



The Actinide Research Quarterly

of the Nuclear Materials Technology Division

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Researcher Offers a Technical Perspective on Plutonium in the Environment

Introduction

Plutonium is often inaccurately identified in the media as the “most toxic substance known to man.” Indeed, the element and its compounds are hazardous, and measures must be employed to protect workers, the public, and the environment. Minimizing the potential for release of plutonium is a primary concern at TA-55. Plutonium already in the environment, however, receives comparatively little attention.

An accurate assessment of the hazard posed by environmental plutonium is difficult, but substantial advances in this assessment are reported. Areas of investigation include determining the amount of environmental plutonium, describing its distribution and



Figure 1: Plutonium in the environment takes a number of forms and can be distributed in a number of ways, depending on the particle size and the dispersal mechanism.

migration, and evaluating its biological and health consequences. These topics are of increasing interest as a result of the expanding use of plutonium in mixed oxide fuels for power generation in other countries. This article attempts to summarize important aspects of the subject for readers of the *Actinide Research Quarterly*.

Quantity, Sources, and Distribution of Environmental Plutonium

Plutonium occurs naturally as a result of neutron capture and fission of uranium in pitchblende ores. The resulting plutonium concentration, about 5×10^{-12} of ^{239}Pu per gram of uranium, constitutes a negligible source of environmental plutonium.

Estimates place the total amount of man-made plutonium in the environment at 4.3 metric tons. This quantity results primarily from atmospheric testing of nuclear weapons and to lesser extents from reprocessing of nuclear fuel and destruction of thermoelectric generators from satellites reentering the atmosphere. The total alpha activity of ^{239}Pu from

aboveground nuclear testing is estimated at 7400 tera (10^{12}) Becquerels (TBq—see box) and accounts for about 3.3 metric tons of plutonium. The total alpha activity resulting from ^{238}Pu in the environment is about 1200 TBq, or about 2 kilograms of plutonium. Approximately 92% of environmental Pu is attributed to atmospheric testing.

A Curie (Ci) is a unit of radioactivity. One Ci corresponds to 3.7×10^{10} disintegrations per second. Because of the differences in half-lives of various radioactive elements, a Ci represents different amounts for different radioactive elements. For example, one Ci amounts to radioactivity resulting from about 16.3 grams of ^{239}Pu with a half-life of about 24,000 years, or about 0.1 milligram of tritium, which has a much shorter half-life. The author uses Becquerel units (Bq); one Bq is equal to 2.7×10^{-11} Ci or about 4.4×10^{-10} grams of ^{239}Pu (.44 billionths of a gram).

“Approximately 92% of environmental Pu is attributed to atmospheric testing.”

Relatively small amounts of plutonium result from accidents. About 15 kilograms (90 TBq) of plutonium was released from the reactor at Chernobyl. The contribution from military sources is even smaller. For example, the aircraft accident involving nuclear weapons near Thule, Greenland, in 1968 released about 0.9 TBq of alpha activity from plutonium.

The rate of plutonium deposition in the environment has varied substantially over the past fifty years. The largest rates were during the period of atmospheric testing in the 1950s and early 1960s. Releases from reprocessing facilities reached a maximum estimated rate of 70 TBq per year during the mid 1970s, but are currently at about 0.1 TBq per year as a result of improved facilities and procedures. A major concern for reprocessing and storage facilities is the potential catastrophic loss of containment and a high, localized release of material.

Global distribution of plutonium is inhomogeneous. Concentrations are high at mid-latitude zones of each hemisphere and are highest in the northern hemisphere, where most atmospheric nuclear tests were conducted. Maximum ^{239}Pu activities (70 Bq–80 Bq per square meter) appear at 35° – 45° north latitude. Activities of the isotope are about 15 Bq per square meter at mid latitudes of the southern hemisphere and are 1 Bq–10 Bq per square meter near the equator and poles.

Behavior of Environmental Plutonium

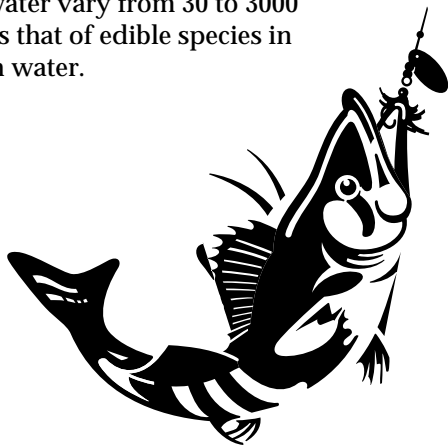
Studies show that Pu exists primarily as an oxide in land deposits and in ocean sediments. Behavior in the environment is strongly dependent on the physical and chemical conditions of both the material and the medium. Important properties of the oxide are particle size and solubility. Plutonium appears in water as ionic species of Pu(IV) and Pu(VI).

When plutonium is released into the atmosphere, its behavior depends on the particle size and the dispersal mechanism. Stratospheric aerosols formed by nuclear testing and satellite burnup distribute globally over a period of years; material released by accidents typically deposit locally within minutes or hours. Airborne redistribution of potentially dispersible particles with geometric diameters less than $10 \mu\text{m}$ is unlikely because such small particles readily adhere to surfaces of large soil particles.

