocarbons have been used in quantity only during this century, most notably since World War II. Virtually all halocarbons are man-made. They have many applications where their unique chemical and physical properties are desirable: lack of flammability and chemical reactivity, high density, and volatility.

Halocarbons are the basis of remarkably strong and inert plastics; Teflon® and polyvinyl chloride are two familiar examples. Another type of halocarbon, the chlorofluorocarbons (CFCs), were essentially an outgrowth of our early nuclear industry. Materials were sought that were inert to uranium hexafluoride, and wartime research and production led to the manufacture of large quantities of CFC-based plastics and refrigerants. Highly volatile and quite chemically inert, CFCs were expected to last indefinitely once produced, and these properties were to eventually cause the difficulties of cleaning up CFC-contaminated materials.

Most halocarbons originate from reaction of a halogen (chlorine, bromine, fluorine) and a hydrocarbon. Industrially, chlorine and bromine are obtained by the electrolysis of brine. The most reactive halogen, fluorine, is obtained from reacting fluorine-containing minerals with sulfuric acid. The resulting hydrogen fluoride (HF) is then used "as is" or electrolyzed to produce fluorine. Chlorine reacts with hydrocarbons to form chlorocarbons and hydrogen chloride (HCl). Totally chlorinated hydrocarbons are common starting species for CFCs. For example, carbon tetrachloride (CCl₄) reacts with pure HF in the presence of a catalyst to produce refrigerant-11 and refrigerant-12.

Today, many halocarbons are listed as hazardous or as ozone-depleting compounds. Biologically, several chlorocarbons are suspect carcinogens. CFCs supply chlorine atoms to the stratosphere, and such atoms have been shown to disrupt the ozone production cycle. Moreover, chlorinated hydrocarbons have contaminated thousands of sites both above and below ground throughout the world.

The general chemical inertness of these compounds precludes an easy solution to their cleanup and destruction.

In nuclear technology, the presence of these compounds in nuclear waste classifies the entire lot as "mixed waste" (waste containing both radioactive and hazardous materials, e.g., plutonium and CCl₄). The work described in this article presents a simple solution to the halocarbon destruction problem and promises many attractive advantages in dealing with mixed wastes.

At TA-55 and other places, soda lime has been used to scrub fluorine gas from the fluorination systems. Similar scrubbing of chlorine, carbon dioxide, HCl, and phosgene has also been done by soda lime. Prior research in plutonium chlorination revealed that soda lime, when added to a chlorination flow loop using CCl₄, apparently trapped or destroyed the CCl₄ along with the acid gases. Later, when heat was applied to a similar soda lime reactor, CCl₄ was completely and reproducibly destroyed. Subsequently, other compounds similar to CCl₄ were successfully destroyed in this fashion. As experiments progressed, it appeared that CFCs were also destroyed completely by this method at temperatures somewhat higher than those for the chlorocarbons.

In all cases, the reactions are exothermic. If the halocarbon vapor is sufficiently concentrated and is delivered constantly, the reaction self-sustains. Reactor core temperatures rose to 600°C during runs with vapor that was continuously fed into the reactor. This phenomenon is very controllable and can maximize process efficiency by using the latent heat produced.

The advantage of this process is particularly noteworthy in the nuclear industry; it has great potential in destroying the hazardous components of mixed waste. Halocarbons are particularly tough to remediate because of their resistance to conventional means of treatment. For example, burning or incineration of halocarbons is difficult to perform totally, and it is necessary to scrub the off-gases of the acids produced, such as HCl and HF. Thermal desorption of the hazardous component is the preferred method to use in tandem with the soda-lime-based system.

The destruction of halocarbons in a soda lime system may be applied in many industries: automotive, aviation, semiconductor, dry cleaning, refrigeration, waste treatment, and environmental restoration. The system can not only destroy halocarbons from liquid inventories and hazardous waste forms, it can be used as a scrubber for off-gases from semiconductor etching and cleaning and for destroying residual gases from dry cleaning and refrigeration systems. For waste solvents and the like, the soda lime system may treat vapors directly from a storage vessel. There is a patent pending for this destruction system, and an industrial partner is being sought to complete development and marketing.

