FY 1970

PLUTONIUM RESEARCH PROGRAM

FY 1970

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Compiled by

THE PLUTONIUM RESEARCH
COORDINATING COMMITTEE

MASTER

UNITED STATES ATOMIC ENERGY COMMISSION

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PLUTONIUM RESEARCH
PROGRAM

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INTRODUCTION

This report contains a compilation of unclassified plutonium programs underway in FY 1970 in the field of materials science. It includes work in ceramics, metallurgy, solid state physics and physical chemistry. Information on each of the programs is given in five sub-headings:

- a) Scope of the work
- b) Technical effort in manyears (M.Y.)
- c) Primary class of materials studied
- d) Person(s) to contact for further information
- e) Reports and publications

All the work listed is restricted to either research or long range development and not applied or hardware-type projects. The report was compiled from information supplied by each laboratory and is part of an overall survey of plutonium research made by the Plutonium Research Coordinating Committee. This Committee is composed of the following members:

L. C. Ianniello, AEC, Chairman
R. D. Baker, LASL
T. D. Chikalla, PNL
Z. Hagedorn, AEC
M. V. Nevitt, ANL
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1. Physical and Mechanical Metallurgy of Plutonium and Other Actinide Metals

a) This program involves the study of the deformation of single crystals of alpha plutonium and the identification of ($\alpha \rightarrow \beta$) transformation mechanism in neptunium. Ancillary work is involved in the preparation of alpha plutonium single crystals and in a feasibility study of the purification of plutonium by electrodiffusion.

- Deformation of alpha plutonium single crystals.

Single crystals are strained in compression or in tension at room temperature and at higher and lower temperatures in an effort to clarify and define the complex deformation behavior, apparently involving a large number of slip planes.

- Transformation mechanisms in neptunium.

High temperature Laue-pattern work, X-ray diffractometry and optical metallography are being employed to analyze habit and correspondence planes in the $\alpha \rightarrow \beta$ transformation of neptunium. Kinetic data have been obtained by electrical resistivity for the same transformation. Lattice constants have been determined from room temperature to 450°C.

- Preparation of single crystals of alpha plutonium.

Production of single crystals of alpha plutonium have been accomplished on a semi-routine basis. Effort is now being devoted to further reduce subgraining in the crystals.

- High purity plutonium by electrodiffusion.

Experimental conditions for refining are being optimized. Resistivity ratios are being correlated with chemical analyses to assess the degree of success that is achieved by this method.

- b) 3 M.Y.
- c) Metallic
- d) N. L. Peterson
- e)

1969

Slip in Crystals of Alpha Plutonium

R. G. Liptai and R. J. Friddle

J. Metals 21 (3), 17A (March 1969), Abstract

ARGONNE NATIONAL LABORATORY

- e) Studies on the $\alpha \rightleftharpoons \beta$ Phase Transformations in Neptunium
J. J. Rechtien, M. B. Brodsky, and D. Lieberman
J. Metals 21 (3), 79A (March 1969), Abstract

High Pressure Crystal Growth of Alpha Plutonium
R. G. Liptai and R. J. Friddle
J. Crystal Growth 5, 216-218 (1969)

Internal Friction of Neptunium
J. E. Selle and J. J. Rechtien
J. Nucl. Mater. 31, 203-210 (1969)

1968

On the Emission of Elastic Waves during Martensitic Transformations
R. G. Liptai
J. Metals 20, 37A (1968), Abstract

The Crystal Geometry of the $\beta \rightarrow \alpha$ and $\alpha \rightarrow \beta$ Displacive Transformations in Neptunium
J. J. Rechtien and D. S. Lieberman
J. Metals 20 (8), 25A (August 1968), Abstract

Deformation of Alpha Plutonium
R. G. Liptai and R. J. Friddle
J. Metals 20, 135A (1968), Abstract

Internal Friction of Neptunium
J. J. Rechtien and J. Selle
J. Metals 20, 120A (1968), Abstract

1967

The Pressure/Temperature Behavior of Pure Plutonium
M. B. Brodsky
Plutonium 1965, Proc. 3rd Intern. Conf. on Plutonium, London, ed. A. E. Kay and M. B. Waldron. Chapman and Hall, London, 1967, pp. 137-139, Discussion

On the Formation of Substructure in Alpha Plutonium at High Pressures
R. G. Liptai and R. J. Friddle
J. Nucl. Mater. 24, 316-322 (December 1967)

- , e) The $\beta \rightarrow \alpha$ Phase Transformation in High Purity Plutonium
Moshe Rosen, L. T. Lloyd, and R. G. Peterson
Plutonium 1965, Proc. 3rd Intern. Conf. on Plutonium, London,
ed. A. E. Kay and M. B. Waldron. Chapman and Hall, London,
1967, pp. 18-38

2. Defect Equilibria in the Compounds of Plutonium and Other Actinides

- a) The study of defect equilibria in PuO_{2-x} has prompted the development of a statistical model, on which a recent attempt has been made to generalize into a new system of spacing statistics.

The partial pressure of oxygen over PuO_{2-x} and a statistical analysis of the lattice defects of PuO_{2-x} is essentially complete. A study of the sorption of oxygen, water and possibly hydrogen has been launched on stoichiometric PuO_2 . Later on, sorption on the hypostoichiometric form of the same compound will be studied. The objectives of this work are to determine the relevant thermodynamic quantities and to identify the processes involved in sorption.

- b) 3 M.Y.
c) Nonmetallic
d) N. L. Peterson
e)

1969

The Distribution of Two Assemblies of Defects on Separate
Sublattices
Leon M. Atlas
Proc. Solid State Chemistry Symp., Tucson, Arizona,
April 1969

1968

Statistical Model of Partially Ordered Defects in UO_{2+x}
Leon M. Atlas
J. Phys. Chem. Solids 29 (8), 1349-1358 (August 1968)

Statistical Model of Partially Ordered Defects in a Hypo-
stoichiometric Metal Oxide
L. M. Atlas
J. Phys. Chem. Solids 29, 91-100 (January 1968)

1967

Defect Equilibria of PuO_{2-x} , 1045°C to 1505°C
L. M. Atlas and G. J. Schlehman
Plutonium 1965, Proc. 3rd Intern. Conf. on Plutonium, London,
ed. A. E. Kay and M. B. Waldron. Chapman and Hall, London,
1967, pp. 838-844

3. Electronic Structure of Plutonium Metal and Other Actinides

a) The electronic structure of plutonium and neighboring actinide metals is delineated by the following types of measurements and their interpretation: electron transport studies; bulk magnetic studies; NGR (Mössbauer) effect; Fermi surface topology.

. Electron transport

Carefully made resistivity measurements have yielded no evidence of a magnetic transition in alpha plutonium. However, additional single-crystal measurements are being made to provide confirmation and clarification, and to give correlative support to the interpretation of Hall coefficient and magnetoresistivity measurements that are currently being done to establish the full resistivity tensor. Thus far, there is no substantive evidence for the existence of an ordered, low temperature, magnetic structure in pure alpha plutonium. Pure americium behaves similarly.

Resistivity measurements being made on a series of dilute alloys of Pu and Np in Pd have shown that these are Kondo-type systems; i.e., magnetic moments localized on the actinide solute are compensated by the conduction electrons of the host giving anomalous resistivity behavior.

. Bulk magnetization

Further confirmation of the absence of a magnetic transition at low temperature is being sought by measurements of polycrystalline susceptibility and single-crystal susceptibility.

Bulk magnetization measurements on the Pu, Np-Pd alloys have supported the local moment explanation of the resistivity behavior.

Correlated, confirmatory measurements are continuing.

. NGR (Mössbauer) effect

Nuclear gamma-ray resonance (NGR) has been observed in ^{243}Am from the beta decay of ^{243}Pu [bred via $^{242}\text{Pu}(n, \gamma)$]. This Mössbauer system and those involving several uranium isotopes with their respective alpha-active plutonium parents, are being studied systematically and used where appropriate in the identification of magnetic transitions. For example, results appear to confirm that 10% neptunium dissolved in plutonium forms an ordered magnetic structure.

Studies of the systematics of existing and potential Mössbauer systems will continue. As more is learned about the NGR and general electronic and magnetic properties of the heavier actinide metals and compounds, more specific problems will be studied. One that has been chosen is ^{237}Np (absorber) in solutions with plutonium and uranium. Additional systems of interest are: $^{246}\text{Cm} \xrightarrow{\alpha} ^{242}\text{Pu}$, $^{243}\text{Am} (n, \gamma) ^{244}\text{Am} \xrightarrow{\beta} ^{244}\text{Cm}$, etc. These studies will be greatly aided by receipt of ^{242}Pu and ^{243}Am metal.

. Fermi surface topology.

De Haas van Alphen and galvanometric measurements are under way on single crystals of plutonium having resistance ratios >30 in 70 kOe magnetic fields. The use of the ^{250}kOe field at the Francis Bitter National Magnet Laboratory is planned. To avoid self damage, a sample of ^{242}Pu has been ordered, from which a single crystal can be made.

- b) 9 M.Y.
- c) Metallic
- d) N. L. Peterson
- e)

1969

NGR Study of Hyperfine Interactions in ^{238}U

S. L. Ruby, G. M. Kalvius, B. D. Dunlap, G. K. Shenoy, D. Cohen, M. B. Brodsky, and D. J. Lam

Phys. Rev. 184, 374-380 (1969)

Magnetic Susceptibility of Pd-Np and Pd-Pu Solid Solutions

M. B. Brodsky

Bull. Am. Phys. Soc. 14 (3), 321 (March 1969), Abstract

Electrical Resistivity of Alpha Uranium at Low Temperatures

M. B. Brodsky, N. J. Griffin, and M. D. Odie

J. Appl. Phys. 40 (2), 895-897 (February 1969), Communication

NGR Measurements with ^{238}U

S. L. Ruby, G. M. Kalvius, B. D. Dunlap, G. K. Shenoy,

M. B. Brodsky, and D. Cohen

Bull. Am. Phys. Soc. 14 (1), 134 (January 1969), Abstract

Mössbauer Isomer Shift in ^{243}Am

G. M. Kalvius, S. L. Ruby, B. D. Dunlap, G. K. Shenoy,

D. Cohen and M. B. Brodsky

Physics Letters 29B, 489-490 (1969)

- e) Hyperfine Interaction and Susceptibility in Some Actinide Metals and Intermetallic Compounds
B. D. Dunlap, M. B. Brodsky, G. M. Kalvius, G. K. Shenoy, and D. J. Lam
J. Appl. Phys. 40, 1495-1496 (1969)

Isomer Shift of the Nuclear γ -Ray Resonance (NGR) in ^{243}Am
G. M. Kalvius, S. L. Ruby, B. D. Dunlap, G. K. Shenoy, D. Cohen, and M. B. Brodsky
Bull. Am. Phys. Soc. 14, 521 (1969), Abstract

Hyperfine Spectra and Anisotropic Lattice Vibrations of ^{237}Np in α -Np Metal
B. D. Dunlap, G. K. Shenoy, G. M. Kalvius, and M. B. Brodsky
Bull. Am. Phys. Soc. 14 (1), 132-133 (January 1969), Abstract

Hyperfine Field of ^{237}Np in NpO_2 ¹⁹⁶⁸
B. D. Dunlap, G. M. Kalvius, D. J. Lam, and M. B. Brodsky
J. Phys. Chem. Solids 29 (8), 1365-1367 (August 1968)

Isomer Shift and Hyperfine Splittings of the 59.6-keV Mössbauer Resonance in ^{237}Np
B. D. Dunlap, G. M. Kalvius, S. L. Ruby, M. B. Brodsky, and D. Cohen
Phys. Rev. 171 (2), 316-321 (July 10, 1968)

Isomer Shifts and Hyperfine Splittings of the 59.6-keV Resonance in ^{237}Np
B. D. Dunlap, G. M. Kalvius, S. L. Ruby, M. B. Brodsky, and D. Cohen
Hyperfine Structure and Nuclear Radiations, Proc. Intern. Conf. on Hyperfine Interactions Detected by Nuclear Radiation, Asilomar, Conf. Grounds, Pacific Grove, California, August 25-30, 1967, E. Matthias and D. A. Shirley, eds., North-Holland Publishing Company, Amsterdam, The Netherlands, 1968, pp. 84-91

¹⁹⁶⁷
Thermoelectric Power of Delta Plutonium, and Resistivity of Americium and Their Relevance to Magnetism in the Actinide Elements
M. B. Brodsky
Plutonium 1965, Proc. 3rd Intern. Conf. on Plutonium, London, ed. A. E. Kay and M. B. Waldron. Chapman and Hall, London, 1967, pp. 210-212, Discussion

e) Transverse Magnetoresistivity of α and β Plutonium

M. B. Brodsky

Phys. Rev. 163, 484-487 (November 10, 1967)

Temperature Dependence of the Hyperfine Field of Np^{237} in NpO_2

B. D. Dunlap, G. M. Kalvius, M. B. Brodsky, and D. J. Lam

Bull. Am. Phys. Soc. 13, 28 (January 1968), Abstract

Hall Effect and Electronic Structure of Various Actinide Metals

M. B. Brodsky

Plutonium 1965, Proc. 3rd Intern. Conf. on Plutonium, London,

ed. A. E. Kay and M. B. Waldron. Chapman and Hall, London,

1967, pp. 286-298

4. Electronic Structure of Plutonium Compounds and Compounds of
Neighboring Actinides

a) Magnetic susceptibility and nuclear magnetic resonance measurements are made on NaCl-type and CaF_2 -type compounds of plutonium with nontransition elements of Groups IV, V, and VI. Corresponding measurements are made on some of the same compounds of uranium, neptunium and americium. The purpose of this experimental work, which is closely coupled to theoretical studies, is to delineate electronic structure. Supporting alloy-chemistry studies have been conducted to define the composition ranges of the relevant compounds.

. Susceptibility measurements

The magnetic susceptibilities of the NaCl-type compounds of plutonium with carbon and phosphorous have been measured, and corresponding measurements on the isomorphous compounds of nitrogen and sulfur are under way. First-principle calculations of the low-lying electronic states for various actinide ions, including plutonium, in the crystalline state are in progress, utilizing a noninteracting ion model. The theoretical model yields good agreement with the experimental results.

. Nuclear magnetic resonance studies

NMR measurements on the NaCl-type compound PuP above the Curie temperature have been performed. Evidence has been collected for the occurrence of ferromagnetic correlations existing in the paramagnetic state of this compound. Corresponding measurements on other isomorphous compounds of plutonium, and also uranium and neptunium, are under way.

b) 3 M.Y.

c) Nonmetallic

d) N. L. Peterson

e) 1969

Magnetic Properties of Plutonium Monophosphide

D. J. Lam, F. Y. Fradin, and O. L. Kruger

Bull. Am. Phys. Soc. 14 (3), 387 (1969), Abstract

Magnetic Properties of Plutonium Monophosphide

D. J. Lam, F. Y. Fradin, and O. L. Kruger

Phys. Rev. 187 (2), 606 (1969)

Alloy Chemistry of Th, U, and Pu Compounds

A. E. Dwight

Structural Developments in Alloy Phases, Plenum Press (1969)

Nuclear Relaxation in Plutonium Phosphide Above the Curie Temperature

F. Y. Fradin

Solid State Commun. 7 (10), 759-761 (1969)

Ground States of Actinide Ions in the Crystal Field of Cubic and Octahedral Symmetry

S. K. Chan and D. J. Lam

Bull. Am. Phys. Soc. 14 (3), 351 (1969), Abstract

1968

The Unit-cell Constants of Some PuNi₃-type Compounds

A. E. Dwight

Acta Cryst. B24 (10), 1395-1396 (1968)

Magnetic Properties of Plutonium Monocarbide Between 5 to 1000°K

D. J. Lam, J. W. Ross, and L. L. Isaacs

Bull. Am. Phys. Soc. 13, 461 (1968), Abstract

1967

The Magnetic Susceptibility of the Monocarbides of Uranium and Plutonium

D. J. Lam, M. V. Nevitt, J. W. Ross, and A. W. Mitchell

Plutonium 1965, Proc. 3rd Intern. Conf. on Plutonium, London, eds. A. E. Kay and M. B. Waldron. Chapman and Hall, London, 1967, pp. 274-285.