Processes for translocation and redistribution of environmental plutonium are both mechanical and chemical. Vertical transport in soil is slow compared to lateral redistribution by processes such as cultivation and erosion by wind and water currents. Other mobilization mechanisms, including biological

transport, depend on the solubility of the plutonium in water. In water, distribution constants, that is the fraction of the total Pu dissolved, are in the 10^{-4} – 10^{-5} range, showing that plutonium is an insoluble solid with a solubility similar to that of glass (SiO_2).

Chemical uptake by biological systems occurs via several pathways. The plutonium fraction transferred by root uptake of plants ranges from 10^{-3} to 10^{-5} . Particle inhalation by grazing animals is of negligible concern compared to their gastrointestinal uptake. The fraction of ingested plutonium absorbed is approximately 10^{-4} , and the combination of the plant uptake and animal ingestion from plant sources shows that the fraction of deposited Pu translocated to herbivores is 10^{-7} to 10^{-9} . Behavior in marine and fresh water systems is similar; however, Pu concentrations in edible species (e.g., fish and crustaceans) in seawater vary from 30 to 3000 times that of edible species in fresh water.



Human Effects of Environmental Plutonium

As with animals, incorporation of plutonium by humans also occurs primarily by inhalation and ingestion. Inhalation is of concern only in instances of accidental release where local concentrations are high for a short time. Studies of human populations give uptake fractions of 10^{-4} to 10^{-6} for ingested Pu.

The effects of plutonium on human health and longevity are the primary concern. Biologically, Pu is classified as a radiotoxin. The risk of cancer death is estimated to increase by 0.2% (2 in 1,000) for a person who breathes highly contaminated air ($0.1 \mu\text{g}$ of respirable oxide per cubic meter) for one hour. Homogeneous dispersal of one kilogram of oxide in a typical municipal water supply would result in a Pu concentration of about 1 nanogram of Pu per liter. The increased risk of cancer death for a person who drinks two liters of that water per day for seventy years is estimated to be 0.01% (1 in 10,000).

Relevant Studies

Although the mission of TA-55 does not include studies on environmental Pu, work being conducted under the Plutonium Repackaging Program is directly related to hazard assessments for incidents involving both facilities and weapons. Unlike Pu metal, PuO_2 is a powdered material with potential for environmental dispersal. Studies are underway to measure the size distributions for oxides from different sources and define the mass fractions of dispersible ($< 10 \mu\text{m}$ geometric diameter) and respirable ($< 3 \mu\text{m}$ geometric diameter) particles.

Results show that the dispersible and respirable fractions of the oxide vary by a factor of about 10^4 depending on the method of preparation. A credible assessment of the dispersal hazard is possible only if the oxide source is considered.

This article is based on "Plutonium in the Environment," LA-UR-96-1261, by John Haschke, in the Safety Series Document, "Safe Handling and Storage of Plutonium," Chapter 5, International Atomic Energy Agency, April 1996.

R.L. Gutierrez, NMT-DO, and M.F. Stevens, Center for Materials Science, contributed this article.

“Plutonium materials science research at Los Alamos now requires renewed commitment and funding to support enhanced stockpile surveillance, recertification of weapons, and replacement manufacturing.”

Plutonium Materials Science Supports Science-Based Stockpile Stewardship and Management

Cessation of nuclear testing and the end of the Cold War caused the DOE weapons design laboratories to rethink their policies and practices for maintaining expertise in nuclear weapons science and for ensuring the integrity and safety of the increasingly smaller nuclear arsenal. In particular, weapons were originally intended for a stockpile life of approximately 25 years, but current knowledge and data indicate that the weapon life cycle may be extended beyond 25 years before the degradation of materials affects their performance.

Extending the stockpile life of existing nuclear weapons requires knowledge of the effects of aging on plutonium. In the past it was possible to remove a unit from the stockpile and test it in an underground explosion to measure any aging effects directly. Thus, much less knowledge of the specific materials and their influence on weapon performance was required. Without the advantage of underground weapons testing, we must now study the material itself.

Within the weapon pit component, clearly, it is the plutonium that causes aging-induced changes. The other components consist of relatively inert and otherwise well-protected materials. The materials-science-related phenomena that are likely to create changes in the plutonium can be categorized into three major areas: decay, equation-of-state, and high chemical reactivity.

Plutonium is radioactive (the 239 isotope has a half-life of approximately 24,000 years) and decays by emitting alpha radiation. Internally trapped alpha particles evolve into helium atoms, which can accumulate to about a thousand atomic parts per million over a 20- to 30-year time frame. Helium is known to cause embrittlement and swelling of metals such as stainless steel. The formation of such helium bubbles could affect the structural stability of the plutonium.

Another event that accompanies the ingrowth of helium during alpha decay is the injection of a uranium atom. Besides the possible chemical effects of the uranium atom on phase stability and physical properties, the recoil part of the nuclear decay process occurs at a high energy level, during which, it is conjectured, the uranium atom displaces many plutonium atoms before coming to rest. The defects thus produced may result in altering the chemical and physical stability of the plutonium.