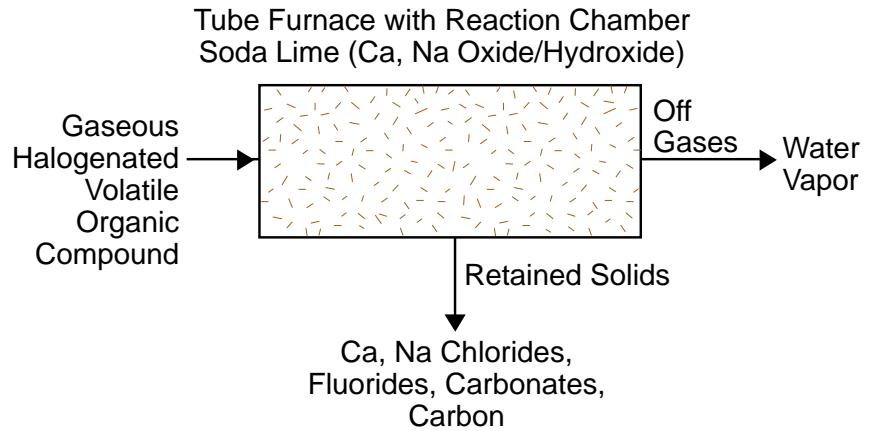


Figure 4. The system uses soda lime to convert hazardous halocarbons to nonhazardous gases and solids.

The halocarbon species listed here react with soda lime to form solids (carbon, calcium halides, calcium carbonates) and volatile products (water, carbon monoxide, and methane). The reaction mechanisms are not yet fully understood; however, the *net* result is formation of solid products and mostly H₂O.

halocarbon
carbon tetrachloride

trichloromethane
dichloromethane
trichloroethene, TCE

tetrachloroethene
1,1,1-trichloroethane, TCA

dichlorodifluoromethane

trichlorofluoromethane

1,1,2-trichlorotrifluoroethane

Researchers Perform Rapid, On-Line, Elemental Analysis Using Laser-Induced Plasma Spectroscopy

Pamela K. Benicewicz of NMT-6 is the principal investigator of this project. Additional contributors to this work include **Thomas K. Gamble** of CST-1 and **Vicente D. Sandoval, James T. McFarlan,** and **D. Kirk Veirs** of NMT-6.

Introduction

As a result of increased regulations, DOE orders, and the Laboratory's own commitment to safeguarding the environment, scientists are increasingly being tasked with developing and modifying their techniques and procedures in order to reduce the amount of waste they generate. We are developing an analytical technique, based on optical emission spectroscopy, that promotes this goal of waste minimization and can be used *in situ* to provide a rapid determination of the elements in actinide-containing materials.

Atomic emission spectroscopy of laser-induced plasmas can be used to detect elements analytically, at ppm concentrations, in harsh or radioactive environments. With this technique a laser beam is focused onto the surface of a sample. A tiny portion of the sample is vaporized, and a high-temperature plasma is formed. Spectral analysis of the plasma light shows the elemental content of the original sample. Because the constituents of the laser-created plasma are not constant in time,

time-resolved spectral analysis, obtaining spectra of the plasma only during a specified time period, allows one to discern atomic emissions from ionic emissions. We can further characterize the laser-generated plasma by studying its spatial evolution. The study is accomplished using a gated, two-dimensional detector, which allows the time-resolved spectroscopic image of the plasma plume to be captured. Because the f-elements, such as actinides, have unique physical properties, we expect that the temporal and spatial evolution of f-element plasmas will differ significantly from the temporal and spatial evolution of other elements, such as the transition metals. The goal is to use this technique for the analysis of trace amounts of actinides in a variety of actinide-containing materials.