5. Neutron Scattering Measurements on Compounds of Plutonium and Neighboring Actinides

a) This study involves determinations of thermal neutron scattering lengths for certain Pu isotopes, and the beneficial use of these data in delineating the crystalline and magnetic ordering of Pu in certain compounds.

The thermal neutron scattering length for ^{240}Pu has been obtained from a neutron diffraction study of $^{240}\text{PuO}_2$. Measurements are now under way to determine the scattering length of ^{242}Pu , using a sample of $^{242}\text{PuO}_2$.

A search for magnetic ordering in PuO_2 and $\text{PuO}_{1.7}$ is now being carried out; both ^{240}Pu and ^{242}Pu isotopes have been found to be suitable for neutron studies.

Corresponding studies on NpO_2 are also being pursued.

- b) 1 M.Y.
- c) Nonmetallic
- d) N. L. Peterson
- e)

1969

Neutron Diffraction Study of NpC

G. H. Lander, L. Heaton, M. H. Mueller, and K. D. Anderson
J. Phys. Chem. Solids 30, 733-737 (1969)

1967

A Neutron Diffraction Determination of the Coherent Scattering Amplitude of Np and the Possible Antiferromagnetism of Neptunium Dioxide

L. Heaton, M. H. Mueller, and J. M. Williams
J. Phys. Chem. Solids 28, 1651-1654 (1967)

6. Thermodynamic Properties of Inorganic Materials (Calorimetric Studies)

a) In this program, enthalpy of formation data are obtained on inorganic materials by the use of calorimetric methods (solution and flow calorimeters and bomb combustion calorimeters using oxygen, fluorine, or chlorine). A significant fraction of our work has been on uranium and plutonium compounds and their combustion products. The plutonium calorimetric facility was operated on a routine basis in 1968 and 1969, data being obtained for PuO_2 , PuN and PuC by the oxygen bomb calorimetric method. Exploratory work with fluorine bomb calorimetry has indicated that enthalpy-of-formation values for PuF_3 and PuF_4 will be necessary auxiliary (or building block) data. However, adequately pure samples have not been available as yet.

Enthalpy data for PuO_2 has been determined. It is hoped a sample of Pu_2O_3 will be made available some time for study. It is hoped, also, both PuS and Pu_2S_3 will be available for study some time in the future. PuN has been studied; PuP is a candidate for our program, and $\text{PuC}_{0.86}$ has been studied. Los Alamos is preparing a sample of Pu_2C_3 for our use; if this sample is adequately characterized, measurements will be made.

- b) 4 M.Y.
- c) Nonmetallic
- d) H. M. Feder
- e) (See composite list)

7. Thermodynamics of Fuel Materials

a) Under this activity, the chemical and thermodynamic properties and phase diagrams of LMFBR fuels, fission products, and fuel additives will be studied. For the near term, this program will concentrate on carbide and oxide fuels and on a few of the more important fission products (fuel swelling and cladding attack) at temperatures from 800°C to the melting point of the reactor fuels. In these studies, chemical potentials, vapor species pressures, transport phenomena, and phase diagrams will be studied as functions of fuel composition and temperature.

- b) 10 M.Y.
- c) Nonmetallic
- d) A. D. Tevebaugh
- e) (See composite list)

8. Reactor Materials Behavior at High Temperatures

a) The program on the behavior of reactor materials at high temperatures consists of physical property measurements needed for fast reactor safety analysis and the theoretical extrapolation of these data to temperatures beyond the temperature limits (~ 3000 - 3500°K) for experimental studies. In the experimental studies, measurements are under way or will be made of the vapor pressure, enthalpy and heat capacity, compressibility, thermal diffusivity, phase equilibria, density, surface tension, and viscosity of reactor fuels and structural materials. In the theoretical studies, existing theories such as corresponding states, scaled particle, significant structures, etc., will be used to extend the physical property data beyond the temperature limits for the experimental studies.

The current experimental studies are limited primarily to measurements of enthalpy and heat capacity, vapor pressure, compressibility, and thermal diffusivity of UO_2 . The enthalpy and heat capacity measurements have recently been extended to $(\text{U Pu})\text{O}_2$, and phase equilibria studies will soon start on $(\text{U Pu})\text{O}_2$ containing nonradioactive isotopes of the fission products and fuel cladding materials. The latter studies will provide information concerning the distribution of fission product heat sources after a reactor meltdown accident.

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- b) 4 M.Y.
- c) Nonmetallic
- d) A. D. Tevebaugh
- e) This list is a composite list for three programs; #6, #7, #8:
 - . Thermodynamic Properties of Inorganic Materials (Calorimetric Studies)
 - . Thermodynamics of Fuel Materials
 - . Reactor Materials Behavior at High Temperatures

Papers describing results of plutonium studies and equipment for plutonium studies are included.

1969

The Enthalpies of Formation of Plutonium Dioxide and Plutonium Mononitride

G. K. Johnson, E. H. Van Deventer, O. L. Kruger, and
W. N. Hubbard

J. of Chemical Thermodynamics 1, 89-98 (1969)

The Enthalpy of Formation of Plutonium Monocarbide

G. K. Johnson, E. H. Van Deventer, O. L. Kruger, and
W. N. Hubbard

To be submitted.

Migration of Plutonium in Fast Reactor Fuels Under the Action of
a Thermal Gradient

M. G. Chasanov and D. F. Fischer
ANL report in preparation.

A Mass Spectrometric Study of the Volatilization Behavior in
the Plutonium-Carbon System

J. E. Battles, W. A. Shinn, P. E. Blackburn, and R. K. Edwards
Accepted for publication in the J. of High Temp. Sci.

A Foreline Trap for Contamination-Free Roughing

P. M. Danielson and F. C. Mrazek
J. Vacuum Sci. Technol. 6 (3), p. 423 (May/June 1969)

Sealing Large Ultrahigh-Vacuum Flanges with Polytetrafluoroethylene
Gaskets

P. M. Danielson
J. Vacuum Sci. Technol. 6, p. 425 (May/June 1969)

A Demountable Induction-Heater Power-Lead Coupling for Use in
Ultrahigh Vacuum

P. M. Danielson
Rev. Sci. Instr. 40 (5), 736-737 (May 1969)

- e) Law of Maxima in Species Partial Pressures for Equilibria in ,
Two-Component Systems
R. K. Edwards
High Temperature Science, 1 (2), 232-237 (July 1969)
- A Demountable, Stainless Steel-to-Aluminum Transition Seal for
Ultrahigh Vacuum
P. M. Danielson
Vacuum, 19 (8), p. 365 (August 1969)
- A Design for Modular Ultrahigh Vacuum Feedthroughs
P. M. Danielson
Vacuum, 19 (9), p. 412 (September 1969)
- An Investigation of Gasket Coating Materials for Ultrahigh
Vacuum Seals
P. M. Danielson
Submitted to the J. Vacuum Sci. Technol.
- Synthesis and Fabrication of (U,Pu)C Nuclear Fuel Pellets from
U,Pu Alloy
P. A. Nelson, D. E. Grosvenor, S. Vogler, N. P. Quattropani,
P. W. Krause
Amer. Cer. Soc. Bull. 48 (9), 863 (1969)
- The Solubility of Uranium and Plutonium in Liquid Alloys
I. Johnson
Proc. Symp. on Reprocessing of Nuclear Fuels, Ames, Iowa,
Aug. 25-27, 1969, Nuclear Metallurgy, Vol. 15, USAEC
Report CONF-690801, pp. 547-564 (1969)
- Plutonium Tetrafluoride Preparation and Separation by Sorption
on Sodium Fluoride
M. J. Steindler, A. A. Jonke
U. S. Patent No. 3,423,190 dated January 1969
- Production of Plutonium Hexafluoride by Fluorination in a
Fluidized-Bed
N. M. Levitz, G. J. Vogel, E. L. Carls, D. E. Grosvenor,
B. J. Kullen, D. J. Raue, and W. A. Murphy
Nucl. Appl. 6 (2), 147-155 (1969)
- Separation of Ruthenium and Plutonium by a Lithium Fluoride
Sorption Technique
J. G. Riha, L. E. Trevorrow, M. J. Steindler
U. S. Patent No. 3,458,291 dated July 29, 1969

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- e) Contribution to the Plutonium-Magnesium Phase Diagram
W. Knoch, J. B. Knighton, R. K. Steunenberg
Proc. Symp. on Reprocessing of Nuclear Fuels, Ames, Iowa,
Aug. 25-27, 1969, Nuclear Metallurgy, Vol. 15, USAEC
Report, CONF-690801, pp. 535-546 (1969)

Ultraviolet-Activated Synthesis of Plutonium Hexafluoride at
Room Temperature

L. E. Trevorow, T. J. Gerding, M. J. Steindler
Inorg. Nucl. Chem. Letters, accepted for publication

The Liquid Scintillation Determination of Plutonium

R. P. Larsen, L. E. Ross, E. C. Bogusch
In preparation; to be submitted for publication

1968

Helium Pumping with a Sputter-ion Pump

P. M. Danielson
J. Vacuum Sci. Technol., 5 (89) (1968)

Fluorination of Uranium, Plutonium, and Neptunium from a
Fluidized Bed of Alumina

L. J. Anastasia, P. G. Alfredson, M. J. Steindler
Abstracts of Papers, 155th Amer. Chem. Soc. Mtg., San
Francisco, March 31-April 5, 1968. Abstract 0-145

Laboratory Investigations in Support of Fluid-Bed Fluoride
Volatility Processes. Part XIX. Reaction of Bromine Penta-
fluoride with Selected Compounds of Uranium and Plutonium

R. L. Jarry, J. J. Stockbar, M. J. Steindler
ANL-7412 (1968)

Kilogram-Scale Production of Plutonium Hexafluoride in a
Fluidized Bed

N. M. Levitz, G. J. Vogel, E. L. Carls, D. Grosvenor, B. Kullen,
D. Raue, W. Murphy
Trans. Amer. Nucl. Soc. 11 (1), 35 (1968)

The Chemistry of Plutonium Hexafluoride

M. J. Steindler
Proc. of Rocky Flats Fluoride Volatility Meeting, Golden,
Colo., June 24-25, 1968, USAEC Report CONF-680610, p. 2
(1968)

Reaction of PuO_2 with Molten MgCl_2

D. A. Wenz, I. Johnson
Inorg. Nucl. Chem. Letters 4, 735-738 (December 1968)

e)

1967

The Volatility of Plutonium Carbides, Part I

P. S. Harris, B. A. Phillips, M. H. Rand, M. Tetenbaum*

UKAEA Report AERE-R-5353 (March 1967)

* ANL-CEN Division

Thermal Gradient Apparatus for Use in a Glovebox

D. F. Fischer and M. G. Chasanov

Rev. Sci. Instr. 38, 1574 (1967)

Extraction of Plutonium from Uranium-Plutonium Alloy with Uranium Trichloride

N. R. Chellew, R. K. Steunenberg

Nucl. Appl. 3 (3), 142-146 (1967)

The Melting-Point Diagram for the System Uranium Hexafluoride-Plutonium Hexafluoride

L. E. Trevorow, M. J. Steindler, D. V. Steidl, J. T. Savage

Inorg. Chem. 6 (5), 1060-1061 (1967), Note

Removal of Plutonium Hexafluoride from Cell Exhaust Air by Hydrolysis and Filtration

R. W. Kessie

Proc. of Ninth AEC Air Cleaning Conf., Boston, Mass.,
Sept. 13-16, 1966, Vol. 2, pp. 741-762, J. M. Morgan, Jr.
and M. W. First (Eds.), USAEC Report CONF-660904
(January 1967)

Plutonium and Uranium Hexafluoride Hydrolysis Kinetics

R. W. Kessie

Ind. Eng. Chem. Process Design Develop. 6 (1), 105-111 (1967)

Fluidized-Bed Technique for Preparation of Uranium-Plutonium Monocarbide

E. J. Petkus, P. A. Nelson, S. Vogler, P. W. Krause

Chem. Eng. Progr. Symp. Ser. 63 (80), 111-120 (1967)

Laboratory Investigations in Support of Fluid-Bed Fluoride Volatility Processes. Part XV. Estimation of Rates of Thermal Decomposition of Plutonium Hexafluoride

L. E. Trevorow, M. J. Steindler

ANL-7347 (1967)

9. Development of Mixed Plutonium Oxide-Uranium Oxide Fast Reactor Fuel

a) This program has as its aim to predict accurately and reliably the performance and lifetime of mixed plutonium-oxide-uranium oxide fuel elements for the LMFBR. Fuel compositions of interest center around UO_2 -20 w/o PuO_2 , with an O/M ratio of 1.97 and a smear density of 80, 85 and 90%. The scope covers: (1) in-reactor experiments and postirradiation examinations to assess fuel swelling behavior and (2) studies of the compatibility of mixed oxide fuel with potential cladding materials

Fuel swelling studies.

The swelling behavior of mixed oxide fuel is being investigated by employing three coordinated approaches:

(1) In-pile measurements of intrinsic swelling rate are being made as a function of temperature, temperature gradient, burnup rate, total burnup and external pressure.

(2) Kinetics of fission-gas bubble nucleation, growth, migration and resolution are being obtained from in-pile experiments as a function of temperature, temperature gradient, cooling rate, burnup rate, and total burnup.

(3) Microscopic postirradiation examination of EBR-II irradiated samples is employed to determine fission-gas distribution and content. Transmission electron microscopy is employed, as are microsampling techniques in conjunction with a quadrupole mass spectrometer.

Compatibility studies.

This work which is essentially completed has involved the out-of-pile study of the interactions of mixed uranium-plutonium oxide fuel material $[(\text{U}_{0.8}\text{Pu}_{0.2})\text{O}_{1.97}]$ in contact with selected vanadium alloys developed at ANL and at Westinghouse. Reaction zones are examined by optical microscopy, microhardness and electron microscopy.

- b) 13 M.Y.
- c) Nonmetallic
- d) B. R. T. Frost

e)

1969

Performance of Mixed-Oxide Fuel Elements with Center Melting
F. L. Brown, L. A. Neimark, W. F. Murphy, and R. Natesh
Trans. Amer. Nucl. Soc. 12 (1), 107-108 (June 1969), Abstract

Compatibility of Vanadium Alloys with (U,Pu)O₂ and (U,Pu)C

T. W. Latimer

J. Metals 21 (3), 107A (March 1969), Abstract

1967

Performance of Vibratorily Compacted Mixed-Oxide Fuel Rods
Under Fast-Reactor Conditions

F. L. Brown, L. A. Neimark, B. J. Koprowski, J. E. Ayer,
J. H. Kittel

Trans. Amer. Nucl. Soc. 10 (1), 101-102 (June 1967), Summary

10. Migration of Major Fuel Constituents and Characterization of Fission
Products in Mixed UO₂-PuO₂

a) The determination of the kinetics of uranium and plutonium migration in mixed-oxide fuel constitutes the scope of this program. The intent is to establish and evaluate those factors that influence the migration of uranium and plutonium in a fast reactor fuel and identify: (a) the migration mechanisms of uranium and plutonium during irradiation in mixed oxide and mixed carbide fuels; priority will be given to the work on mixed oxides; (b) measurement of chemical diffusion of uranium and plutonium under in-reactor conditions; and (c) calculation of the effective diffusivity coefficient (D) and heat of transport (Q*) for uranium and plutonium, using a shielded microprobe analyzer. By selection of fuel samples, the following variables are being studied: density, burnup, power rating, fabrication history, maximum temperature and temperature gradient. The principal material in the near-term phase of the study is UO₂-20 w/o PuO₂.