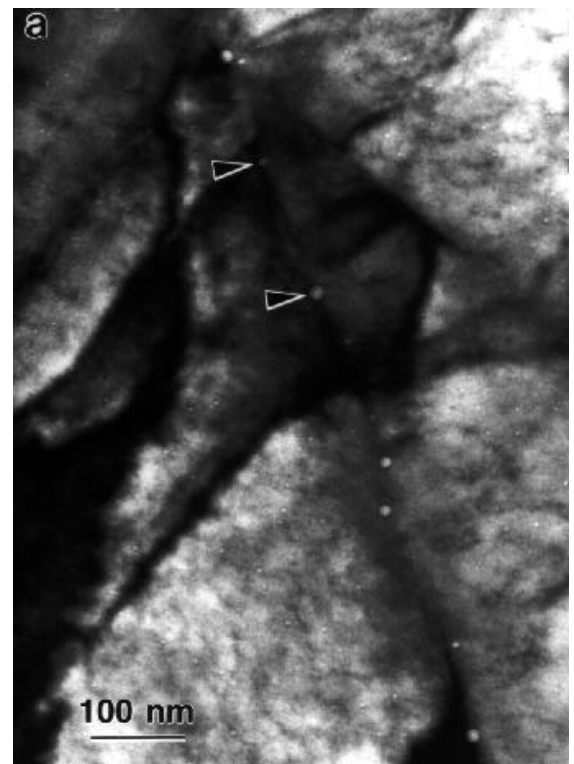


Figure 2: Helium bubbles are shown in 20-year-old plutonium. Helium generation affects plutonium properties in weapon components.

Another measurable radiolytic process involves the decay of residual ^{241}Pu , which results in the ingrowth of americium in the metal matrix. Americium may affect plutonium phase stability, but this phenomenon has not been studied conclusively.

Plutonium research efforts at Los Alamos have accomplished many noteworthy goals, such as determining many known plutonium binary phase diagrams, thorough study of the physical and mechanical properties of the stable metallurgical phases, and development of the “tailwind” and “trunk” alloys, now standards in the stockpile. In the middle to late 1980s, new techniques such as transmission electron microscopy and neutron scattering were applied to plutonium, resulting in detailed studies of the crystallography and structure of the $\delta \rightarrow \alpha$ martensitic phase transformation, the first observation of helium bubbles in aged plutonium (Figure 2), and the determination of Debye-Waller factors for both α - and δ -phase alloys. In the last few years small efforts have resulted in new technique breakthroughs, such as the advent of ultraviolet photoelectron spectroscopy, the laser-induced miniflyer technique (shock compression), and the extended x-ray absorption fine structure technique, which hold even further hope for understanding and predicting the behavior of plutonium under storage conditions. Such studies are important for thorough stockpile stewardship.

Several well-defined requirements within the new Stockpile Stewardship and Management Program necessitate renewed plutonium materials science activity. These requirements include recertification of existing weapons, a replacement-level pit manufacturing capability, and enhanced surveillance. Plutonium materials science research at Los Alamos now requires renewed commitment and funding to support enhanced stockpile surveillance, recertification of weapons, and replacement manufacturing.

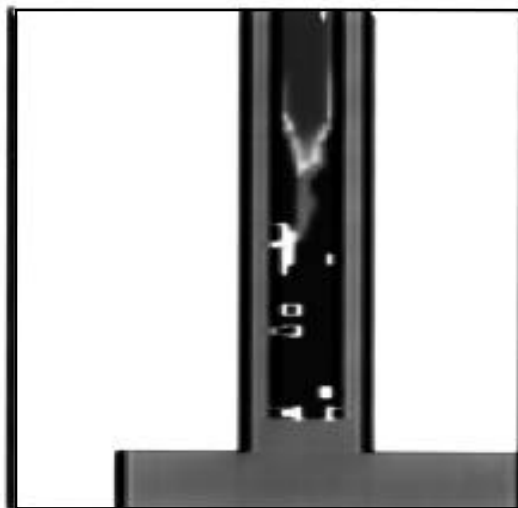


Figure 3. Computer simulation of plutonium casting. Such simulations are used before plutonium parts are cast to study the behavior of the material in the casting process and to avoid defects.

Specific plutonium research topics at Los Alamos that are underway or proposed are divided into four major areas:

- Characterization of the effect of radiolytic helium on plutonium properties,
- Effect of aging on phase stability and density of plutonium,
- Surface physics and chemistry of plutonium metal, and
- Dynamic studies (shock compression) of plutonium and other weapons and materials.

Experiments will require close interaction by the major participating divisions within the Laboratory. We suggest that a committee chaired by a major funding program manager and a representative from each of the participating divisions coordinate this enhanced stockpile surveillance effort.

Division Director Discusses Plutonium Future *Part 2*

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

Presently there is no international consensus on the disposition of excess plutonium released from weapons dismantlement. An approach that is being considered by most nuclear states, but not by the U.S. at this time, is utilizing plutonium as fuel to extract the available energy. In this context, weapons plutonium can serve as an initial step towards the recycling of plutonium for use in the civilian fuel cycle. There are no *technical* barriers to this option, but a myriad of timing, political, and fiscal barriers exist. Some of the world's weapons materials are currently vulnerable to diversion, and because of the urgency to reduce the potential of nuclear threats, we can

ill afford to wait for the political debate. Current U.S. policy is not to reprocess and recycle plutonium in commercial reactors, and the financial incentives to burn plutonium in nuclear power reactors are lacking in this, the world's most energy-intensive nation.