This particular method of actinide and hazardous material analysis has significant advantages over other, well-established analytical techniques. It is a fairly straightforward method that requires little or no chemical preparation of the sample, and only a very small amount of material, on the order of nanogram or microgram quantities, is required for analysis. Thus, this technique approaches the ideal from the perspective of waste minimization. Measurements can be carried out rapidly and on-line, thereby avoiding the long lag times associated with some other analytical techniques. In fact, elements can be identified in minutes once a reference database has been established and calibrations have been made. Because the laser beam is focused to a small spot ($1\mu\text{m}$ – $50\mu\text{m}$), the technique provides spatial resolution of the sample composition for the evaluation of inhomogeneities. This method can also be adapted for remote analysis, which is particularly useful in radioactive environments.

Although laser-induced plasma spectroscopy has been in use for decades, recent improvements in the instrumentation supporting this technique have been substantial. The pulse-to-pulse stability of the output of current lasers is much better than the output of lasers made only a few years ago, resulting in greater accuracy and reproducibility of analytical

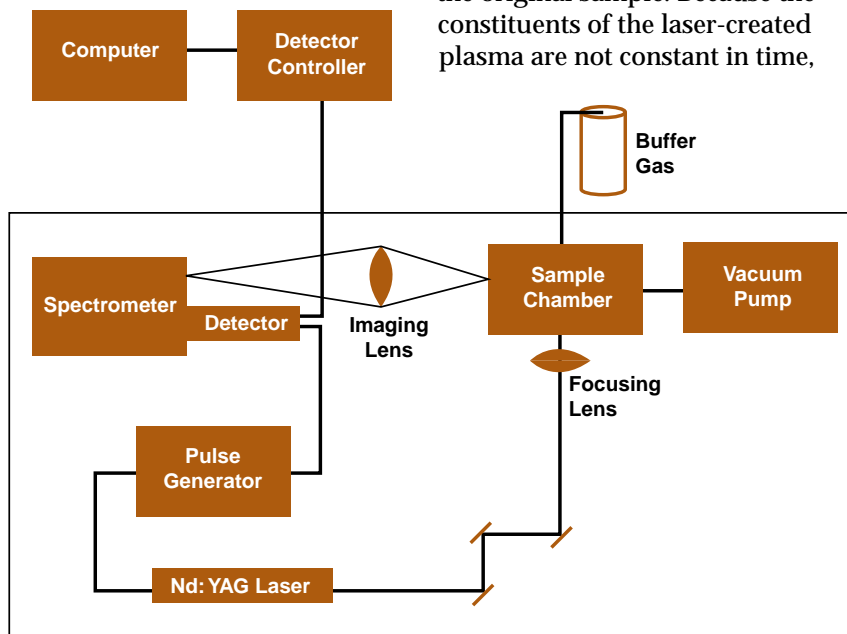


Figure 5. A schematic diagram of the experimental setup used for laser-induced plasma spectroscopy. Atomic emission spectroscopy of laser-induced plasmas can be used to analyze small samples on-line for elements at ppm concentrations in harsh or radioactive environments, even remotely.

results. Detector advancements are also contributing to the improvements in this technique. Data collection can be accomplished using a personal computer, and mathematical software is available for rapid analysis of the data and manipulation of the spectra. Finally, very compact lasers now produce the pulse energy required for this technique. Thus, it is feasible to develop portable instrumentation to detect and analyze actinides and hazardous materials on-site.

Results to Date

Experiments were conducted to study the time-resolved emission spectra and images of laser-generated plasma plumes from a target of gadolinium (Gd). We chose Gd for our experiments because many of its physical properties (e.g., ionization potential, vapor pressure, heat of vaporization, etc.) that are important to the processes of laser ablation and plasma formation and evolution are similar to those of plutonium (Pu). The objective of this work was to determine the initial conditions to be used for future experiments with Pu by optimizing signal detection and element identification by varying laser energy, buffer gas, and buffer gas pressure. The ultimate goal of our research is to optimize this method to detect trace amounts of actinide elements and determine relative actinide concentrations.