The study also covers the identification of the crystal chemistry of solid fission-product phases and a quantitative assessment of the swelling contributed by these phases.

b) 6 M.Y.

c) Nonmetallic

d) B. R. T. Frost

e)

1969

Solid Fission Product Behavior in Uranium-Plutonium Oxide Fuel
Irradiated in a Fast Neutron Flux

D. R. O'Boyle, F. L. Brown, and J. E. Sanecki

J. Nucl. Mater. 29 (1), 27-42 (January 1969)

- e) 1968
Observations on Metallic Ingots Formed During Irradiation of Mixed-Oxide Fuel
D. R. O'Boyle and F. L. Brown
Trans. Amer. Nucl. Soc. 11 (1), 101-102 (June 1968), Abstract

1967
Microanalysis of Solid Fission Products in Mixed-Oxide Fuel Irradiated in EBR-II
D. R. O'Boyle, F. L. Brown, and J. E. Sanecki
Trans. Amer. Nucl. Soc. 10 (2), 426-463 (November 1967)

11. Chemistry of Irradiated Fuel Materials

a) This program concerns the use of analytical techniques and instrumentation needed for studies on fuel microstructure, cladding-fuel interactions and fuel-coolant interactions that occur in irradiated fuel samples. For the near term, the major effort will be on the irradiated mixed uranium oxide-plutonium oxide fuel. The data obtained are correlated with observations on fuel swelling, fuel fabrication variables (e.g. O/M, impurities, additives, etc.) and irradiation variables.

An electron microprobe, laser sampler, a spark source mass spectrometer, alpha, gamma and X-ray spectrometer, a metallograph, a quadrupole mass spectrometer and more conventional tools are employed.

- b) 3 M.Y.
c) Nonmetallic
d) C. E. Crouthamel
e)

1969
Distribution of Alpha and Gamma Emitting Nuclides in a Fast Reactor Irradiated UO_2 - PuO_2 Fuel
M. D. Adams
Abstracts of Papers Presented at the 158th Amer. Chem. Soc. Meeting, New York, September 8-12, 1969, NUCL-52

Electron Microprobe Studies of the Migration of Elements in Irradiated Oxide Fuels
C. E. Crouthamel, N. R. Stalica, C. E. Johnson, C. A. Seils
Abstracts of Papers Presented at the 158th Amer. Chem. Soc. Meeting, New York, September 8-12, 1969. NUCL-47

- e) Studies in Mixed-Oxide Irradiated Fuels: Transport of Cladding Components

C. E. Johnson, C. E. Crouthamel, H. Chen, P. E. Blackburn
Trans. Amer. Nucl. Soc. 12 (2), 565 (1969), Abstract

Cladding Interactions in Mixed-Oxide Irradiated Fuels

C. E. Johnson, C. E. Crouthamel
J. Nucl. Mater. (In press)

Compatibility of Carbide Fuels with Potential LMFBR Claddings

T. W. Latimer, L. A. Neimark, C. E. Johnson, C. E. Crouthamel
Trans. Amer. Nucl. Soc. 12 (2), 594 (1969), Abstract

Electron Probe Microanalysis of Irradiated Oxide Fuels

N. R. Stalica, C. A. Seils, C. R. Crouthamel
Amer. Cer. Soc. Symp. Nucl. Fuels, Vol. 2, p. 211 (1969)

1968

Determination of Isotope Distribution in the Microstructure of Irradiated Ceramic Fuels

M. D. Adams, S. C. Tong, L. E. Ross
Abstracts of Papers, 155th Amer. Chem. Soc. Meeting, San Francisco, March 31-April 5, 1968, Abstract 0-19

12. High Temperature Mechanical Properties of Ceramic Fuels

a) This program covers long-time creep behavior and short-time mechanical properties of mixed uranium oxide-plutonium oxide fuel materials. There are three parts in the study: (1) irradiation-induced creep; (2) thermal creep; and (3) stress/strain data.

. Irradiation-induced creep.

This part of the study investigates the effect of neutron irradiation and simultaneous fission on the creep behavior of pure UO_2 and later UO_2 -20 w/o PuO_2 specimens having a helical geometry. The major experimental variables are fission rate (1×10^{12} to 2×10^{13} fissions/cm³-sec), torsional stress (2000 to 10,000 psi), temperature (below 600°C), stoichiometry (O/M from 2.00 to 2.10 in UO_2) and density (85 to 95% TD).

. Thermal creep.

The dependence of thermal compressive creep of mixed UO_2 -20 w/o PuO_2 upon temperature from 1000°C to 2200°C will be studied. Variables are stoichiometry, density, homogeneity, grain size and

pore dispersion. Cylindrical specimens are employed. A principal purpose is to acquire data for swelling models and fuel-element behavior codes.

- . Mechanisms of flow and fracture.

Employing strain rates of 0.1, 1.0 and 10.0 cm/cm/hr and temperature increments of 125°C from room temperature to 1800°C, samples of UO_2 -20 w/o PuO_2 are subjected to cross bending and the flow and fracture behavior is observed. Materials variables are solid solution homogeneity, stoichiometry, density, grain size, pore size and pore dispersion. The data is analyzed to predict the dominant mechanisms in the stress strain behavior.

Elastic and inelastic response in polycrystalline mixed oxide has been studied by the Förster resonance technique.

- b) 10 M.Y.
- c) Nonmetallic
- d) B. R. T. Frost
- e)

1969

Plastic Yielding and Fracture in UO_2 - PuO_2

J. T. A. Roberts and B. J. Wrona

To be published in the Bull. Amer. Cer. Soc. Meeting,
May 1970, Philadelphia

Stress Strain Behavior of UO_2 at Elevated Temperatures

R. J. Beals and R. F. Canon

Bull. Amer. Cer. Soc. 48 (9) 879, 881 (September 1969)

1968

Elastic and Anelastic Studies on Polycrystalline $(\text{U,Pu})\text{O}_2$

A. W. Nutt, A. W. Allen, and J. H. Handwerk

Bull. Amer. Cer. Soc. 47 (9), 845 (1968), Abstract

13. Uranium-Plutonium Metal Fuels for the Fast Reactor*

a) This program covers the investigation of the feasibility of using a metallic fuel system in an LMFBR. Involved are studies of fuel/cladding compatibility, physical metallurgy of the alloy fuel, and burnup and thermal performance capabilities of a sodium-bonded fuel-element system. Fuel/cladding compatibility

* This is the phase-out segment of an earlier, larger effort at ANL on metallic fuels for fast reactor applications. No work will carry beyond FY 1970.

is assessed by out-of-pile diffusion couple experiments and by post-irradiation examination of prototypal fuel elements. EBR-II irradiations have been used to assess performance limits. Virtually all recent emphasis has been on the composition U-15 w/o Pu-12 w/o Zr. Earlier work also gave attention to alloys in the U-Pu-Zr system.

- b) 2.5 M.Y.
- c) Metallic
- d) B. R. T. Frost
- e)

1968

The Irradiation Behavior of High-Burnup Uranium-Plutonium Alloy
Prototype Fuel Elements
W. N. Beck, R. J. Fousek, and J. H. Kittel
ANL-7388 (May 1968)

Thorium-Uranium-Plutonium Alloys as Potential Fast Power-
Reactor Fuels. Part I. Thorium-Uranium-Plutonium Phase
Diagram
B. Blumenthal, J. E. Sanecki, and D. E. Busch
ANL-7258 (September 1968)

Constitution of Plutonium Alloys
F. H. Ellinger, W. N. Miner, D. R. O'Boyle, and
F. W. Schonfeld
USAEC Report LA-3870, Los Alamos Scientific Laboratory
(December 1968)

Compatibility Between Metallic U-Pu-Base Fuels and Potential
Cladding Materials
S. T. Zegler and C. M. Walter
Nuclear Metallurgy, Proc. 1967 Nucl. Met. Symp., Scottsdale,
Arizona, October 4-6, 1967, K. E. Horton, R. E. Macherey,
and R. J. Allio, eds., IMD-AIME, New York, 13, 335-344 (1968)

Uranium-Plutonium Alloy Fuel Fabrication
A. B. Shuck
Nuclear Metallurgy, Proc. 1967 Nucl. Met. Symp., Scottsdale,
Arizona, October 4-6, 1967, K. E. Horton, R. E. Macherey,
and R. J. Allio, eds., IMD-AIME, New York, 13, 263-278 (1968)

Irradiation Performance of Fast Reactor Uranium-Plutonium Metal
Fuels
W. N. Beck, F. L. Brown, B. J. Koprowski, and J. H. Kittel
Nuclear Metallurgy, Proc. 1967 Nucl. Met. Symp., Scottsdale,
Arizona, October 4-6, 1967, K. E. Horton, R. E. Macherey,
and R. J. Allio, eds., IMD-AIME, New York, 13, 507-523 (1968)

- e) Transformation Kinetics of U/10 wt% Pu/10 wt% Fz Alloy
H. Savage
J. Nucl. Mater. 25 (3), 241-248 (1968)

The Heat Content and Specific Heat of Some Metallic Fast-Reactor Fuels Containing Plutonium

H. Savage
J. Nucl. Mater. 25 (3), 249-259 (1968)

1967

Technology of Sodium Cooled Fast Reactor Fuels in the U.S.

J. M. Simmons, E. R. Astley, E. A. Evans, J. H. Kittel,
D. A. Moss, and A. Strasser
Fast Breeder Reactors, Proc. London Conf. on Fast Breeder
Reactors, London, England, May 17-19, 1966, P. V. Evans,
ed. Pergamon Press, Oxford, England, pp. 615-630 (1967)

Examination Techniques for High Burnup U-Pu Fuel Elements for
Fast-Breeder Reactors

B. J. Koprowski and F. L. Brown
Proc. 15th Conf. on Remote Systems Technology, Amer. Nucl.
Soc., Hinsdale, Illinois, pp. 63-67 (1967)

Irradiation Behavior of Plutonium Alloy Fuels for Fast Reactors

W. N. Beck, W. F. Murphy, J. H. Kittel, and F. G. Foote
Plutonium 1965, Proc. Third Intern. Conf. on Plutonium,
London, England, 1965, A. E. Kay and M. B. Waldron, eds.
Chapman and Hall, London, pp. 933-948 (1967)

Discussion on Alloy Systems - 2

D. R. O'Boyle
Plutonium 1965, Proc. Third Intern. Conf. on Plutonium,
London, England, 1965, A. E. Kay and M. B. Waldron, eds.
Chapman and Hall, London, pp. 532-534 (1967)

Further Work on ANL on Fast Reactor Fuels

L. R. Kelman
Plutonium 1965, Proc. Third Intern. Conf. on Plutonium,
London, England, 1965, A. E. Kay and M. B. Waldron, eds.
Chapman and Hall, London, pp. 525-526 (1967)

Reviews of Status of Technology of Fast Reactor Fuels

E. L. Zebroski, Howard Kittel, and D. Moss
ANS Topical Meeting on Fast Reactors, San Francisco, Calif.,
April 10-12, 1967, ANS-101, pp. 2-63 - 2-80 (1967)

- e) Status of Metallic Plutonium Fast Power-Breeder Fuels
L. R. Kelman, H. Savage, C. M. Walter, B. Blumenthal,
R. J. Dunworth, and H. V. Rhude
Plutonium 1965, Proc. Third Intern. Conf. on Plutonium,
London, England, 1965, A. E. Kay and M. B. Waldron, eds.
Chapman and Hall, London, pp. 458-484 (1967)

Performance of Advanced U-Pu-Zr Alloy Fuel Elements Under
Fast-Reactor Conditions

W. N. Beck, F. L. Brown, B. J. Koprowski, and J. H. Kittel
Trans. Amer. Nucl. Soc. 10 (1), 106-107 (June 1967),
Abstract

Potential of Advanced Fuels for a Fast Test Reactor

W. J. Bailey, P. D. Cohn, D. T. Aase, F. G. Foote, F. M. Heck,
J. H. Kittel, G. A. Last, W. E. McHugh, A. A. Strasser, and
C. W. Wheelock
Trans. Amer. Nucl. Soc. 10 (1), 108-109 (June 1967),
Abstract

14. Technology of Nuclear Fuels

a) The "Technology of Nuclear Fuels" program has only a relatively minor amount of direct plutonium research incorporated at this time, although the emphasis of the program is shifting in that direction. Specifically, the tasks directly involving plutonium have as their objectives: (1) the investigation of the irradiation behavior of UO_2 and UO_2 - PuO_2 at surface temperatures from 1200 to 1900°C; (2) the determination of fission-gas diffusion, solubility and bubble nucleation and agglomeration processes in refractory UO_2 - PuO_2 fuels; (3) the performance of a combined theoretical and experimental effort to define chemical activities and to establish sublimation-condensation mechanisms for candidate uranium-plutonium fuels to provide guidance for selection of fuel compositions and fuel-cladding combinations that might release gas more readily during irradiation.

b) 2 M.Y.

c) Nonmetallic

d) W. Chubb, V. H. Storhok

e) Since the program has only recently changed emphasis toward plutonium work, there are no publications or topical reports from the effort. However, the results have been reported over a period of the last 1-1/2 years in the BCL quarterly report series, Progress on High-Temperature Fuels Technology. BMI-1875 is the most recent issue in this series.

15. Creep of UO_2 - PuO_2

a) The objective of this program is to determine the elevated temperature creep behavior of fuels of interest for liquid-metal fast-breeder reactors (LMFBR). Major emphasis is on UO_2 - PuO_2 fuels, although initial experiments were conducted on UO_2 to demonstrate feasibility of the design of the apparatus. In-pile creep experiments will determine the mechanical behavior of fuel under actual irradiation conditions. Out-of-pile creep experiments on both unirradiated and irradiated specimens are being performed to determine to what extent out-of-pile behavior can be used to predict in-pile creep behavior and to provide information on basic creep mechanisms which can be used to help understand the effect of fissioning on the creep of oxide fuels.

The program involves an initial intensive effort culminating in the first in-pile creep test during the first year. UO_2 - PuO_2 (and UO_2) will be the only fuel considered during the first three years; but other fuels believed to have greater long-range potential for fast-breeder reactors, such as UC-PuC and UN-PuN , will be considered in the fourth and fifth years. Objectives of current tasks are:

To design, fabricate, and operate capsules for determining the in-pile compressive creep behavior of UO_2 - PuO_2 fuels as affected by fission rate, stress, temperature, and burnup.

To generate and analyze creep data obtained from in-pile creep capsules.

To provide the analytical means for determining compressive creep constants of UO_2 - PuO_2 from anisothermal in-pile creep experiments so that they can be compared to isothermal creep constants of unirradiated material, and therefore in-pile creep behavior can be predicted.

To fabricate and characterize UO_2 - PuO_2 specimens for pre-irradiation and in-pile compressive creep experiments.