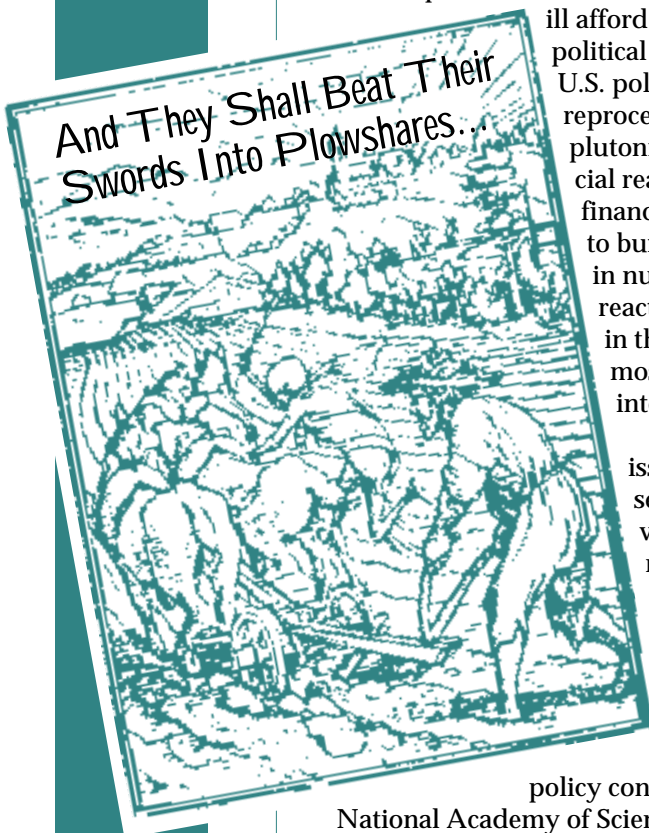
The urgent issue is how to secure excess weapons plutonium against potential theft and diversion while the international debate on disposition

policy continues. The National Academy of Sciences has stated

that the excess plutonium in the world's stockpile represents a clear and present danger to national security. So the question is: How can we preserve this potentially valuable resource for the future while dealing today's realities? There is a logical appeal to the idea that the solution for dealing with excess plutonium from weapons has a parallel path to that for spent reactor fuel, and that the two paths converge at a decision point 50 years in the future.

The top-level tasks for the weapons plutonium project would be

1. dismantle weapons,
2. stabilize residues,
3. separate plutonium from weapons components and residues,
4. discard the low-level and nonnuclear wastes in permanent repositories,
5. convert the plutonium into inspectable forms of oxide or metal, and
6. store the plutonium in containers that can be certified and safeguarded for 50 or 100 years. Then let the political and economic debates rage and settle, and *assuming the conclusion is the peaceful use of the nuclear fuel cycle*,
7. convert the plutonium to mixed-oxide fuel,
8. recycle it in reactors, and eventually
9. reprocess and recycle plutonium in advanced reactor fuel cycles.



Technical solutions needed to complete this sequence have been developed as follows:

- disassembly, stabilization, and separation technologies for dismantled weapons and the 50-year legacy wastes are well established;
- disposal of nonnuclear or low-level nuclear waste is economically attractive and more environmentally acceptable than storing high-level radioactive wastes;
- the long-term stability of plutonium oxide and metal is well understood; and storage containers designed to last for 50 years have been fabricated and are in use.

Safeguarding the plutonium seems fairly simple because the amount of estimated excess weapons-grade plutonium metal could fit in two or three pickup trucks. (Of course the trucks would collapse, and the mass would be supercritical, but the image helps to visualize the magnitude of the problem.) One or two storage facilities containing 99.5 metric tons of separated plutonium could be readily safeguarded against theft and diversion for 50 years.

The actions for dealing with spent fuel are somewhat more complicated because of the profuse numbers and the dispersion of spent fuel assemblies around the nation and the globe. An additional 800 metric tons of reactor plutonium are contained—and still growing at about 50 tons per year—in spent nuclear reactor fuel. The spent nuclear fuel, along with about 90 metric tons of separated, reactor-grade plutonium, is presently stored in the 30 countries with civil nuclear power programs. Ironically, the intense radioactivity of spent fuel reduces the threat of its theft and diversion and therefore decreases the urgency of dealing with it. The parallel, top-level tasks for dealing with spent nuclear fuel might be

1. temporary storage in pools, followed by
2. retrievable dry storage in aboveground containers, thereby allowing time to debate the use of plutonium as a future energy source, and *assuming the conclusion is the peaceful use of the nuclear fuel cycle*,
3. reprocessing spent fuel,
4. discarding the fission product wastes in permanent repositories,
5. fabricating mixed-oxide fuel,
6. recycling the fuel in reactors, and
7. reprocessing and recycling plutonium in advanced reactors.

The energy content of the weapons plutonium in the U.S. has about the same energy content as 100,000,000 metric tons of oil; the energy content of spent fuel could increase the energy available from the once-through uranium cycle by nearly 100 times. This energy source could contribute to worldwide economic stability and reduce worldwide pollution from burning of fossil fuels. The barriers to achieving the vision of Isaiah with weapons-grade plutonium are significant but relatively simple: overcoming the aversion to separate, store, and use weapons-grade plutonium for peaceful uses, and reversing the restrictions on reprocessing and recycling reactor-grade plutonium.

Success depends on nations agreeing with nations on strict safeguarding and security of reactor-grade plutonium; I'm not optimistic, but I'm hopeful. In the meantime, the mission that we are facing is every bit as challenging, technically exciting, and important as the Manhattan Project. Whatever the political decisions and economic drivers; TA-55 will play a key role in developing new technologies for utilizing, managing, and ensuring the safety of the world's plutonium.

Bruce Matthews

The ideas in this editorial are not original; they are a synthesis from many national and international studies, reports, and publications and from conversations with today's prophets. The recommendations, however, are mine, they do not necessarily represent the opinion of Los Alamos National Laboratory, the University of California, the Department of Energy, or the U.S. Government.