A schematic of the experimental setup is given in Figure 5. A 0.25-in. Gd rod containing known quantities of trace elements (200 ppm Tb, 50 ppm Zr, 20 ppm Cu, 20 ppm Cr, 10 ppm Ca and 1 ppm–3 ppm Al, Fe, Si, Mn and Mg) was housed in a sample chamber, which was evacuated and backfilled with a flowing buffer gas. The 1.064-mm, 5-ns output of a Nd:YAG laser, having 15 mJ/pulse was focused onto the Gd target by a single lens. The plume, formed at a right angle to the target, was imaged onto the front entrance slit of the spectrometer. The results of single-shot spectra and images taken of the plasma plume show that both temporal and spatial evolution are highly dependent on buffer gas and buffer gas pressure. Ar rather than He enhances atomic emissions in the red wavelength range. Three different buffer gases (Ar, He, and N) at three

different pressures (100, 300, and 500 torr) were compared. The intensity of the atomic Gd emissions in He was over a factor of 30 less than the intensity observed in Ar. Although the intensity of the Gd emissions in Ar is greater than the intensity in N at all

three pressures, at 500 torr the intensity in both gases is essentially the same. Because air is composed primarily of nitrogen and because 500 torr is approaching atmospheric pressure (at Los Alamos), it is reasonable to suggest that these experiments can be performed successfully in air with no special control of the pressure.

We also wanted to determine whether any of the trace elements present in the Gd rod could be detected. Figure 6 is a spectrum obtained using a 5 μ s integration time 4 μ s after the laser hit the target. All of the known Gd lines published in the National Bureau of Standards (now National Institute of Standards and Technology) wavelength tables in the wavelength range of 792 nm to 812 nm were observed, as indicated in the figure. Unknown peaks above 2000 most likely represent Gd emissions. Both strong Cu I emissions that occur in this wavelength range were observed, suggesting detection limits of Cu of at least 20 ppm.

Future Directions

We are currently in the process of setting up our laser spectroscopy system inside the Plutonium Facility of Building PF-4. We will begin characterizing actinide materials by their atomic emissions from laser-induced plasmas in the near future, and emphasis will be on detecting trace amounts of actinide elements. In addition, we are developing a technique based on laser-induced fluorescence of laser-created plasmas that uses a second, tunable solid-state laser to determine individual isotopes.

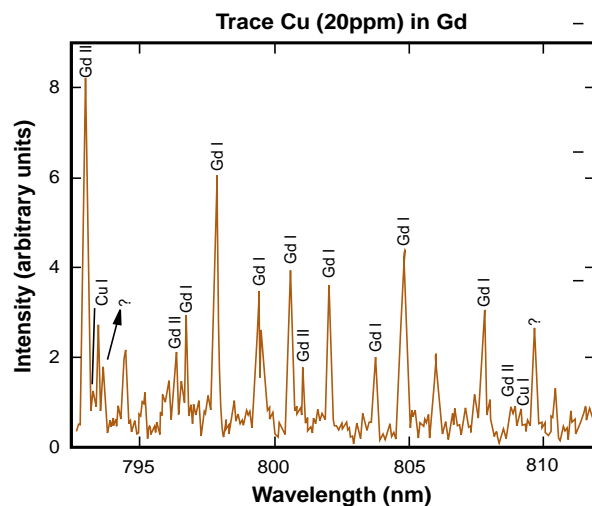
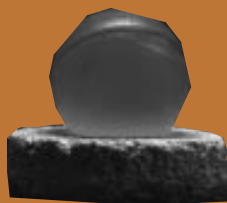


Figure 6. A spectrum obtained using a 5- μ s integration time 4 μ s after the laser hit the target. All of the known Gd lines were observed. The ultimate goal of the research is to optimize this method to detect trace amounts of actinide elements and determine relative actinide concentrations.



This article was contributed by **Laura Linford**.

NMT's Contributions to the Cassini Saturn Mission Follow Division's Space Exploration Tradition (*cont.*)

the heat of reentering the earth's atmosphere. Each RHU weighs only 40 grams and generates one watt of thermal energy through radioactive decay, operating at about body temperature (35°C–40°C). That may not seem like much, but it is a vast difference from the subzero temperatures of space.