To determine the amount of primary creep and the minimum compressive creep rates for unirradiated UO_2 - 22 wt % PuO_2 identical to the UO_2 - PuO_2 that will be creep tested in-pile.

To determine reference out-of-pile creep data (including the amount of data scatter) on the enriched UO_2 to be tested in-pile and to determine the effect of specimen geometry on creep behavior.

To determine the primary and secondary compressive creep behavior of unirradiated UO_2 under temperature conditions as close as possible to those experienced by identical UO_2 in in-pile tests in order to compare the obtained data with the in-pile creep data.

To provide a method for predicting the resulting of neutron irradiation on the compressive creep behavior of UO_2 and UO_2 - PuO_2 by combining a program of creep experiments on unirradiated materials with an analysis of the known effects of the fission process on the properties and microstructures of these materials.

- b) 13 M.Y.
- c) Nonmetallic
- d) J. S. Perrin
- e) Progress is reported quarterly in the series entitled Progress on Development of Materials and Technology for Advanced Reactors. The most recent report is BMI-1872.

16. Uranium - Plutonium Mononitride Fuel Materials

a) These studies are being conducted to evaluate the potential of mixed UN-PuN fuels for high-gain, liquid-metal fast-breeder reactor (LMFBR) applications. Emphasis is being placed on the study of solid solution uranium - plutonium mononitrides. The use of mixed nitride fuels in LMFBR applications is desirable because of the high physical and fissile density of the materials, high melting point, high thermal conductivity as compared to that of the oxides, and the high breeding ratio and short doubling time attainable. The program is divided into efforts involving the preparation of materials, property determinations, thermal and fast-flux irradiation studies, and supporting studies. The evaluation of the sodium-bonding concept is also of major importance in the program. Nitride-fueled LMFBR systems are considered to be optimized in their performance when utilizing 1200°F exit sodium and with linear heat ratings on the order of 40 kW/ft with heat fluxes approaching 2×10^6 Btu/hr/ft². These criteria are believed to dictate the use of a sodium-liquid-metal heat transfer medium at the fuel-clad interface to protect against fuel melting and allow for fuel swelling under irradiation without attendant clad distortion. Objectives of current tasks include the following:

To determine those properties pertaining to plutonium - nitride-bearing fuel compositions which allow predictions of fuel stability, transport, and other factors needed to evaluate the behavior of the fuel under possible reactor environmental conditions of both transient and steady-state nature.

To determine those solid-state reactions that occur between UO₂ and U₂N₃-type phases at temperatures of 1500°C and below to determine approximate reaction compositions and isotherms.

To investigate the compatibility of UN-PuN fuels with potential cladding materials under simulated LMFBR conditions.

To develop a reliable analytical method for the determination of nitrogen in the sodium used to bind (U,Pu)N irradiation fuel specimens.

To investigate the self- and interdiffusion behavior in nitride fuels of interest to LMFBR applications.

To fabricate ($U_{0.8}Pu_{0.2}$)N pellets by powder metallurgical techniques for use in irradiation experiments designed to evaluate fuel performance under thermal flux conditions.

To irradiate UN-PuN fuels to burnups of 100,000 MWD/T at fission-heat-generation rates and temperatures typical of sodium-cooled fast-reactor operations.

To evaluate the stability of mixed nitride fuels irradiated in a thermal neutron flux at cladding temperatures up to 700°C, heat generation rates up to 40 kW/ft, and burnups up to 100,000 MWD/T.

To fabricate solid solution ($U_{0.8}Pu_{0.2}$)N pellets by powder metallurgical techniques and clad them in well characterized Type 316 stainless steel for irradiation in the EBR-II.

To design and fabricate irradiation capsules, encapsulate fuel pins, perform required nondestructive examinations, prepare safety analyses for the irradiations, and ship the completed capsules to the EBR-II.

To perform transient irradiation tests on (U,Pu)N fuels in the TREAT reactor.

- b) 22 M.Y.
- c) Nonmetallic
- d) W. M. Pardue
- e) Vaporization Characteristics of Uranium Mononitride
C. A. Alexander, J. S. Ogden, and W. M. Pardue
Submitted for publication in J. Nucl. Matls.

Potential of Mixed Nitrides (U,Pu)N as a Fast Reactor Fuel

W. M. Pardue, A. A. Bauer, and D. L. Keller

Presented at the International Symposium on Ceramic Nuclear Fuels, American Ceramic Society, Washington, D. C., May 6, 1969

Sintering Studies on UN and ($U_{0.8}Pu_{0.2}$)N

M. U. Goodyear, R. A. Smith, and D. E. Kizer
Trans. ANS, 11 (2) (1968), 521

- e) Cladding-Fuel Compatibility, A Compendum of Properties and Characteristics for Selected LMFBR Cladding Materials
D. E. Price, J. M. Fackelmann, J. C. Hebeisen, R. D. Koester, and M. D. Houston
USAEC Report No. BMI-1900, May 15, 1968

Thermodynamics of Interactions in Sodium-Bonded Nitride Fuels
C. A. Alexander, J. S. Ogden, W. M. Pardue, and D. E. Price
Presented at the American Ceramic Society Nuclear Division Meeting, Pittsburgh, Pennsylvania, October 6-9, 1968

Thermodynamics and Mass Transport in the Plutonium-Uranium-Nitrogen System
C. A. Alexander, J. S. Ogden, and W. M. Pardue
Presented at the ANS 1968 Winter Meeting, Washington, D. C., November 10-15, 1968

Irradiation of Mixed Nitride Fuel
A. A. Bauer, E. O. Fromm, J. H. Saling, and V. W. Storhok
Presented at the ANS 1968 Winter Meeting, Washington, D. C., November 10-15, 1968

Sintering Studies on UN and $(U_{0.8}Pu_{0.2})N$
M. U. Goodyear, R. A. Smith, and D. E. Kizer
Presented at the ANS 1968 Winter Meeting, Washington, D. C., November 10-15, 1968

Nitride Fuel-Cladding Compatibility
J. E. Hebeisen, D. E. Price, and D. P. Moak
Presented at the ANS 1968 Spring Meeting, Toronto, Canada, June 9-13, 1968

Nitride Fuel-Cladding Compatibility
J. C. Hebeisen, D. E. Price, and D. P. Moak
Presented at the ANS Winter Meeting, Washington, D. C., November 1968

Nitride Fuel-Cladding Compatibility
D. E. Price and D. P. Moak
Presented at the ANS 1968 Winter Meeting, Washington, D. C., November 10-15, 1968

- e) Synthesis, Fabrication, and Properties of Plutonium Mononitride .
W. M. Pardue, V. M. Storhok, and R. A. Smith
Preparation of Nuclear Fuels, Nuclear Engineering - Part XVIII,
A. D. Tevebaugh and D. L. Keller (Eds.), Chemical Engineering
Progress Symposium Series, Vol. 63 (1967), 142

Fabrication of Uranium-Plutonium Mononitride

M. U. Goodyear and R. A. Smith

Presented at the American Ceramic Society Nuclear Division
Meeting, Urbana, Illinois, October 1967

Compatibility of (U,Pu)N with Reactor Cladding

W. M. Pardue

Presented at the American Ceramic Society Nuclear Division
Meeting, Urbana, Illinois, October 1967

Uranium-Plutonium Mononitride Fuels

W. M. Pardue, R. A. Smith, and F. A. Rough

Presented at the AIIME Topical Meeting on Plutonium, Phoenix,
Arizona, October 1967

17. Dislocation Damping in Plutonium

- a) Measurement of dislocation damping in Pu as a function of temperature. Measurements will be made in the kilocycle range using a longitudinal resonant bar.
- b) 0.5 M.Y.
- c) Metallic
- d) C. R. Heiple

18. Solid State Diffusion

- a) Determination of solid state diffusion coefficients in various plutonium alloys and phases.
- b) 0.75 M.Y.
- c) Metallic
- d) A. L. Rafalski
- e) A. L. Rafalski, M. R. Harvey, D. H. Riefenberg
ASM Trans. Quart. 60, 721 (1967)

M. R. Harvey, A. L. Rafalski, D. H. Riefenberg
ASM Trans. Quart. 61, 629 (1968)

M. R. Harvey, A. L. Rafalski, D. H. Riefenberg
Submitted to ASM Trans. Quart.

19. Elevated Temperature Strength and Ductility of Pu-1 wt. % Ga Alloy

- a) Determination of the effects of iron and nickel on strength and ductility of Pu-1 wt. % Ga alloy.
- b) 0.2 M.Y.
- c) Metallic
- d) S. Beitscher
- e) RFP-1053
S. Beitscher
The Dow Chemical Company, Rocky Flats Division, Nov. 1967

20. Low Temperature Deformation of Pu-1 wt. % Ga Alloy

- a) Study of the mode of plastic deformation in the Pu-1 wt. % Ga alloy at low temperatures (-80 to 196°C). Examination is by metallography, x-ray, hardness, and analysis of stress-strain curves.
- b) 1.0 M.Y.
- c) Metallic
- d) W. N. Seery

21. Pu-Ga Phase Diagram

- a) Investigation of solubilities of gallium in α , β and γ phases of plutonium and study of the mechanism of phase transformation between these phases and the δ phase.
- b) 0.5 M.Y.
- c) Metallic
- d) R. L. Moment

22. Rolling Texture of Pu-Ga Alloys

- a) Comprehensive study of rolling texture of various delta-stabilized Pu-Ga alloys. Determination of rolling textures at various temperatures, recrystallization textures, elastic modulus anisotropy.
- b) 1.0 M.Y.
- c) Metallic
- d) R. L. Moment
- e) The Rolling Texture of Pu-1 w/o Ga
R. L. Moment and J. E. Schindler
RFP-1278, Paper presented at Spring Meeting of AIME
(Pittsburgh, May 1969)

The Rolling Texture of Some Pu-Ga Alloys

J. E. Schindler

RFP-1315, The Dow Chemical Company, Rocky Flats Division,
February 1969

23. Oxidation Kinetics of Bulk Plutonium Metal

a) An ellipsometer will be used to determine the kinetics of the initial stages of oxidation of bulk plutonium metal. The optical constants of films of PuO , $\alpha\text{-Pu}_2\text{O}_3$, $\beta\text{-Pu}_2\text{O}_3$, and PuO_2 will be measured. The influence of gas species, pressure, temperature, etc., on the oxidation behavior of plutonium will be investigated.

b) 1.5 M.Y.

c) Metallic

d) L. A. Lott

e) Ellipsometer Studies of Plutonium Oxidation

D. T. Larson and D. L. Cash

J. of Phys. Chem. 73, 2814 (1969)

A Fortran Program for Ellipsometer Measurements of Surface Films

R. W. Loser and D. T. Larson

RFP-1392, October 1969

Optical Constants of Plutonium at 5461 Angstroms

D. T. Larson and D. L. Cash

J. of Nucl. Materials 24, 232, (1967)

Characteristics of Uranium Oxides Produced by Thermal and Oxygen Ion Bombardment Reactions

D. T. Larson, N. W. Taylor, and K. Terada

RFP-863, January 1967

Ellipsometer Studies on the Cleaning and Oxidation of Uranium

D. T. Larson and N. W. Taylor

RFP-674, June 1966

24. Studies of Physical Properties of Plutonium and Plutonium Compounds

a) Investigation of the electrical and optical bulk and surface properties of plutonium and plutonium compounds will be conducted. Determinations of the electrical resistivity, Hall coefficient, thermoelectric power, and optical constants of a thin film of plutonium oxide will be made.

b) 2.0 M.Y.

c) Nonmetallic

d) L. A. Lott

e) Work Function of Ultrahigh Vacuum Deposited Beryllium Films

R. D. Dixon and L. A. Lott

To be published in J. of Appl. Phys., November 1969

25. Studies of Surface Compounds

a) To study the mechanism of and conditions for growth of plutonium surface compounds and to investigate the properties of those compounds.

b) 4.5 M.Y.

c) Nonmetallic

d) R. O. Adams

e) Thermal Desorption Study of Surface Interactions Between Water and Plutonium Dioxide

ACS National Meeting, September 1969

High Temperature X-ray Investigation of Plutonium Oxidation
J. of Nucl. Materials 30, 340-342 (1969)

Desorption from Plutonium Dioxide

RFP-1248, November 21, 1968

26. Basic Studies of Surface Properties

a) To investigate interactions of various gases and vapors with plutonium surfaces. Specific studies to determine the kinetics and thermodynamic characteristics of the adsorption phenomena are done.

b) 1.5 M.Y.

c) Metallic

d) R. O. Adams

GENERAL ELECTRIC COMPANY
Breeder Reactor Development Operation
Sunnyvale, California

27. CREEP STUDIES OF $(\text{Pu}_{0.2}\text{U}_{0.8})\text{O}_2$

a) Creep properties of mixed oxides of plutonium and uranium in the composition range of interest to LMFBR's are being investigated in a compression test apparatus. These tests will primarily be conducted on coprecipitated $(\text{Pu}_{0.2}\text{U}_{0.8})\text{O}_2$. Test variables include grain size (7-25 μ), pellet density (90-97%), stoichiometry (1.93-2.00), and strain rate. Experimental procedures and test apparatus are the same as those noted in GEAP-10054, "Compressive Creep Characteristics of Stoichiometric Uranium Dioxide," May 1969.

- b) 1 M.Y.
- c) Nonmetallic
- d) W. E. Bailey

GENERAL ELECTRIC COMPANY
Nucleonics Laboratory
Vallecitos Nuclear Center
Vallecitos, California

28. THERMODYNAMIC DATA PROGRAM FOR URANIA-PLUTONIA SOLID SOLUTIONS

a) This program is a thermodynamic study of urania-plutonia solid solutions for fast reactor applications. It is directed primarily toward understanding and controlling the demixing or segregation behavior of plutonium in a realistic temperature environment. Four ways for segregation to occur are by liquid-solid transformation, by thermal diffusion in the solid (Soret Effect), by preferential vapor transport in the gas, or by selective reaction with sodium. Each of these processes is governed by the thermodynamic forces in the system, namely the chemical potentials and the temperature gradient. Consequently, the approach to an understanding is through the determination of the thermodynamic behavior of the solid solution.

Since segregation most likely will occur in the temperature extremes of a fuel element, this study emphasizes the temperature region from about 1600°C to the melting temperature. Specific measurements which are being performed are: Liquidus-Solidus temperatures, component activities of the fuel as a function of composition, oxygen, and plutonium migration in an imposed temperature gradient and sodium-fuel thermodynamics.

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- b) 2 M.Y.
- c) Nonmetallic
- d) E. A. Aitken
- e) A Thermodynamic Data Program Involving Plutonia and Urania at High
Temperatures
E. A. Aitken and S. K. Evans
GEAP-12027 (GE-VNC Quarterly Report No. 7, August 1969).

LAWRENCE RADIATION LABORATORY

P. O. Box 808

• Livermore, California 94550

29. MECHANICAL PROPERTIES

a) To study the creep and torsion behavior of delta-plutonium alloys as a function of temperature, load or strain rate, impurity content and micro-morphology, and to correlate these data with existing mechanical property data.

b) 2.5 M.Y.

c) Metallic

d) J. L. Robbins

e) A. D. Wheeler and J. L. Robbins
J. Nucl. Mat., 32, 57 (1969).