Does the Interaction of Plutonium Oxide with Water Pose a Potential Storage Hazard?

Editor's note:

This article brings up an interesting debate. Independent reviewers of the article point out that other researchers have tried to produce PuO_{2+x} and higher oxides by similar methods using oxygen, ozone, nitrous oxide, and other strong oxidizing agents. No higher oxide has been produced.

Recent world events have resulted in downsized nuclear arsenals and focused attention on additional ways of reducing the nuclear danger. Proper disposal of the surplus weapons-grade plutonium from dismantled weapons is necessary to prevent its diversion and reuse in weapons by proliferant states or terrorists. Most nations plan to use plutonium from both commercial reactors and weapons for nuclear power generation; the United States is considering various alternatives. Since selection and completion of a disposal option will probably take many years, surplus plutonium must be stored safely for an extended interim period.

DOE standards are established for storing surplus plutonium as metal (Pu) or as dioxide (PuO_2). After appropriate processing and certification, these materials are doubly confined in leak-tight stainless steel containers. According to popular belief, oxide is a stable material and the preferred form for storing plutonium. However, storage of oxide is more complicated than storage of metal. If hydrocarbons such as oils and plastics are present with the stored plutonium, whether metal or plutonium oxide, they undergo radiolytic decomposition by alpha particles from the radioactive decay of the plutonium, and this decomposition generates hydrogen gas. Whereas metal consumes the hydrogen gas by forming plutonium hydride, hydrogen produced in an oxide container pressurizes the vessel and may lead ultimately to rupture and the release of plutonium.



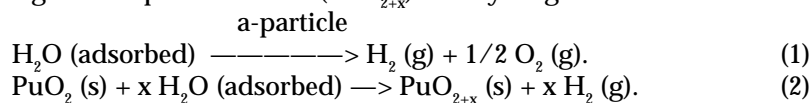
Figure 4. This failed storage container (left) containing Pu metal was packaged in air containing some moisture. Whereas one end bulged, the weld on the other end (right) failed and ruptured. The drastic failure occurred after just a few days.

The Problem

Plutonium dioxide typically exists as a fine powder, and various hydrogen-containing species adsorb on its surface. Although oxide is processed by firing in air at 950°C to remove water, hydrocarbon, and other species before storage, water readily re-adsorbs when the oxide is exposed to room air during packaging operations. The fate of adsorbed water during storage is a concern.

Does residual water on the oxide pose a potential storage hazard? The answer to that question is a qualified "No." If oxide is prepared, certified, and packaged according to procedures developed by NMT Division, the amount of residual water is far too small to cause problems. However, the situation would be different if oxide were to be packaged mistakenly without being fired.

Two processes by which adsorbed water might generate pressure in oxide-containing storage vessels are being investigated by members of the Applied Weapons R&D Team in NMT-5. As described by the following reactions, these processes are (1) radiolytic decomposition of adsorbed water into oxygen and hydrogen and (2) chemical reaction of dioxide and water to form a higher-composition oxide (PuO_{2+x}) and hydrogen:



Reaction 1 is similar to the radiolytic decomposition of hydrocarbon compounds noted above. Reaction 2 is suggested by early studies of plutonium chemistry and by recently

The work presented here implies that water is a better oxidizing agent than oxygen, which is difficult to prove. The question also remains whether the extra oxygen in PuO_{2+x} is actually adsorbed on the surface or incorporated into the lattice.

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published results showing that a PuO_{2+x} phase forms on the dioxide surface in water vapor at 250°C to 300°C. The product is a mixed-valence oxide of Pu (IV) and Pu (VI) with $x \leq .3$.

A number of questions remain to be answered:

- Does any significant reaction occur at room temperature?
- Does some other unanticipated reaction occur?
- How fast are these reactions?
- Are their rates fast enough to present a potential storage problem?
- Do O_2 and H_2 combine to form water?
- Is the recombination rate faster than the rate of radiolytic decomposition?

Experimental Approach

With so many questions to be answered, a large research effort might be anticipated. However, essential questions relevant to the storage issue are readily addressed by simple pressure-volume-temperature (PVT) experiments. After reactants are sealed in a vessel of known volume, the temperature and pressure are monitored over time. Reaction rates are defined by changes in pressure with time. The reaction is identified by analysis of the gaseous products.

Two PVT experiments were conducted using PuO_2 prepared by reacting high-purity Pu with oxygen. In one test the oxide-containing reaction vessel was evacuated and backfilled to the saturation pressure of liquid D_2O at room temperature. In the other test the vessel was backfilled with approximately 125 torr of a mixture with D_2 and O_2 in a 2:1 ratio.

Analytical results for the test with D_2O are the key to defining chemical behavior. Radiolytic decomposition, Reaction 1, is the important reaction if D_2 and O_2 appear in a 2:1 ratio. Likewise, the formation of pure D_2 shows that water reacts according to Reaction 2. Occurrence of both reactions produces a mixture with a hydrogen:oxygen ratio greater than 2.

The importance of radiolytic decomposition is most easily shown by comparing the pressure changes for the two tests. Radiolysis cannot result in pressurization of an oxide storage container if the oxygen and hydrogen recombine to form water (reverse of Reaction 1) at a faster rate than that of the forward reaction.

Results and Conclusions

Results of the test with D_2O show that adsorbed water reacts chemically with PuO_2 to form D_2 according to Reaction 2. The pressure increases at a constant rate over time, and hydrogen is the only gaseous product. At the measured rate a high hydrogen pressure can be produced in a few years in a typical storage container.