Originally, Savannah River was to produce the GPHSs for the Cassini mission, but since their plutonium fuel forms facility was not operational, Los Alamos was selected by the DOE in 1990 to produce the heat sources. After performing the necessary operational readiness reviews, NMT Division began producing plutonium-238 heat sources in 1993. All 157 RHUs necessary for Cassini and the Huygens probe, plus 23 spares, were completed in about two years. Just recently, the 216 GPHSs were also completed. During the last few months of production, workers were on the project seven days a week and were able to complete approximately 25 GPHSs in one month. NMT-9 will make from 20 to 30 more GPHSs for spares, safety testing, etc. In July GPHSs were sent to EG&G/Mound Technologies, where they will be installed on the three RTGs for the Cassini orbiter. The RHUs will be shipped directly to the Kennedy Space Center in April 1997.

A large group of scientists, engineers, and technicians has been working on this project. Beside the 45 people dedicated to the project from NMT-9, many others throughout NMT and other Laboratory divisions have contributed to the effort. NMT-9 Group Leader **Tim George** and Deputy Group Leader **Liz Foltyn** have headed up the GPHS project, and **Gary Rinehart** is project leader for the LWRHU Program. **Egan McCormick** and **Mike Lopez** have been leading the fabrication effort of processing the PuO₂ and hot-pressing it into pellet form. **Bob Mathews** has been in charge of welding the special cladding and shipping containers for the heat sources. **Becky Guillen** leads the nondestructive safety testing of RHUs and GPHSs, such as checking for leakage and correct thermal output, **Mary Ann Reimus** oversees the nondestructive safety testing, and **Jim Jones** provides critical systems maintenance support.

Cassini will not be the first space mission for which Los Alamos has made heat and/or electricity sources. In the mid-70s, before TA-55 or NMT Division existed, Los Alamos developed and tested heat sources for Voyager I and II. The heat source used for these missions was known as the multi-hundred watt, or "MHW," which was about the size of a golf ball. The new RHUs and GPHSs reflect a move toward a more modular approach; these heat sources are more like small building blocks that can be used in varying quantities depending on specific needs. These newer heat sources (both RHUs and GPHSs) were designed here at TA-55 in the late 70s for the Galileo mission to Jupiter, and while the GPHSs were fabricated at Savannah River, the RHUs were made here. RTGs were also used in the Ulysses mission to orbit the poles of the sun.

The future holds still more opportunities for Los Alamos to contribute heat and energy sources to space missions. The upcoming Mars Pathfinder mission, set to launch in December of 1996, will be using three RHUs from TA-55 on a small rover that will explore the Mars surface, and a Pluto fast fly-by mission set to launch around the year 2000 will probably use around 50 RHUs, which NMT Division plans to produce. Los Alamos scientists and engineers from NMT and other divisions are also considering the possibilities of other energy sources beside the RTG that is standard today. Some of these possibilities include the conversion of PuO₂ heat to electricity through a dynamic engine with moving parts, such as a Brayton or Stirling motor. Such motors would greatly increase the efficiency of Pu energy sources (from about 7% efficiency to 20%), but the use of moving parts presents problems in the event of breakdown. Other possibilities include the use of infrared photovoltaic cells or alkali metal thermoelectric conversion.

The Cassini project is nearing its close at TA-55, but NMT scientists and engineers will continue to play their traditional role in space exploration, advancing mankind's efforts to understand our solar system and the universe beyond.

Publications, Presentations, and Reports (April 1996–June 1996)

Conference Presentations

The following papers were presented at The Combined 45th Annual X-Ray Conference and Powder Diffraction Satellite Meeting of the XVII Congress of the International Union of Crystallography, Denver, CO, August 3–8, 1996: L. D. Calvert (deceased), P. L. Wallace (LANL), T. C. Huang (IBM), J. A. Kaduk (AMOCO), J. N. Dann (Osram Sylvania), M. H. Mueller (ANL), and A. C. Roberts (Geological Survey of Canada), "Test Data for the Calculation of Powder Patterns for Intermetallic Phases"; J. N. Dann (Osram Sylvania), A. C. Roberts (Geological Survey of Canada), P. L. Wallace (LANL), and M. H. Mueller (ANL), "Use of Calvert's Test Data for the Evaluation of Powder Pattern Calculation Programs"; and L. D. Calvert, P. L. Wallace, T. C. Huang, J. A. Kaduk, J. N. Dann, M. H. Mueller, and A. C. Roberts, "Test Data for the Calculation of Powder Patterns for Intermetallic Phases," LA-UR-96-2547, published in *Advances in X-ray Analysis*, Vol. 40, 1996.