30. X-RAY DIFFRACTION STUDIES

a) To use x-ray diffraction to investigate the micro-morphology (grain size, dislocation density, stacking fault density, etc.) of plutonium and its alloys.

b) 1 M.Y.

c) Metallic

d) P. L. Wallace

31. MISCELLANEOUS (Less than 0.5 MY per year per topic)

a) Thermal expansion of plutonium alloys, diffusion in plutonium, elastic moduli, Pu-Mg phase behavior, neutron diffraction by Pu^{242} .

b) 1 M.Y.

c) Metallic

d) J. L. Robbins

32. Plutonium Phase Diagrams

a) This is a continuing program which has as its goal the determination of the phase relationships in plutonium and in alloy systems containing plutonium as one of the alloying elements. Phase diagrams are constructed from data obtained by optical and X-ray metallography, thermal analysis, and dilatometry.

Currently the binary systems of plutonium with ruthenium, osmium, zirconium, and hafnium are being studied in detail in order to reconcile discrepancies in the various versions of the phase diagrams as published by different investigators; and the unpublished systems of plutonium with the heavy rare-earth elements are being examined. Also, the thermal expansion-vs-composition relationships in the stable, plutonium-gallium delta phase are being determined by the lattice-parameter method.

- b) 4 M.Y.
- c) Metallic
- d) R. D. Fowler, F. W. Schonfeld
- e) Binary Systems of Plutonium with Praseodymium, Neodymium and Samarium
F. H. Ellinger, C. C. Land and K. A. Johnson
J. Nucl. Mat. 29, 178 (1969)

Constitution of Plutonium-Rich Plutonium-Hafnium Alloys
F. H. Ellinger and C. C. Land
J. Nucl. Mat. 28, 291 (1968)

Constitution of Plutonium Alloys
F. H. Ellinger, W. N. Miner, D. R. O'Boyle and F. W. Schonfeld
LA-3870 (1968)

Thermodynamics of the Plutonium Carbides
W. M. Olson and R. N. R. Mulford
Thermodynamics of Nuclear Materials, 1967, IAEA, Vienna, 1968, p. 467; also DTIE, 1967, 19 pp.

The Plutonium-Oxygen and Uranium-Plutonium-Oxygen Systems: a Thermochemical Assessment
R. N. R. Mulford
Contribution to IAEA Panel Report: Technical Report Series No. 79, IAEA, Vienna, 1967

e) Thermodynamics of the Plutonium Carbides

W. M. Olson and R. N. R. Mulford

Gmelin, 1967, 18 p.

The Plutonium-Lanthanum System

F. H. Ellinger, C. C. Land, and K. A. Johnson

Trans. Met. Soc. AIME, 239, 895 (1967)

The Ternary System Plutonium-Cerium-Cobalt

F. H. Ellinger, C. C. Land, K. A. Johnson, and V. O. Struebing

Trans. Met. Soc. AIME, 236, 1577 (1966)

33. Experimental Metal Physics

a) This program is concerned with the electronic and metallurgical behavior of solids, and considerable effort is being directed toward determining the transport properties of pure plutonium, its alloys and compounds. Work that is now in progress includes: positron annihilation experiments to gain information about the electron momentum distribution and the Fermi surface of plutonium; thermal conductivity, electrical resistivity, and thermoelectric power measurements on plutonium compounds, which will shed light on the electronic structures of these systems; investigation of systems and techniques for preparing thin films of plutonium, as a preliminary step toward studying the optical properties of plutonium in order to gain further knowledge about the band structure of plutonium; and low-temperature electrical resistivity measurements on nondilute alloys of plutonium with rare-earth elements, which will reveal further information about the electronic structure of plutonium.

b) 4 M.Y.

c) Metallic

d) R. D. Fowler, F. W. Schonfeld

e) Hall Effect in Oriented Alpha Plutonium

T. R. Loree and H. T. Pinnick

Bull. Am. Phys. Soc., Ser. 2, 14, 306 (1969), Abstract

Thermal Conductivity of Some Delta-Plutonium Alloys

J. F. Andrew

J. Nucl. Mat. 30, 343 (1969)

Transformation-Induced Precipitation of Delta Plutonium in Metastable Cerium-Plutonium Alloys

R. O. Elliott, W. N. Miner, R. B. Roof, Jr., and C. C. Land

J. Less-Common Metals, 15 (1), 106 (1968)

- e) Self-Irradiation Effects in a Cerium Alloy Containing 15 at. % Plutonium
R. O. Elliott, W. N. Miner, F. W. Clinard, Jr.
Rare Earth Research Conf. 6th, Gatlinburg, Tenn.,
May 1967. Proc. p. 405

Self-Irradiation Effects in a Cerium Alloy Containing 15 at. % Plutonium
R. O. Elliott, W. N. Miner, and F. W. Clinard, Jr.
J. Less-Common Metals, 13 (3), 274 (1967)

Annealing of Self-Irradiation Induced Defects in Alpha Plutonium Stored at 87°K
W. N. Miner and R. O. Elliott
J. Nucl. Mat. 23, 115 (1967)

Magnetic Susceptibility of Delta-Plutonium Alloys
J. L. Lunsford and E. A. Kmetko
Plutonium 1965, Kay and Waldron, Eds., Chapman and Hall, London (1967), pp. 214-21

Self-Irradiation of Plutonium and Its Alloys
R. O. Elliott and C. E. Olsen
Studies in Radiation Effects, Series A, Physical and Chemical, Vol. 1, Gordon and Breach, New York (1966), pp. 1-71

Thermal Conductivity of Plutonium Metal
J. F. Andrew
J. Phys. and Chem. Solids, 28, 577 (1967)

34. Crystal Structure Determinations

- a) This program is a long-term effort directed toward the detailed determination of the crystal structures of plutonium-containing compounds, mainly through X-ray studies of single crystals.

Currently under investigation are the three different single crystals, characterized by cubic, hexagonal, and rhombohedral unit cells, that have been found in the phase B region of the cerium-plutonium-cobalt alloy system at about the composition $\text{Ce}_5\text{Pu}_2\text{Co}_3$. Several compounds, Y_2Zn_{19} , $\text{Gd}_2\text{Zn}_{19}$, and Y_2Cd_{19} , that are similar to the plutonium-zinc η phase, $\text{Pu}_2\text{Zn}_{19}$, but which do not show the same superlattice characteristics that the plutonium-zinc compound does, are being examined in order to elucidate the structures of

this type of compound. Also, the anomalous scattering factors and the photoelectric absorption cross sections are being computed for plutonium, as part of a larger computational program directed toward calculating these properties for all the elements, and involving a more rigorous treatment than has been used previously. A comprehensive atlas of all the presently known crystal structures, including those involving plutonium, is being prepared in cooperation with Professor W. B. Pearson.

- b) 1.5 M.Y.
- c) Metallic
- d) R. D. Fowler, F. W. Schonfeld
- e) Compton Scattering Factors for Aspherical Free Atoms
D. T. Cromer
J. Chem. Phys. 50 (11), 4857 (1969), also LA-4079 (1969)

Compton Scattering Factors for Spherically Symmetric Free Atoms
D. T. Cromer and J. B. Mann
J. Chem. Phys. 47 (6), 1892 (1967); also LA-3689 (1967)

The Crystal Structure of $\text{Pu}_2\text{Zn}_{\sim 9}$
A. C. Larson and D. T. Cromer
Acta Cryst. 23, 70 (1967)

Crystal Structure of Beta Plutonium
W. H. Zachariasen and F. H. Ellinger
(Abstract), American Crystallographic Association Annual Meeting, MIT, Cambridge, Mass., March 1963. Program and Abstracts, p. 23

Crystal Structures of PuNi_3 , CeNi_3 , and Ce_2Ni_7
D. T. Cromer, A. C. Larson, and C. E. Olsen
(Abstract), American Crystallographic Association Annual Meeting, Cornell University, Ithaca, N. Y., July 1959. Program and Abstracts, p. 34

Crystal Structure of $\text{Pu}_2\text{Zn}_{\sim 9}$
A. C. Larson and D. T. Cromer
(Abstract), Acta Cryst. Suppl. 21, A84 (1966)

35. Defects in Plutonium Metal, Alloys and Compounds

a) Studies of irradiation induced defects in actinide metal oxides are being undertaken. Films for transmission electron microscopy have been prepared by electrothinning and by evaporation, and

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equipment for ion bombardment thinning and cathodic etching is under construction. Pure plutonium, delta-phase plutonium alloys, and PuO_2 have been examined, and these materials as well as others are now under study after having been irradiated with alpha particles from a $^{238}\text{PuO}_2$ source.

- b) 0.5 M.Y.
- c) Nonmetallic
- d) R. D. Fowler, F. W. Schonfeld
- e) Grain Growth in Thin Films of ThO_2 , NpO_2 , and PuO_2
S. E. Bronisz and D. L. Douglass
Phys. Stat. Sol. 29, K95 (1968)

Discussion of 'Transmission Electron Microscopy of Pu-1 wt % Ga' by A. W. Brewer and R. J. Fraikor
D. L. Douglass and S. E. Bronisz
J. Nucl. Mat. 23, 107 (1967)

Modified Techniques for Preparing Specimens of Alpha-Active Materials
D. L. Douglass
DTIE, 1967, 10 pp.

36. Compatibility of Plutonia and Other Ceramics with Metallic Materials

a) This project concerns the evaluation of the compatibilities of various metallic container materials with radioisotopic heat source ceramics and cermets that are being developed for space electric power applications. Encapsulated pellets, about $\frac{1}{8}$ in. dia by $\frac{1}{8}$ in. thick, of the materials under test are held for several thousands of hours at temperatures as high as 1800°C , and then are examined by optical and X-ray metallography, electron microscopy, and electron microbeam analysis to detect and identify any reactions that may have taken place as a result of the treatment. Currently, the compatibilities of $^{238}\text{PuO}_2$, $^{238}\text{PuO}_2\text{-ZrO}_2$ and $^{238}\text{PuO}_2\text{-ThO}_2$ solid solutions, and $^{238}\text{PuO}_2\text{-ThO}_2\text{-Mo}$ cermets with TZM, tantalum-tungsten and platinum-rhodium alloys, and molybdenum are under investigation, but other materials are likely to be included as the fuel development program progresses.

- b) 1 M.Y.
- c) Nonmetallic
- d) R. D. Baker, F. W. Schonfeld

37. Helium Behavior in Isotopic Fuels ($^{238}\text{PuO}_2$)

a) This project, which pertains mainly to ^{238}Pu -bearing materials, embraces three research areas: helium release behavior, the effects of alpha self-irradiation on crystallite size and lattice strain, and the relation of alpha particle bombardment to structural defects.

In the first area, the kinetics of helium release are being studied from both fundamental and engineering standpoints. This work mainly involves isothermal and temperature-ramp experiments on pure $^{238}\text{PuO}_2$ and on $^{238}\text{PuO}_2$ - ThO_2 solid solutions and cermets (e.g. with molybdenum). Most of the information obtained so far has had engineering value and has served to define the problem. Considerably more work will be required to provide understanding of the complex release process.

In the second area, a computer program has been developed for the purpose of analyzing the data from an X-ray diffractometric study of a high-purity $^{238}\text{PuO}_2$ disk in order to determine the changes in the effective crystallite size and strain parameters that result as self-irradiation damage and the concentration of helium increase while the sample ages at room temperature. Over the period of several months that this study has been in progress, a slight increase in the lattice parameter of the $^{238}\text{PuO}_2$ has been observed, but, because of the slow rate at which the change is taking place, no X-ray line broadening that can unequivocally be associated with a change in crystallite size has yet been detected.

In the third area, a study concerned with the effect of 5 MeV alpha-particle bombardment, in a Van de Graaff accelerator, on ThO_2 is nearing completion, and the experience thus gained will be of considerable value in conducting a somewhat similar study of radiation induced defects in PuO_2 . Principally, spallation (fracture) effects, lattice dilation phenomena, and atomic scale damage effects are being investigated. Since ThO_2 and PuO_2 both have the fluorite crystal structure, comparison of the results for the two materials should be enlightening.

- b) 1.5 M.Y.
- c) Nonmetallic
- d) R. D. Baker, F. W. Schonfeld

38. Solidification and Transformation Studies on Pu

- a) The objective of this program is to study the effect of the solidification and transformation rates and mechanisms upon the properties of cast, high purity Pu metal. Chill casting and subsequent heat treating and quenching by unusual and "drop smashing" methods are being investigated.
- b) 2 M.Y.
- c) Metallic
- d) R. D. Baker, W. J. Maraman
- e) Unusual Problems Encountered in Casting High Density-High Purity Plutonium
D. R. Harbur, J. W. Anderson, and W. J. Maraman
Modern Castings, 53, 60 (1968)
- Rapid Quenching Drop Smasher
D. R. Harbur, J. W. Anderson, and W. J. Maraman
Trans. Met. Soc. of AIME, 245, 1055 (1969)
- Preparation of Sound High Purity Plutonium Rods. Pt. I. Effect of Chill Casting and Subsequent Heat Treatment on Micro-cracking
D. R. Harbur, J. W. Romero, J. W. Anderson, and W. J. Maraman
J. Nucl. Materials, 25, 160 (1968)
- The $\alpha \rightarrow \beta$ Transformation in High Purity Plutonium
D. R. Harbur, J. W. Anderson, and W. J. Maraman
J. Nucl. Materials, 30, 289 (1968)
- Preparation of Sound High Purity Plutonium Rods. Pt. II. Observed Phase Transformations During Quenching
D. R. Harbur, M. C. Fleming, J. W. Anderson, and W. J. Maraman
J. Nucl. Materials (in press)

39. Ceramic Plutonium Fuel Materials

- a) This is a continuing program on the synthesis, fabrication and characterization of plutonium fuel materials. Compounds being investigated include plutonium monocarbide, plutonium sesquicarbide, plutonium dicarbide, plutonium dioxide, plutonium sesquioxide, plutonium mononitride and plutonium sulfide, as well as solid solutions of most of these compounds with the corresponding uranium compound. Anion solid solutions such as carbonitrides also are to

be investigated. Complete chemical analyses are made for major constituents, trace impurities, constancy of composition, and phase relationships by microprobe examination.

The principal goal is to determine high temperature properties of well characterized pure materials. Properties that are being determined include thermal conductivity, thermal stability, transformation by thermal and differential thermal analysis, structure by x-ray, high temperature x-ray diffraction and by neutron diffraction, thermal expansion, density, hardness as a function of temperature, plastic deformation, compatibility utilizing electron microprobe analysis, and self-irradiation damage. In addition to phase equilibria and general properties, specific thermodynamic properties such as free energy of formation by vaporization equilibria measurement in the 1200° to 2000°C temperature range with mass spectrometer identification of vapor species, free energy of formation by electromotive force measurement in the 450° to 1200°C temperature range, and heat capacity and heat of transformation at temperatures up to 2900°C are being determined. High temperature physical properties also are determined.

Development of new chemical methods of analysis and improvements in existing methods with emphasis on precision will be continued and applied to the characterization of fuel candidate materials. Electron microprobe examinations and x-ray diffraction measurements will be included. Of particular importance to mixed oxide fuels is a precise method for determining O/M ratios.