An explanation for the apparent absence of radiolytic decomposition is suggested by kinetic results for the D_2+O_2 mixture. As shown in Figure 5, the pressure in the mixture dropped rapidly during the first few days and gradually slowed to an almost constant decrease that has continued for more than a year. The rate at which hydrogen and oxygen combine on the catalytic surface of PuO_2 is so fast that radiolytic decomposition is not observed. Occurrence of Reaction 1 is not a concern for oxide storage. The $\text{D}_2:\text{O}_2$ ratio of the gas mixture is progressively increasing over time because the D_2O product is reacting with the oxide as described in Reaction 2.

The results demonstrate that excessive pressurization of a storage container might occur if the oxide were improperly fired or if unfired oxide were mistakenly packaged. Although all questions presented above have been answered, additional studies are being conducted to determine how the rate of Reaction 2 depends on temperature and on the concentration of water adsorbed on the oxide.

Spinoff

The importance of these findings extends far beyond their relevance to storage safety. Further study of the formation of PuO_{2+x} might help to explain why environmental plutonium appears as high-oxidation-state ions in solution and why high-fired oxide tends to be insoluble. Results of the oxygen-hydrogen combination study have already been used to explain why atmospheric corrosion of plutonium metal is accelerated by moisture.

This work was done by members of the Applied Weapons R&D Team led by **John Haschke**.

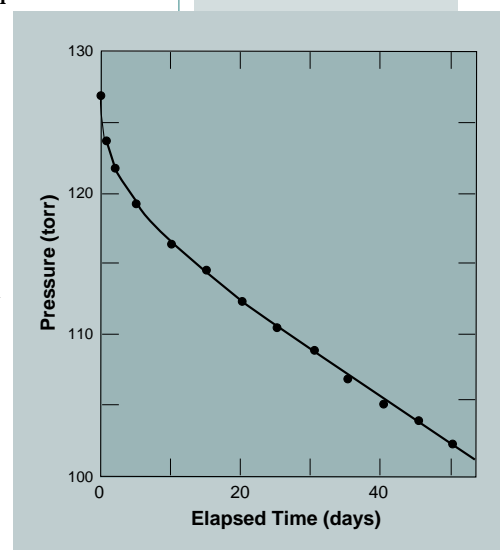


Figure 5. Time dependence of the D_2+O_2 pressure over plutonium oxide at 25°C. Pressure dropped rapidly the first few days and gradually slowed to a nearly constant decrease that has continued for more than a year.

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

Journal Publications

P. J. Allen, D. K. Veirs, S. D. Conradson, C. A. Smith, and S. F. Marsh, "Characterization of Aqueous Plutonium (IV) Nitrate Complexes by Extended X-ray Adsorption Fine Structure Spectroscopy," *Inorg. Chem.* **35**, 2841–2845, 1996.

R. N. Mulford and K. C. Kim, "Measurement and Analysis of the Fourier Transform Spectra of the v_3 Fundamental and $v_1 + v_3$ Combination of NpF_6 ," *J. Mol. Spectros.* **176**, 369–374, 1996.

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," *Advances in X-ray Analysis*, **40**, 1996.

J. M. Haschke and T. E. Ricketts, "Adsorption of Water on Plutonium Dioxide," LA-UR-96-1399, submitted to *J. Alloys and Compounds*, April 1996.

Invited Talks

R. B. Matthews, "The Plutonium Legacy," The University of California, Berkeley, CA, April 15, 1996.

D. C. Christensen, "The Future of Plutonium Science," The University of California, Davis, CA, May 20, 1996.

Conference Presentations

The following papers were presented to The 1996 American Nuclear Society Meeting, Reno, Nevada, June 16–20, 1996: D. E. Wedman and W. H. Smith, "Application of Electrodialysis to the Recovery of Spent Electrorefining Salts"; L. A. Worl, J. M. Berg, D. D. Padilla, S. M. Bowen, and M. Cisneros, "TRU Removal from Underground Storage Tank Waste"; K. K. S. Pillay, "Plutonium Management for the Future"; L. D. Schulte, S. D. McKee, and R. R. Salazar, "Application of Extraction Chromatography to Actinide Decontamination of Hydrochloric Acid Streams"; D. C. Christensen and R. B. Matthews, "Actinide Sciences in the Future of the United States Defense Complex"; B. F. Flamm and G. M. Isom, "Hydride Dehydride Recycle Process." L. D. Schulte, S. D. McKee, and R. R. Salazar, "Application of Extraction Chromatography to Actinide Decontamination of Hydrochloric Acid Effluent Streams."

S. M. Long and S. T. Hsue, "Design and Fabrication of Non-destructive Assay Standards," American Chemical Society, Nuclear and Isotopic Methods of Analysis for Safeguards and Security Symposium, New Orleans, LA, March 24–28, 1996.

P. K. Benicewicz and D. K. Veirs, "Characterization of Gadolinium via The Temporal and Spatial Evolution of Emissions From Laser-Created Plasmas," The Conference on Lasers and Electro-Optics (CLEOS '96), Anaheim, CA, June 2–7, 1996.

J. Foropoulos, Jr., "Solid Alkali Destruction of Volatile Waste Halocarbons," LA-UR 95-4302, Eighth National Technology Information Exchange Workshop, Santa Fe, NM, April 16–18, 1996.