The following were presented at the American Glovebox Society Conference, San Diego, CA, July 22–25, 1996: T. O. Nelson, L. E. Bronisz, R. F. Hinde, J. Y. Huang, L. S. Kreyer, R. Laskie, H. E. Martinez, R. F. Meehan, P. Sayka, A. R. Schake, and L. H. Stapf, "Design of a Glovebox Conveyor System"; T. O. Nelson, D. A. Romero, J. Berg, D. Ford, J. Y. Huang, H. E. Martinez, W. R. Romero, A. R. Schake, L. H. Stapf, R. Vaughn, and D. E. Wedman, "Removal of Radioactive Material Containers from a Glovebox Using Electrolytic Decontamination"; D. W. Gray, S. M. Dinehart, W. D. Smyth, W. B. Smith, Bryan Howell, and G. Fisher, "Evaluation of an Alternative Adhesive System for Lined Gloveboxes Using Rare Earth Oxides to Enhance Radiation Shielding"; T. O. Nelson, H. E. Martinez, J. Y. Huang, R. F. Meehan, L. H. Stapf, W. G. Brough, and D. K. Dennison, "Retrofit of Existing Gloveboxes to Address Shielding Concerns"; M. A. Chavez, M. S. Palmer, W. B. Smith, and W. Smyth, "Expanded Roles for Lined Gloveboxes at LANL"; L. E. Bronisz, R. F. Hinde, J. Y. Huang, L. S. Kreyer, R. Laskie, H. E. Martinez, R. F. Meehan, T. O. Nelson, P. Sayka, L. H. Stapf, A. R. Schake, and D. K. Dennison (LLNL), "Design of a Glovebox Conveyor System"; and V. D. Sandoval, Y. M. Rivera, M. A. Martinez, R. A. Fernandez, and S. Pierce, "Methods for Leak Testing a Helium Atmosphere Glovebox."

The following were presented to the American Chemical Society, I&EC Division Special Symposium, Birmingham, Alabama, September 9–11, 1996: K. K. S. Pillay, "Waste Minimization from Plutonium Processing"; and L. R. Avens and K. K. S. Pillay, "Plutonium: It's Past, Present, and Future."

D. E. Wedman, T. O. Nelson, and K. R. Weisbrod, "Some Chemistry of the Electrolytic Decontamination process," the 212th National Meeting of the American Chemical Society, Orlando, Florida, August 25–29, 1996.

J. R. Hurd, F. Hsue, and P. M. Rinard, "Shuffler Measurements of Previously Unverified and Unconfirmed Inventory Items," LA-UR-96-341, 37th Annual Meeting of the Institute of Nuclear Materials management (INMM), Naples, FL, July 28–31, 1996.

L. R. Avens, D. D. Padilla, L. W. Worl, F. C. Prenger, and D. D. Hill, "Use of High Gradient Magnetic Separation for Actinide Applications," NATO Advanced Study Institute on Actinides and the Environment, Crete, Greece, July 7–19, 1996.

L. Morales and J. M. Haschke, "Investigation of the Plutonium Oxide-Water Reaction," High Temperature Chemistry Gordon Research Conference, Tilden School, New Hampshire, July 21–26, 1996.

M. A. Williamson, "Accelerator-Driven Transmutation of Waste: Should High Temperature Scientists Be Interested," Gordon Research Conference: High Temperature Chemistry, Tilton, NH, July 21–26, 1996.