- b) 18 M.Y.
- c) Nonmetallic
- d) R. D. Baker, J. A. Leary
- e) Refractory Plutonium Fuel Materials

J. A. Leary, W. C. Pritchard, R. L. Nance and M. W. Shupe
Plutonium 1965, Proceedings of the Third International
 Conference on Plutonium, Institute of Metals, Chapman and
 Hall, pp. 639-653, London (1965)

Thermodynamic Properties of Plutonium Mononitride from Electro-
 motive Force Measurements

G. M. Campbell and J. A. Leary
 J. Phys. Chem. 70, pp. 2703-2705 (1966)

Recent Development in High-Temperature Plutonium Compounds

J. A. Leary
 Trans. Amer. Nucl. Soc. 9, p. 37 (1966)

- e) Preparation and Properties of Plutonium Mononitride and Uranium Mononitride-Plutonium Mononitride Solid Solutions
J. A. Leary, R. L. Nance, W. C. Pritchard, J. G. Reavis,
M. W. Shupe, and A. E. Ogard
American Ceramic Society, Portland, October (1966)

Refractory Plutonium Fuel Materials

J. A. Leary, et al.
3rd Int. Conf. on Plutonium, London, Nov. 22-26, 1965,
639-53, Inst. Metals (1967)

A Low Temperature Magnetic Transition in Plutonium Monocarbide

J. L. Green, G. P. Arnold, J. A. Leary, and N. G. Nereson
J. Nucl. Materials, 23 (1967)

Phase Relations in the High-Carbon Portion of the U-Pu-C System

J. G. Reavis, M. W. Shupe, C. W. Bjorklund, and J. A. Leary
Trans. Amer. Nucl. Soc. 10, 111 (1967)

High-Temperature Heat Content and Heat Capacity of Uranium Dioxide and Uranium Dioxide-Plutonium Dioxide Solid Solutions

A. E. Ogard and J. A. Leary
Thermodynamics of Nuclear Materials, 1967, Inter. Atomic
Energy Agency, Vienna (1968)

Thermodynamic Properties of Plutonium Compounds by Electromotive Force Techniques

G. M. Campbell, L. J. Mullins and J. A. Leary
Thermodynamics of Nuclear Materials, 1967, Inter. Atomic
Energy Agency, Vienna (1968)

Crystallography and Magnetic Ordering in Plutonium Carbides by Neutron Diffraction

J. L. Green, G. P. Arnold, J. A. Leary, and N. G. Nereson
Bull. Amer. Cer. Soc. 46, 893 (1967)

Self-Irradiation Damage in Plutonium Ceramics

C. W. Bjorklund, R. M. Douglass and J. A. Leary
Bull. Amer. Cer. Soc. 46, p. 901 (1967)

Thermodynamic Properties of PuN by Galvanostatic Potential Determination

G. M. Campbell
J. Phys. Chem., in press (1968)

- e) Crystallography and Magnetic Ordering in Plutonium Carbides by Neutron Diffraction

J. L. Green, G. P. Arnold, J. A. Leary, and N. G. Nereson
J. of Nucl. Materials, in press (1968)

Mass Spectrometric Studies of Plutonium Compounds at High Temperatures. III. The Vapor Pressure of Plutonium

R. A. Kent

J. of High Temp. Sci. (in press)

Mass Spectrometric Studies of Plutonium Compounds at High Temperatures. IV. The Vaporization of PuN

R. A. Kent and J. A. Leary

J. of High Temp. Sci. (in press)

Present Status of the Uranium-Plutonium-Carbon Phase Diagram

J. A. Leary

Amer. Cer. Soc. Mtg., Wash., D. C. (May 3-8, 1969)

Determination of Oxygen in Refractory Oxides

C. S. MacDougall, M. E. Smith, and G. R. Waterbury

Anal. Chem. 41, 372 (1969)

Combustion Micro-Monometric Determination of Carbon in Plutonium Sulfide

C. S. MacDougall, M. E. Smith, and G. R. Waterbury

LA-4047-MS, 1968. (Presented at Twelfth Conf. on Analy. Chem. in Nuclear Technology, Oct. 8-10, 1968, Gatlinburg, Tenn.)

An Evaluation of Two Methods for Determining Uranium and Plutonium in Ceramic Reactor-Fuel Materials

G. B. Nelson, K. S. Bergstresser, G. R. Waterbury, and C. F. Metz

Twelfth Conf. on Analy. Chem. in Nuclear Technology, Oct. 8-10, 1968, Gatlinburg, Tenn.

Studies on the Formation of Pu_2O_3 in the Sintering of PuO_2

W. C. Pritchard and R. L. Nance

LASL Report No. LA-3493 (1966)

Mass Spectrometric Studies of Plutonium Compounds at High Temperatures. I. The Heats of Vaporization of Gold and Plutonium and the Heat of Decomposition of Plutonium Mononitride

R. A. Kent and J. A. Leary

LASL Report No. LA-3902 (1968)

- e) Thermodynamic Properties of PuN by Galvanostatic Potential Determination
G. M. Campbell
LASL Report No. LA-3945 (1968)

A Computer Program for Calculation of Particle Size Distribution from Sedimentation Data
J. D. Olson and G. M. Campbell
LASL Report No. LA-4001 (1968)

Differential Thermal Analysis Apparatus for Observation of Refractory Plutonium Compounds
J. G. Reavis, J. F. Buchen, and J. A. Leary
LASL Report No. LA-4103 (1969)

The Determination of Free (Uncombined) Carbon in Plutonium Carbides
T. K. Marshall and G. R. Waterbury
LA-3603 (1966)

The Potentiometric Titration of Cerium and Manganese Following Separation from Plutonium and Cobalt
R. G. Bryan and G. R. Waterbury
LA-3402 (1966)

40. Preparation and Evaluation of Plutonium-238 Radioisotopic Heat Sources

a) The successful utilization of plutonium-238 in a radioisotopic heat source for biomedical use depends on demonstrating that an economic fuel can be produced reliably in pure form, and that the fuel capsule can be used safely. The primary requirements are minimal external radiation and reliable containment of radioactive material.

Fuel composition depends mainly on the operating temperature of the fuel capsule. For relatively low temperature applications, a plutonium alloy will be satisfactory. However, for systems that require fuel temperatures in excess of about 600°C, a ceramic fuel form will be required. Results obtained in this project during FY 1968 and FY 1969 have shown that the external radiation from metal sources can be reduced to acceptable levels by elimination of light element impurities commonly present in conventional

plutonium-238. Moreover, neutron and photon emission rates in ceramic compositions are excessive unless target nuclides such as oxygen-18 and nitrogen-14 that are present in the natural elements are eliminated from the oxide or nitride fuel.

Metal and ceramic compositions are being synthesized and characterized in small scale (4-10 watt) preparations in order to select the best fuel form for the artificial heat application. These materials are evaluated by determining properties with screening tests that include chemical purity, reliability of synthesis, fabricability, external radiation, power density, behavior on thermal cycling, melting point, helium release, and compatibility with container materials. Based on experience obtained in the screening phase, several 30-watt power sources will be fabricated with the preferred fuel composition by the best process in order to evaluate fabricability and to provide preliminary cost data on a practical scale of operation. The shelf life and operating life characteristics of these prototypical capsules will be determined. In addition, the advantages of utilizing plutonium that contains less than 1 ppm of plutonium-236 are being evaluated.

Reliable synthesis of the fuel material and fabrication of source capsules will be demonstrated by producing a few dozen capsules on an interim basis. This interim production experience will serve as a basis for providing assistance to the Commission and its designated contractors in order to ensure the timely commercial production of reliable radioisotopic heat sources. Concurrently, development of improved and alternative power source materials will be continued.

- b) 9 M.Y.
- c) Metallic
- d) R. D. Baker, J. A. Leary
- e) Refined Plutonium-238 for Biomedical Applications
L. J. Mullins and J. A. Leary
Trans. Amer. Nucl. Soc. 11, 91-92 (1968)

Plutonium-238 for Biomedical Applications
L. J. Mullins and J. A. Leary
Nucl. Applications, 6, 287-297 (1969)

Plutonium-238 Materials as Radioisotopic Heat Sources for Human Use
L. J. Mullins, G. M. Matlack, R. L. Nance, C. F. Metz, and
J. A. Leary
Amer. Nucl. Soc. Mtg., Seattle, Washington (June 1969)

- e) Neutron and Gamma Radiations from ^{238}Pu
J. Bubernak, G. M. Matlack, and C. F. Metz
Trans. Amer. Nucl. Soc. 11, 457 (1968)

Ion Exchange Extraction Separation and Radiochemical Determination of Neptunium-237 in Plutonium-238
J. Bubernak, M. Lew, and G. M. Matlack
Twelfth Conf. on Anal. Chem. in Nucl. Technol.,
October 8-10, 1968, Gatlinburg, Tenn.

Detection of Light Element Impurities in High Purity Plutonium-238 by Gamma Spectroscopy
J. Bubernak, M. Lew, and G. M. Matlack
Twelfth Conf. on Anal. Chem. in Nuclear Technology,
October 8-10, 1968, Gatlinburg, Tenn.

Neutron and Gamma Radiations from Plutonium-238
J. Bubernak, G. M. Matlack, and C. F. Metz
Trans. Amer. Nucl. Soc. 11, 457 (1968)

Plutonium-238 Materials as Radioisotopic Heat Sources for Human Use
L. J. Mullins, G. M. Matlack, R. L. Nance, C. F. Metz, and J. A. Leary
Trans. Amer. Nucl. Soc. 12, 47 (1969)

Radiation Characteristics of Plutonium-238
G. M. Matlack and C. F. Metz
LA-3696 (1967)

41. Plutonium-238 Space Electric Power Fuel Development

a) The primary objective of this project is to develop advanced plutonium-238 radioisotopic heat source materials for space electric power applications. Four categories of fuel compositions are to be investigated:

- . Sintered monolithic $^{238}\text{PuO}_2$ (The formula " $^{238}\text{PuO}_2$ " includes $^{238}\text{PuO}_{2-x}$.)
- . $^{238}\text{PuO}_2\text{-MO}_2$ solid solution (M = Th or Zr)
- . Molybdenum - $^{238}\text{PuO}_2$ cermets
- . Alternate $^{238}\text{PuO}_2$ fuels, combining advantages of both solid solutions and cermets.

In view of the fact that numerous fuel forms are to be developed simultaneously, program schedules and specific objectives must remain flexible in order to allow increased emphasis on the best fuel type as development progresses.

Modular segments of each fuel composition will be fabricated and evaluated with regard to intended application. Typically, the segment consists of a disc of the order of 0.5 to 2.5 in. in diameter and 0.2 to 0.5 in. tall. In addition to modular segments, special shapes will be fabricated as required for product characterization in order to establish quality assurance under operating environments. Specific properties to be determined include the following:

- crystallography
- microstructure (including electron microprobe examination)
- chemical composition, purity, and uniformity
- density, and uniformity of density
- thermal analysis under oxidizing, reducing, and vacuum environments
- helium release rate
- thermal diffusivity
- thermal conductivity
- thermal shock behavior
- mechanical properties, including fragment analysis
- rate of dissolution in surface waters
- compatibility with potential clads
- external radiation
- basic thermodynamic properties including vapor pressure
- radiochemical changes during fuel lifetime (theoretical)

An important aspect of the characterization of fuel materials is the continuing theoretical and experimental work required to assess the effects of fuel aging on fuel properties. Methods for reducing external radiation from fuels, such as by use of oxygen-16, also will be investigated theoretically and experimentally.

Additional fabrication and properties evaluations will be done in order to establish product performance under potential accident environments. Such properties include impact behavior, solubility in surface waters, and the effects of excessive thermal environments associated with launch pad fireball and reentry conditions.

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- Monolithic $^{238}\text{PuO}_2$ and of $^{238}\text{PuO}_2\text{-MO}_2$ solid solution fuel forms will be synthesized and fabricated by the economical method of powder blending, cold pressing, and sintering. Synthesis of molybdenum cermetts involve first either (1) coating $^{238}\text{PuO}_2$ microspheres by chemical vapor deposition in a fluidized bed reactor, or (2) coating PuO_2 sintered particles followed by hot pressing a compaction of coated oxide to high density. Methods for synthesizing and fabricating alternate fuels will depend on fuel compositions and requirements.

In addition to the fabrication of specimens for evaluation, selected fuel forms will be produced on a pilot plant scale in order to establish quality control and assurance, as well as to provide a basis for reliable cost estimates for eventual commercial production. Assistance in establishing commercial capabilities will be provided as requested by the Commission.

A second phase of this project involves a fundamental study of helium release. Gross kinetics of helium release, as well as helium precipitation and changes in crystallographic characteristics will be investigated theoretically and experimentally.

- b) 9 M.Y.
- c) Nonmetallic
- d) R. D. Baker, J. A. Leary
- e) Plutonium-238 Pyrolytic Coated Particles
W. J. McCreary and W. C. Pritchard
70th Annual Meeting of the American Ceramic Society,
Chicago, Illinois (1968)

The LASL Plutonium-238 Fuel Development Program

J. A. Leary

Given at U.S.A.E.C. Plutonium-238 Isotopic Fuels and Heat
Source Program Review held in Washington, D. C.,
Oct. 29-30, 1968

Operating Experience on an Economical Plutonium-238 Processing Facility

M. W. Shupe, et al.

IAEA Symp. on Radiation Safety Problems in the Design and
Operation of "Hot" Facilities, Saclay, France (Oct. 13-17,
1969)

The LASL Plutonium-238 Fuel Development Program SEPO/SNS/AEC Document CMB-11-9455 (Oct. 1968)

42. Electrometallurgy and Molten Salt Chemistry of Plutonium

a) Three general areas of research are included in this program:

1. Electrorefining of liquid plutonium in molten salt systems
2. Thermodynamic properties of selected elements in liquid plutonium
3. Liquid plutonium -- molten salt equilibrium studies

In addition, research on preparation of neptunium and americium metal has been conducted.

Electrorefining studies have been directed toward developing a reliable process for producing high purity plutonium metal on the multikilogram scale from impure plutonium, including gallium alloys. In addition to the electrochemical aspects, the behavior of impurity elements during casting and sample has been characterized. High purity plutonium has been produced and supplied to most of the plutonium laboratories in the U.S. and U.K. Currently the NBS distributes high purity plutonium prepared under this program as a chemical standard. Recent efforts have been devoted to electrorefining special isotopic compositions such as plutonium-242 for use in basic plutonium research.

The equilibrium solubilities of selected elements such as carbon, vanadium, chromium, titanium, manganese, niobium, zirconium, molybdenum, rhenium, thulium, tantalum, and tungsten in liquid plutonium and in liquid plutonium-gallium alloys have been determined experimentally. The results have been treated theoretically using several liquid state models. The results have been of direct application to selection of container materials, alloying of tracer elements with plutonium, and phase diagram studies.

The chemistry of molten salts, particularly when in contact with plutonium metal, have been studied extensively. These studies have led to a detailed knowledge of phase relationships and thermodynamic properties in these systems. A process for selectively removing americium from liquid plutonium was based on these results.

A process for producing high purity americium metal by reduction of americium oxide with lanthanum metal, followed by vacuum distillation of americium, was developed and used to produce metal on the multigram scale.