N. G. Pope, W. J. Turner, R. E. Brown, R. A. Bibeau, R. R. Davis, and K. B. Hogan, "Upgrade of the Los Alamos Plutonium Facility Control System," LA-UR 96-534, Proceedings from the 1996 International Topical Meeting on Nuclear Plant Instrumentation, Control & Human Machine Interface Tech., Pennsylvania State University, May 5–9, 1996.

S. M. Dinehart, K. A. Qubat-Martin, D. W. Gray, V. A. Hatler, and S. W. Jones, "Radioactive Source Recovery Program: Responses to Neutron Source Emergencies," 28th Annual National Conference on Radiation Control, Albuquerque, NM, May 5–9, 1996, and Hazwaste World/Superfund XVII, Washington, D.C., October 15–17, 1996.

The following papers were presented at the Twentieth Compatibility, Aging and Stockpile Stewardship Conference, Allied Signal, Kansas City, MO, April 30–May 2, 1996: P. D. Kleinschmidt, J. J. Park, and B. R. Kniss, "Composition Changes in Plutonium Weapon Alloys as a Function of Time (U)," LA-CP-96-47; J. C. Martz and J. M. Haschke, "Oxidation, Dynamic Heating and Explosive Dispersal of Plutonium"; J. M. Haschke, T. H. Allen, and J. L. Stakebake, "Enhancement of Plutonium Corrosion in Moist Air."

D. G. Kolman, D. K. Ford, T. O. Nelson, and D. P. Butt, "General and Localized Corrosion Behavior of 304 Stainless Steel Exposed to Room and High Temperature Nitric Acid/Halide Solutions," NACE '97, Symposium on Corrosion Issues in Liquid Radioactive Waste Storage, New Orleans, LA, March 9–14, 1997.

The following papers were presented to The 1996 Actinide Separations Conference, Itasca, Illinois, June 11–13, 1996: L. D. Schulte, S. D. McKee, and R. R. Salazar, "Application of Extraction Chromatography to Actinide Decontamination of Hydrochloric Acid Streams"; T. O. Nelson, D. D. Hill, and H. E. Martinez, "The Use of Cryogrinding to Size Reduce Materials for Chemical Processing and Volume Reduction"; T. O. Nelson, "Nuclear Weapons Dismantlement for Fissile Material Disposition"; T. O. Nelson, A. N. Morgan, and M. E. Lucero, "Contamination Weeping Tests of Plutonium on Uranium Surfaces Using Alpha Spectroscopy"; D. G. Kolman, T. O. Nelson, D. K. Ford, and D. P. Butt, "Investigation of General and Localized Corrosion on Stainless Steel from Processing Simulated Pyrochemical Actinide Chloride Salts by Nitric Acid"; E. Garcia, J. A. McNeese, V. R. Dole, and W. J. Griego, "Pyrochemical Salt Distillation," LA-CP-96-110; L. Worl, D. Padilla, D. K. Veirs, S. Buelow, and J. Roberts, "Hydrothermal Oxidation for the Treatment of Combustibles"; L. A. Avens, K. K. S. Pillay, and S. M. Dinehart, "The 94-1 Core Technology Research and Development Program."

The following papers were presented at JOWOG 22 Pu Focused Exchange, Lawrence Livermore National Laboratory, May 6-7, 1996: P. D. Kleinschmidt and J. J. Park, "Composition Changes in Weapon Alloys as a Function of Time"; S. J. Hale, "Direct Into Shell Casting"; J. M. Haschke, "Moisture-Enhanced Corrosion of Plutonium"; B. Cort, A. Lawson, and J. A. Roberts, "Neutron Diffraction Studies of Plutonium Alloys," J. P. Baiardo, "Acoustic Resonance Spectroscopy in Surveillance"; L. E. Cox and S. D. Conradson, "EXAFS and X-Ray Diffraction of Pu Alloys"; A. C. Lawson, B. Cort, and J. A. Roberts, "Neutron Diffraction Studies of Plutonium Alloys," and S. J. Hale, "Bonding Techniques."

The following papers were presented at JOWOG 22 Conference on Actinide Stabilization, Packaging and Storage, Los Alamos National Laboratory, May 21-22, 1996: J. M. Haschke, "Discussion of Known Container Failure Mechanisms," "Plutonium Metal, Oxide and Residue Stabilization Parameters," "Pressurization and Condensation in Long-Term Storage Containers," and "Recent Findings."

Reports

F. N. Schonfeld and R. E. Tate, "The Thermal Expansion Behavior of Unalloyed Plutonium," Los Alamos National Laboratory report LA-13034-MS, June 1996.

M. H. Reimus, J. E. Hinckley, and T. G. George, "General-Purpose Heat Source: Research and Development Program; Radioisotope Thermoelectric Generator Impact Tests: RTG-1 and RTG-2," U.S. Department of Energy, Office of Special Applications, April 1996.

P. J. Rodriguez, "Characterization and Refinement of Carbide Coating Formation Rates and Dissolution Kinetics in the Ta-C System," Masters Thesis, The University of New Mexico, March 29, 1996.

C. A. Smith and T. R. Mills, "Hydrochloric Acid Recycle: Completion of Cold Test Phase for Acid Evaporator," SERDP (Strategic Environmental Research and Development Program) Interim Report, LA-UR-96-1226, March 1996.

J. M. Haschke, "Nuclear, Physical and Chemical Properties of Plutonium," LA-UR-96-1228, Safety Series Document, Safe Handling and Storage of Plutonium, Chapter 3, International Atomic Energy Agency, April 1996.