T. O. Nelson, D. E. Wedman, and H. E. Martinez, "Electrolytic Methods for the Decontamination of Metal Surfaces," 23rd national Meeting of the Federation of Analytical Chemistry and Spectroscopy Societies (FACCS), Kansas City, Missouri, Sept. 29–Oct. 4, 1996.

L. A. Foster, D. G. Langner, and L. O. Ticknor, "Performance of the waste Crate Assay System at the Los Alamos National Laboratory Plutonium Facility," *Proceedings of the Institute of Nuclear Materials Management Conference*, Naples, FL, July 1996.

Reports

J. M. Macdonald, H. L. Nekimken, R. E. Hermes, J. M. Castro, M. E. Evans, and J. D. Olivas, "A New Glove for Glove Box Workers," Los Alamos National Laboratory report LA-UR-96-2366 (August 1996).

"94-1 Research and Development Project Lead Laboratory Support Status Report, April 1–June 30, 1996," (compiled by Mark Dinehart).

Journal Publications

B. Cort and P. G. Klemens, "Thermodynamic Properties of Helium Bubbles in Aged Plutonium," submitted to *J. Alloys and Compounds*, July 1996.

New NMT Web Pages



Look on the Los Alamos National Laboratory Web pages for issues of Actinide Research Quarterly and the new NMT Home Page! Click on Info by Organization, then on Divisions, then Nuclear Materials

Technology Division, or go directly to <http://www.lanl.gov/Internal/organizations/divisions/NMT/nmtdo>. Look for many new links to division research in the future, and if you have any questions or any suggestions for these pages, please e-mail kck@lanl.gov (K. C. Kim).

NewsMakers

■ Thirteen NMT researchers received awards for their distinguished contributions of noteworthy publications, patents, technology transfers, major program developments, successful research proposals, or other technological innovations. The NMT Division's Science and Technology Award Program is administered as part of the Los Alamos Award Program, approved by the DOE on a two-year trial basis, to recognize exceptional employee contributions and achievements. This year \$4700 from NMT Division's \$16,700 awards program fund was earmarked for the science and technology awards.

Three individual award winners are **John M Haschke** (NMT-5) for his publications and study of long-term storage of nuclear materials, **Steven M. "Mark" Dinehart** (NMT-6) for significant program development, and **Jerry Foropoulos** (NMT-6) for his new, innovative technology to treat volatile halocarbons (see article on page 6).

There were three team awards: The salt distillation team members (NMT-2) are **Vonda R. Dole, Eduardo Garcia, Walter J. Griego, and James A. McNeese** for demonstrating waste-minimizing, innovative technology that can partition plutonium and clean salts. The team working on new separations technology development (NMT-6) includes members **Gordon D. Jarvinen, Mary E. Barr, and Geraldine M. Purdy** for developing selective extraction of actinides and novel anion exchange resins and polymer filtration. The third team (NMT-6) award was given to **Arthur N. Morgan, Mary Esther Lucero, and Timothy O. Nelson** for their work in electrolytic decontamination.

■ **Karen W. Hench** (NMT-4) has received her Ph.D. in Industrial Engineering from New Mexico State University. Her dissertation was entitled "Modeling Fabrication of Nuclear Components: An Integrative Approach." It presents a two-stage approach to modeling the TA-55 foundry, one stage for optimizing foundry layout for reduced personnel exposures and the second for assessing the impact of different layouts on operational performance. The work was conducted under the supervision of **David Olivas** (NMT-5).

Plutonium Futures The Science

Plutonium Futures—The Science, a three-day topical conference on plutonium and the actinides is well into the planning stages for next August 25–27, 1997. It is sponsored by the Laboratory in cooperation with the American Nuclear Society. The conference will address issues of national and international importance including the safe storage and ultimate disposal of surplus weapons materials and the management of large inventories of actinides from civilian nuclear power generation. The focus of the conference will be on the technical knowledge base necessary for addressing these issues. The conference will be

held at the Santa Fe Hilton. For more information, please send e-mail to puconf97@lanl.gov or sign on to the conference Web page at <http://www.lanl.gov/PuConf97/Welcome.html>. This page is updated frequently with information about the conference.

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