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- b) 6 M.Y.
- c) Metallic
- d) R. D. Baker, J. A. Leary
- e) Practical Application of Thermodynamics to Plutonium Process Reactions at High Temperature
J. A. Leary and L. J. Mullins
Symp. on Thermodynamics with Emphasis on Nuclear Materials and Atomic Transport in Solids, Vienna, July 1965

Partial Phase Diagram of the $\text{PuCl}_3\text{-PuOCl}$ System
J. G. Reavis and J. A. Leary
J. Inorg. Nucl. Chem. 28, pp. 1205-1208 (1965)

The Solubilities of Refractory Elements in Liquid Plutonium
D. F. Bowersox and J. A. Leary
Trans. Amer. Nucl. Soc. 9, p. 6 (1966)

The Solubilities of Carbon, Tantalum, Tungsten, and Rhenium in Liquid Plutonium
D. F. Bowersox and J. A. Leary
J. Nucl. Materials, 21, 219-224 (1967)

The Solubilities of Selected Elements in Liquid Plutonium
D. F. Bowersox and J. A. Leary
Trans. Amer. Nucl. Soc. 10, 106 (1967)

Distribution of Americium Between Liquid Plutonium and a Fused Salt. Evidence for Divalent Americium
L. J. Mullins, A. J. Beaumont, and J. A. Leary
J. Inorg. Nucl. Chem., 30, 147 (1968)

The Solubilities of Selected Elements in Liquid Plutonium II. Titanium, Vanadium, Chromium, Manganese, Zirconium, Niobium, Molybdenum, and Thulium
D. F. Bowersox and J. A. Leary
J. of Nucl. Materials, 27, 181-185 (1968)

Plutonium
J. A. Leary and L. J. Mullins
Encyclopedia of Chemical Technology, 15, 879-896 (1968)

Electrometallurgy of Plutonium
J. A. Leary and L. J. Mullins
Presented at Electrometallurgy Symp. held by AIME, Cleveland, Ohio, Dec. 2-3, 1968

e) Vacuum and the Actinide Elements

K. W. R. Johnson

Presented to Oklahoma Section of the American Vacuum
Society, Oklahoma City, Okla., Dec. 12, 1968

The Partial Molar Enthalpies of Solution for Tungsten and
Tantalum in Liquid Rare-Earth Metals

D. F. Bowersox

J. of Nucl. Mater., in press

Thermodynamics of Solution of Selected Elements in Liquid
Plutonium

D. F. Bowersox and J. A. Leary

J. of Nucl. Mat. (in press)

Thermodynamic Properties of Selected Solutes in Liquid Metal
Solutions

D. F. Bowersox

1969 Nuclear Metallurgy Symp., V. 15, "Reprocessing of
Nuclear Fuels"(in press)

Molten Salt Method of Separation of Americium from Plutonium

L. J. Mullins and J. A. Leary

Patent No. 3,420,639 (Jan. 1969)

Preparation of Ultra High Purity Plutonium

J. A. Leary and L. J. Mullins

IASL Report No. LA-3356-MS (1965)

The Solubilities of Selected Elements in Liquid Plutonium VI.
Rhenium

D. F. Bowersox and J. A. Leary

IASL Report No. LA-3359 (1965)

The Solubilities of Selected Elements in Liquid Plutonium VII.
Niobium

D. F. Bowersox and J. A. Leary

IASL Report No. LA-3436 (1966)

The Solubilities of Selected Elements in Liquid Plutonium
VIII. Molybdenum

D. F. Bowersox and J. A. Leary

IASL Report No. LA-3523 (1966)

- e) Distribution of Americium between Liquid Plutonium and a Fused Salt. Evidence for Divalent Americium
L. J. Mullins, A. J. Beaumont and J. A. Leary
LASL Report No. LA-3562 (1966)

The Solubilities of Selected Elements in Liquid Plutonium
IX. Vanadium

D. F. Bowersox and J. A. Leary
LASL Report No. LA-3594 (1966)

The Solubilities of Selected Elements in Liquid Plutonium
X. Thulium

D. F. Bowersox and J. A. Leary
LASL Report No. LA-3623 (1966)

The Solubilities of Selected Elements in Liquid Plutonium
XI. Zirconium

D. F. Bowersox and J. A. Leary
LASL Report No. LA-3742 (1967)

The Solubilities of Selected Elements in Liquid Plutonium
XII. Chromium

D. F. Bowersox
LASL Report No. LA-3850 (1967)

The Enthalpies of Selected Elements in Liquid Plutonium
D. F. Bowersox and J. A. Leary

LASL Report No. LA-3951 (1968)

The Solubilities of Selected Elements in Liquid Plutonium
XIII. Titanium and Manganese

D. F. Bowersox
LASL Report No. LA-3995 (1968)

The Solubilities of Selected Elements in Liquid Plutonium
XIV. Niobium in Gallium-Plutonium Alloys

D. F. Bowersox
LASL Report No. LA-4078 (1968)

43. ^{242}Pu Investigations

- a) High purity electro-refined ^{242}Pu is now available in reasonable quantities and will be characterized by X-ray diffraction, dilatometry, thermal analysis, measurement of electrical properties over

a wide temperature range, etc. The purpose of the work is to establish the basic characteristics of Pu metal in a relatively damage-free state.

- b) 1 M.Y.
- c) Metallic
- d) R. D. Fowler, F. W. Schonfeld

44. Mechanical Property Investigations

a) This project includes two research areas. One concerns the determination of internal friction in plutonium, and the other has to do with the development of a technique to orient or index single-crystal rotation photographs of alpha plutonium.

The internal friction work currently involves the design and construction of instrumentation that will utilize a beam vibrating in the first mode, held at the nodes, and driven in the center by an electrostatic probe. The drive/pick-up transducer is of novel design and is expected to be reasonably quantitative. The transducer circuitry, which employs a bridge input, allows measurement of the probe-to-specimen separation as well as the peak-to-peak displacement of the specimen. Thus, this transducer allows measurement of the mechanical impedance facing the probe. In order to accommodate the large changes in volume that occur in plutonium specimens during transformation, the probe-to-specimen separation will be servo controlled. The support electronics are being designed along phase lock principles to attain feedback control of the drive frequency, and, in this manner phase locked resonance dwell will also be achieved.

Because we have been unable to index unequivocally Laue back-reflection photographs of individual grains of alpha plutonium in studies of slip, we are developing a new technique for indexing a combined Laue-oscillation pattern. This treatment permits the construction of a crystal model of the reciprocal lattice and provides for indexing by use of a computer-interactive oscilloscope display.

- b) 1 M.Y.
- c) Metallic
- d) R. D. Fowler, F. W. Schonfeld
- e) Diffusion in Stabilized Delta Plutonium. II. Aluminum
R. E. Tate, G. R. Edwards and E. A. Hakkila
J. Nucl. Mat. 29, 154 (1969)

- e) Diffusion in Stabilized Delta Plutonium. I. Gallium
G. R. Edwards, R. E. Tate, and E. A. Hakkila
J. Nucl. Mat. 25, 304 (1968)

Slip in Alpha Plutonium

S. E. Bronisz and R. E. Tate

Plutonium 1965, Kay and Waldron, Eds., Chapman and Hall,
London (1967), pp. 558-63

45. Vaporization of Plutonium Oxides

a) The goal of this project is to provide a basic understanding of the vaporization process in plutonium oxides, and the experimental work mainly involves mass spectrometry and Knudsen effusion measurements. Although sufficient data are now available to permit quantitative determination of equilibrium vapor species as a function of composition and temperature, a completely satisfying thermodynamic description that can embrace all the data has yet to be discovered. In order to aid in solving this problem, many independent determinations concerning the thermodynamics of plutonium vaporization will be useful. Currently, a series of mass spectrometric measurements of the isomolecular exchange reactions of PuO(g) with other gaseous oxides is being undertaken. These reactions are of the type:

Measurement of ion intensities for all four species in the mass spectrometer yields the equilibrium constant, K , and the temperature dependence of K yields the heat of reaction. Thus, the heat of formation of PuO(g) may be compared with that of MO(g) . So far, M has included gadolinium and neodymium.

- b) 1 M.Y.
c) Nonmetallic
d) R. D. Fowler, F. W. Schonfeld
e) Thermodynamic Properties of Plutonium Oxides
R. N. R. Mulford and C. E. Holley, Jr.
DTIE, 1967, 41 pp.

Pressures over Pu Oxides

R. N. R. Mulford

CMF-5-A-25, January 26, 1968

46. Theoretical Metal Physics

a) In connection with a continuing program devoted to studies of the theoretical aspects of the electronic structures of solids, a relativistic, self-consistent calculation of the band structure of plutonium is being undertaken. This particular calculation will be a rather large effort, in terms of the computer time and facilities required, and is expected to tie in directly with the wealth of experimental data that is available.

Also underway is a study that considers the effects of changes in the topology of the Fermi surface on the thermodynamic properties of the electron gas. From this work, it now seems evident that it will be possible to utilize features of the Fermi surface to provide a mechanism which will explain some of the peculiar properties of delta plutonium, such as the negative thermal expansion coefficient, the algebraic increase in the expansion coefficient as the electron-to-atom ratio is lowered, the stabilization accompanying a lowered electron-to-atom ratio, and the shrinkage of the phase field by applied hydrostatic pressure as well as by an increase in the electron-to-atom ratio.

b) 1.5 M.Y.

c) Metallic

d) R. D. Fowler, F. W. Schonfeld

e) Electronic Band Structure of Plutonium and Its Monocarbide

E. A. Kmetko and J. T. Waber

Plutonium 1965, Kay and Waldron, Eds., Chapman and Hall,
London (1967), pp. 244-73

Band Structure, Specific Heat, Magnetic Susceptibility, and
Anomalous Thermal Expansion of Delta Plutonium

E. A. Kmetko

Plutonium 1965, Kay and Waldron, Eds., Chapman and Hall,
London (1967), pp. 222-43

Self-Consistent Field Hartree Calculations for Atoms and Ions

A. C. Larson and J. T. Waber

LA-4297, Dec. 1969

47. HELIUM RELEASE FROM $^{238}\text{PuO}_2$ FUEL FORMS

a) This program will include helium release studies from $^{238}\text{PuO}_2$ sol gel microspheres and from $^{238}\text{PuO}_2\text{-ThO}_2$ solid solution sol gel microspheres.

Most of the experimental effort will be directed to determining the helium release characteristics of these fuel forms in the temperature range 800° to 1400°C . Experimental results will then be used in an attempt to derive a theoretical diffusion model helpful to design and safety engineers.

Earlier studies on $^{238}\text{PuO}_2$ plasma-fired microspheres at Oak Ridge National Laboratory, Los Alamos Scientific Laboratory and Mound Laboratory have failed to yield reproducible data; as a consequence, no satisfactory theoretical model of helium release from microspheres could be formulated. One of the reasons for the difficulties has been the variability of the physical characteristics of the plasma-fired microspheres. The physical characteristics of the sol gel and solid solution sol gel microspheres should be much more consistent from sample to sample and as a result, test data should be reproducible. When reproducible helium release data are available, it should then be possible to derive a theoretical model of helium release from PuO_2 .

- b) $\frac{1}{2}$ M.Y.
- c) Nonmetallic
- d) R. E. Zielinski

48. RADIATION DAMAGE AND HIGH TEMPERATURE X-RAY STUDIES

a) The effect of self-irradiation damage on the lattice parameters as a function of time of $^{238}\text{PuO}_2$ (80% ^{238}Pu) plasma formed microspheres have been obtained for three different temperatures (23°C , 100°C and 400°C). Isochronal and isothermal annealing studies have been conducted on these damaged microspheres at temperatures up to 1400°C .

A high temperature X-ray diffraction study of $^{238}\text{PuO}_2$ microspheres has been conducted in the temperature interval of $14\text{-}1505^\circ\text{C}$. Fourth degree polynomial expressions for fractional changes in the lattice parameter and coefficient of thermal expansion were obtained. Presently expansion data of powdered $^{238}\text{PuO}_2$ are being obtained. Future studies will involve the effects of certain additives upon the thermal expansion of the PuO_2 .

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- b) $\frac{1}{2}$ M.Y.
- c) Nonmetallic
- d) W. C. Wyder
- e) Thermal Expansion of $^{238}\text{PuO}_2$ Plasma-Formed Microspheres
R. J. DeSando and W. C. Wyder
submitted to J. Nucl. Materials.

49. COMPATIBILITY

a) This study is concerned with the compatibility of $^{238}\text{PuO}_2$ and various refractory metals and alloys, noble metals and alloys and superalloys, at temperatures between 800 and 2500°C. These studies have been conducted on production grade fuel which contains between 0.5 and 2.00 percent impurities since this was the fuel available when the studies began. Impurities at these levels complicate the interpretation of resulting microstructures in some systems. Selected additional compatibility studies are being conducted, therefore, with higher purity PuO_2 .

- b) 5 M.Y.
- c) Nonmetallic
- d) J. E. Selle
- e) A Summary of Phase Diagrams and Compatibility of PuO_2 with Various Materials
J. E. Selle
MLM-1589 (1969).

50. PHYSICAL PROPERTY MEASUREMENTS

a) This study is directed toward the measurement of the high temperature physical properties, which have not been determined, for the actinide elements, particularly uranium, neptunium and plutonium. These measurements are necessary for comparison with the same properties of the common metals and an interpretation of the solid to liquid phase change. Current studies are directed toward measurement of the thermal conductivity of plutonium in the delta, epsilon and liquid phases, and measurements of the heat of transformations for the three phase changes in neptunium. Future work in this study will be directed toward the measurement of the heat capacity of Np in all phases including the liquid, measurement of the electrical conductivity of liquid Pu and measurement of the heat of fusion for the later-transuranium elements, particularly curium. Also, the high temperature body-centered cubic phase of Pu exhibits an unusual self-diffusion rate and a large compressibility. (Consequently, the elastic moduli of the high temperature phases of U, Np and Pu should be determined).

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- b) $\frac{1}{2}$ M.Y.
- c) Metallic
- d) L. J. Wittenberg
- e) Calorimetric Studies of α , β and γ Plutonium
T. K. Engel
J. Nucl. Materials, 23, 25 (1967).

The Heat Capacities of Al_2O_3 , UO_2 and PuO_2 From 300 to 1100°K
T. K. Engle
J. Nucl. Materials, 31, 211 (1969).

Application of Differential Thermal Analysis to the Study of Phase
Equilibrium in Metal System
D. E. Etter, P. A. Tucker and L. J. Wittenberg
Thermal Analysis, V. 2, Academic Press, New York, 1969.

51. LIQUID METAL PROPERTIES

a) This study is directed toward the determination of the density and viscosity of the liquid transuranium metals. Investigations of these properties for liquid plutonium and its alloys with Fe, Ce, Ga and U have shown that plutonium is an unusual behaving liquid metal. This behavior is associated with the electronic structure of plutonium derived from its position in the periodic table. Measurements of these properties for other transuranium elements will aid in a better understanding of the chemical behavior of the actinide elements. This information will contribute also to better knowledge of liquid metals. Apparatus and techniques have been revised in order to improve the precision of the data. With the improved apparatus, the density and viscosity of neptunium and several additional plutonium alloys will be determined.

- b) 1 M.Y.
- c) Metallic
- d) L. J. Wittenberg
- e) Liquid Plutonium - A Review of Its Physical Properties
L. J. Wittenberg, D. Ofte and W. G. Rohr
Nuclear Applications, 3, 550 (1967).

Fluid Flow of Liquid Plutonium Alloys in an Oscillating Cup
Viscosimeter
L. J. Wittenberg, D. Ofte and C. F. Curtiss
J. Chem. Phys., 48, 3253 (1968).

Density of Liquid Uranium
W. G. Rohr and L. J. Wittenberg
submitted to J. Phys. Chem.

Density and Viscosity of Liquid Plutonium-Uranium Alloys

L. J. Wittenberg, D. Ofte, W. G. Rohr and D. V. Rigney
submitted to Trans. Metallurgical Soc., AIME.

52. STRUCTURE OF LIQUID ACTINIDE METALS

a) The primary objective of this research is the determination of the structures of liquid actinide metals, particularly plutonium and neptunium. The liquid structure of cerium will be obtained initially since it is a convenient non-radioactive metal which exhibits the same type of volume decrease during melting as exhibited by plutonium. Later studies will involve liquid plutonium and neptunium. Information obtained from these studies is expected to reveal correlations between the structures of these liquid metals and their physical properties and help explain such anomalies as the volume decrease during melting and the high viscosity of liquid plutonium.