J. G. Watkin, D. L. Clark, G. Cloke (University of Sussex), B. W. Eichorn (University of Maryland), J. C. Gordon, S. K. Grumbine, R. L. Hollis, J. T. McFarlan, F. McNamara (Sandia National Laboratory), and B. D. Zwick, "Metal Vapor Synthesis in Organometallic Chemistry," Los Alamos National Laboratory report LA-UR-96-1006, April 1996.

E. M. Foltyn, "Monthly Progress Report: Heat Source Technology Programs, December 1995," U. S. Department of Energy, Space and National Security Programs NE50, April 1996.

S. M. Dinehart and N. A. Rink, "94-1 R&D Status Report, Second Quarter 1996," April 1996.

J. M. Haschke, D. R. Horrell, C. W. Hoth, K. W. Fife, S. W. Pierce, T. E. Ricketts, N. A. Rink, and M. A. Robinson, "Nuclear Material Stabilization and Packaging," Quarterly Status Report, January 1-March 31, 1996, May, 1996.

J. M. Haschke, "Plutonium in the Environment," LA-UR-96-1261, Safety Series Document, Safe Handling and Storage of Plutonium, Chapter 5, International Atomic Energy Agency, April 1996.

T. G. George, "Monthly Progress Report: Heat Source Technology Programs, May 1995.

J. E. Hinckley, F. M. Guerra, and R. L. Gonzales, "3-Dimensional Hydrocode Analysis of RTG Impact Tests (Abstract)," United States Department of Energy/Space and National Security Programs, Engineering and Technology Development, May 1996.

S. D. McKee, "Ash Trade Study Performance Factors for Los Alamos," DOE/EM66 (Nuclear Materials Stabilization Task Group), June 1996.

■ **The Science and Technology Panel of the University of California President's Council on the National Laboratories** visited Los Alamos on April 19 to be briefed on programmatic activities of several divisions. For NMT, Division Director Bruce Matthews gave an hour-long presentation on NMT's responses to the observations and concerns raised by the DRC in its most recent report, several new programmatic initiatives undertaken by the division, the funding outlook for both science and technology, and facility and infrastructure support.

■ LDRD News

A Laboratory Directed Research and Development (LDRD) proposal by **Larry Avens** (NMT-6) and a number of co-principle investigators has been selected for funding at one million dollars for five years. The title of this winning proposal is "Actinide Molecular Science," and it proposes a fundamental investigation into a broad range of actinide science topics.

As of May 20, the last day for the 1997 LDRD proposals, NMT technical staff members submitted a record number of individual projects and program development proposals. **Ed Heighway**, LDRD Office Leader, sends a message that he is delighted to see NMT is playing an improved and significant role in the Laboratory's LDRD program.

NewsMakers

Division Director **Bruce Matthews** presented a colloquium titled “The Plutonium Legacy” at the University of California (UC), Berkeley, on April 15. The audience came from both the Nuclear Engineering and Materials Science and Mineral Engineering Departments. Several faculty members and students expressed strong interest in continued dialogue and potential collaborative work with Los Alamos.

Bruce Matthews was elected to the rank of Fellows of the American Nuclear Society. The citation reads, “For his unique contributions in nuclear fuels technology development (fabrication, testing, and demonstration) and being an ardent proponent of safe, beneficial uses of nuclear technologies for the nation and the world.”

Paul Cunningham served as Science Counsel in Vic Reis’ Washington office during March, April, and May. Reis is the Assistant Secretary for Defense Programs in the Department of Energy (DOE). One aspect of Cunningham’s assignment was to arrange a “Plutonium Workshop” for Defense Programs contractors and DOE personnel.

Dana Christensen gave colloquia at UC Davis (May 20), New Mexico State University (NMSU) (April 2), and New Mexico Institute of Mining and Technology (April 3). He informed his audiences on the state of current nuclear materials and nuclear weapon activities. He also discussed potential collaborations with faculty and students at all three campuses, including summer and sabbatical assignments and support for campus projects.

Consultant **Gerd Rosenblatt** spent two days with us on March 21 and 22, and two days on May 13 and 14. Rosenblatt is the former Deputy Director of Lawrence Berkeley National Laboratory (retired) and was Division Leader of the Los Alamos Chemistry Division. He will advise the NMT Division Director and the Nuclear Materials and Stockpile Management Program Director on science and technology issues, facilitate university interactions, and help in recruiting scientists and students.

NMT has a new Division Review Committee member. **Todd R. LaPorte** is Professor of Political Science and formerly Associate Director of the Institute of Government Studies at UC, Berkeley. He teaches and publishes in the areas of public administration, organization theory, and technology and politics, with particular emphasis on the decision-making dynamics of large, complex, technologically intensive (and hazardous) organizations. Sound familiar to you at TA-55? We are very pleased that Professor LaPorte has joined us as a member of the NMT External Advisory Team. LaPorte’s first visit is scheduled July 11 and 12.

Keith W. Fife (NMT-2) submitted his Ph.D. dissertation in chemical engineering to NMSU’s Graduate School. Fife did his graduate work while maintaining a full-time career at TA-55. The dissertation is entitled “A Kinetic Study of Plutonium Dioxide Dissolution in Hydrochloric Acid Using Iron (II) as an Electron Transfer Catalyst.” Congratulations, Keith, for your hard work and for the successful completion of your Ph.D. program.

Los Alamos

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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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