- b) $\frac{1}{2}$ M.Y.
- c) Metallic
- d) C. R. Hudgens

53. Development of LMFBR Oxide Fuels

a) Physical properties of LMFBR fuels will be determined as follows:

Electrical resistivity measurements using a four-probe technique on $(U,Pu)O_2$ as a function of temperature and stoichiometry (300-1900°K on rod-shaped specimens).

Thermal conductivity measurements on $(U,Pu)O_2$ as a function of temperature and stoichiometry using the comparative longitudinal heat flow apparatus (77-400°K on rod-shaped specimen approximately $\frac{1}{4}$ in. diam. by 2 in. long) and using an absolute radial heat flow apparatus for measuring on solid as well as powder specimens (300-1200°K on a specimen approximately 2 in. diam. by 8 in. high under a pressure of up to 10 atm).

Kinetics of the reaction between PuO_2 microspheres and carbon. Phase diagram studies for the $(U,Pu)O_2$ -fission product systems in the temperature range 800-1400°C using x-ray and DTA measurements.

The following fabrication-related studies are being performed:

The stoichiometry of mixed oxides during sintering by measuring the H_2O in the effluent Ar-4% H_2 furnace gas.

Hydrogen gas release from low-density mixed oxides sintered in Ar-4% H_2 by monitoring the water content in effluent gas as CO_2 and oxygen are passed over the specimen during heating.

The thermochemical reactions which occur during the calcining of mixed oxide sol-gel shards using the TGA technique are being studied.

- b) 2 M.Y.
- c) Nonmetallic
- d) A. L. Lotts
- e) Thermodynamic Applications to $(U,Pu)O_{2-x}$ Fuel Systems
K. E. Spear, A. R. Olsen, and J. M. Leitnaker
ORNL-TM2494 (April 1969).

54. High-Performance Plutonium Fuels

a) Physical properties of LMFBR fuels will be determined as follows:

Electrical resistivity measurements on $(U,Pu)N$, $(U,Pu)(N,C)$ and PuN using a comparative eddy current method (4-300°K) and an absolute

54. High-Performance Plutonium Fuels (continued)

four-probe method (300-1900°K). The comparative method can be used for small pellets ($\frac{1}{4}$ in. diam.) whereas the four-probe method requires rod-shaped specimens.

Thermal conductivity measurements on (U,Pu)N, (U,Pu)(N,C), and PuN using a comparative longitudinal heat flow apparatus (300-500°K on $\frac{1}{4}$ in. diam. by $\frac{1}{4}$ in. high specimens), an absolute longitudinal heat flow apparatus which will yield thermal conductivity, electrical resistivity and Seebeck coefficient data (77-400°K on a rod-type specimen approximately $\frac{1}{4}$ in. diam. by 2 in. long), and an absolute radial heat flow apparatus for measuring heat flow in solids and powders (300-1200°K on a specimen 2 in. diam. by 8 in. high either powder or stacked disks at pressures to 10 atm).

Studies of the nitrogen vapor pressure over (U,Pu)N, (U,Pu)(N,C) and (U,Pu)(N,O,C) using an ultrahigh vacuum system equipped with a mass spectrometer at temperatures between 800 and 2400°C.

The thermodynamics of the Pu(C,N) system will be studied by measuring the lattice parameter as a function of composition for specimens annealed under equilibrium conditions in the range 1300-1900°C.

Thermodynamic compatibility between PuN and the elements in potential LMFBR cladding materials in the temperature range 700-2000°C.

The following fabrication-related studies are contained in the program scope.

The correlation of sintering and hot pressing variables with microstructure for (U,Pu)N and PuN is in progress. Lattice parameter measurements as a function of sintering and heat treatment variables for (U,Pu)N, PuN, and (U,Pu)(N,C).

- b) $2\frac{1}{2}$ M.Y.
- c) Nonmetallic
- d) A. L. Lotts
- e) A Consistent Set of Thermodynamic Rules for PuN
K. E. Spears and J. M. Leitnaker
Jn. American Ceramic Society, V. 51, pp 706-709, 1968.

Review and Analysis of Phase Behavior and Thermodynamic Properties of the Pu-N System
K. E. Spears and J. M. Leitnaker
ORNL-TM 2106, 1968.

PACIFIC NORTHWEST LABORATORY
Battelle Memorial Institute
P. O. Box 999
Richland, Washington 99352

55. Plutonium Alloy Research

a) The research activities in the plutonium alloy program are directed primarily at the investigation of physical and mechanical properties of plutonium alloys. The studies have been concentrated primarily on the Pu - 1 wt. % Ga alloy. Specifically, thermal expansion, transformation behavior, phase stability, temperature and strain rate dependence of the mechanical properties, and microstructural variations resulting from mechanical and thermal treatments are being studied. These studies are supported by electron microprobe analyses, electron microscopy, conventional metallography, density determinations, dilatometry, differential thermal analyses, electrical resistivity measurements, elevated temperature metallography, and a wide variety of mechanical property measurements.

- b) 7 M.Y.
- c) Metallic
- d) R. W. Stewart
- e) A Sputtered Plutonium-Gallium Alloy
R. A. Busch
BNWL-3025 (1970)

Phase Transformation in Delta Phase Pu-Ga Alloys under
Hydrostatic and Cold Working Conditions
J. M. Taylor
BNWL-1296 (1970)

An Automatic Ultrasonic Micrometer with an Application to
EDM Control
D. R. Newman and L. T. Lamb
BNWL-1022, February 1969

The Plutonium-Zirconium Equilibrium Diagram from 0 to 10
Atomic Percent
J. M. Taylor
J. Nucl. Matls., 30 (1969), 346

Thermal Expansion of Some Plutonium-Gallium Alloys
J. M. Taylor
J. Nucl. Matls., 31 (1969), 339

Electrolytic Polishing and Etching of Thin Films
H. W. Arrowsmith and D. G. Rencken
Presented at the 2nd Annual Mtg. of the Inter. Metallographic
Soc., San Francisco, Calif., September 1969

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- e) Spallation Threshold Tests on Niobium, Tantalum, and Zirconium

G. P. Hanneman

BNWL-756, September 1968

Elevated Temperature Hardness Testing of Plutonium-Gallium Alloys

D. D. Hays

BNWL-682, March 1968

A New Procedure for Revealing the Microstructure of Alpha Plutonium

D. G. Rencken

Inter. J. of Metallography, 1 (1968), 365

A Study of Phase Equilibria with Plutonium-Zirconium Alloys

J. M. Taylor

BNWL-402, September 1968

Microhardness of Plutonium-Gallium Alloys

D. D. Hays

J. Nucl. Matls., 22 (1967), 352

Compositional and Pressure Effects in the Plutonium-Gallium System

H. R. Gardner

Plutonium 1965, A. E. Kay and M. B. Waldron (Eds.), Chapman and Hall (1967), 118-132

Fatigue Behavior of Plutonium - 1.0 wt% Gallium Δ -Stabilized Plutonium

H. R. Gardner

Plutonium 1965, A. E. Kay and M. B. Waldron (Eds.), Chapman and Hall (1967), 535-542.

Application of Sputtering to the Fabrication of Neutron Detector Anodes

E. D. McClanahan, R. W. Moss, and R. A. Busch

BNWL-553, November 1967

56. Plutonium Metallurgy Research

a) The objectives in plutonium metallurgy research at Battelle-Northwest are to obtain fundamental information on the metallurgy, structure, and properties of plutonium and plutonium alloys in support of the Commission's basic plutonium program. The major activities are directed principally to: (1) study of the phase transformations and properties of high purity plutonium and selected plutonium alloys, (2) determination of the precise mechanisms by which monoclinic plutonium deforms, (3) metallurgy of sputter-deposited plutonium, and (4) development of research techniques

- applicable to alpha-radioactive materials. Specific information concerning these interrelated activities and publications produced are summarized below:

- Phase Transformations

The major objectives of this activity are (a) to determine the nature, kinetics, and mechanisms of solid-state reactions in plutonium and plutonium alloys; (b) to investigate differences in the structure of beta formed from alpha versus beta formed from gamma in unalloyed plutonium; and (c) to investigate phase transformations and properties of sputter-deposits.

- Deformation Processes

The objectives of this activity are (a) to determine the mechanisms of deformation of the allotropes of plutonium and the annealing of alpha and beta plutonium; (b) study the anisotropy of physical and mechanical properties of plutonium; and (c) investigate deformation processes of fine-grained α -Pu and of sputter-deposits.

- Metallurgy of Sputter-Deposited Plutonium

The mechanical properties and phase changes in sputter-deposited unalloyed plutonium are being studied. The structure and properties of stable and metastable alloys are being investigated. The properties, phase transformations, and deformation processes of sputter-deposits are being compared to cast and heat treated material to better understand the metallurgy of this complex metal.

- Technique Development

The main objectives in this activity are to develop techniques that will allow accurate evaluations pertaining to the studies mentioned previously and to provide specimens for investigation. The techniques being developed are (a) metallography (especially elevated temperature metallographs); (b) electron microscopy; and (c) sputter-deposition of the α , β , and δ phases of plutonium.

- b) 5.5 M.Y.
- c) Metallic
- d) R. D. Nelson
- e) Optical-Electrolytic Etching

H. W. Arrowsmith and D. G. Rencken

Submitted for publication in Inter. J. of Metallography

Recrystallization of Alpha Plutonium by Extrusion

M. D. Merz

J. Nucl. Matls., 34 (1970), 108

- e) The Crystallography of the Alpha to Beta Transformation in Plutonium

R. D. Nelson and F. E. Bowman
Trans. AIME, 245 (1969), 967

Large Creep Ductility of Alpha Plutonium

R. D. Nelson and F. E. Bowman
Trans. AIME, 245 (1969), 1656

High Elongations of Alpha Plutonium

M. D. Merz
Presented at the 1969 AIME Spring Meeting, Pittsburgh, Pennsylvania, May 12-15, 1969

Replica Electron Micrographs of Sputter Delta-Stabilized Plutonium Alloys

S. D. Dahlgren
J. Nucl. Matls., 28 (1968), 333

Superplasticity in Unalloyed Plutonium

S. D. Dahlgren
Trans. AIME, 240 (1968), 126

Recrystallization in Unalloyed Plutonium

R. D. Nelson
Trans. AIME, 240 (1968), 331

Deformation of Alpha Plutonium

R. D. Nelson and S. D. Dahlgren
Trans. AIME, 242 (1968), 1865

Microstructure of Deformed Alpha Plutonium

R. D. Nelson and S. D. Dahlgren
BNWL-981 (1968)

Televised Observation of Plutonium Transformations

H. W. Arrowsmith
Presented at the International Metallographic Society Mtg., Denver, Colorado, November 11-13, 1968

Electrolytic Polisher-Etcher for Standard Metallographs

H. W. Arrowsmith and D. G. Rencken
Presented at the International Metallographic Society Mtg., Denver, Colorado, November 11-13, 1968

Deformation in Alpha Plutonium

S. D. Dahlgren
Presented at the Annual Meeting of the Metallurgical Society of AIME, New York, February 1968

PACIFIC NORTHWEST LABORATORY

e) Grain Size Dependency of β -Pu Plasticity

R. D. Nelson

Presented at the AIME Fall Meeting, Detroit, Michigan,
October 13-17, 1968

Superplasticity in Unalloyed Beta Plutonium

S. D. Dahlgren

Presented at the Fall Meeting of the Metallurgical Society
of AIME, Cleveland, Ohio, October 1967

Recrystallization in Alpha Plutonium

R. D. Nelson

Presented at the Fall Meeting of the Metallurgical Society
of AIME, Cleveland, Ohio, October 1967

Transformation and Deformation in Plutonium

R. D. Nelson, F. E. Bowman, and T. K. Bierlein

Plutonium 1965, A. E. Kay and M. B. Waldron (Eds.), Chapman
and Hall (1967), 1-17

Steady-State Creep of Plutonium

R. D. Nelson

Plutonium 1965, A. E. Kay and M. B. Waldron (Eds.), Chapman
and Hall (1967), 564-570

57. Fast Reactor Ceramics Research

a) The major objective of this program is to develop research data needed for an accurate understanding of the solid-state behavior of fast-reactor fuel materials. The program currently emphasizes research on mixed uranium-plutonium oxides near the composition $UO_2 - 25 \text{ wt. \% } PuO_2$. The program includes: (1) direct, in-reactor property studies, (2) postirradiation property studies, and (3) preirradiation, base-line property studies. A significant goal of the program is to appraise the effects of irradiation on creep behavior, thermal conductivity, thermal expansion, melting point, and liquid surface tension of the mixed oxide fuel, and fuel - sodium - cladding compatibility. In addition to irradiation, major experimental variables include temperature, stoichiometry, density, and composition (U/Pu ratio, purity). Studies to determine appropriate phase relationships as a function of U/Pu ratio near 3 and O/M ratios near 2.00 are also planned to permit accurate interpretations of property studies in terms of composition and phases present in the system. The program is divided into the tasks shown below:

- Mechanical Properties of Fast-Reactor Oxide Fuels

The objective of this work is to measure those mechanical properties of fast-reactor oxide fuels required for the analysis of stress buildup occurring during reactor operation due to fuel swelling and short-time temperature transients. High temperature out-of-reactor creep measurements are being made on mixed oxides from 900 to 1650°C. The effects of porosity, grain size, temperature, stoichiometry, and applied load on the creep behavior are being analyzed. In-reactor isothermal creep studies are also being conducted to evaluate the effect of fission on the creep rate. Elastic properties of hot-pressed (U,Pu)O₂ and single crystal UO₂ are being determined using ultrasonic velocity measurements.

- Thermophysical Properties of Fast-Reactor Oxide Fuels

The objective of this work is to determine the effects of irradiation and materials variables (stoichiometry, composition, and density) on the thermophysical properties (thermal conductivity, thermal expansion, melting point, and electrical conductivity) of fast-reactor mixed oxides. The thermal conductivity of (U,Pu)O₂ is being determined to 2500°C using thermal diffusivity measurements in conjunction with heat capacity data. To better understand the mechanism(s) of heat transfer in (U,Pu)O₂, an attempt is being made to correlate electrical conductivity and optical property measurements on UO₂, PuO₂, and mixed oxides. Thermal expansion is being determined to 2500°C using high temperature x-ray diffraction and melting behavior is evaluated using high temperature differential thermal analysis. Of particular interest is the effect of increasing fission product concentration due to burnup on the above properties.

- Properties of Molten Fast-Reactor Oxide Fuels

The objective of this research is to measure those properties of molten uranium-plutonium oxides which relate to mechanical design and safety of LMFBR fuels. Volume change on melting and liquid density to 3000°C are being measured using x-radiography at high temperature to observe a change in liquid column height as a function of temperature. Viscosity of molten mixed oxides will be measured using an oscillating cup viscometer, while surface tension is being measured via either a pendant drop technique or through evaluation of the meniscus shape at temperatures above the melting point. Compressibility and volume viscosity of molten mixed oxides are calculated from measured values of ultrasonic velocity and attenuation.

• Phase Studies of Fast-Reactor Oxide Fuels

The objective of this work is to determine the various phases which exist in the U-Pu-O ternary system in the region of 25 wt. % PuO_2 and O/M of 1.90 - 2.05. Of particular interest is definition of the temperature and composition limits of the nonstoichiometric MO_{2+x} phase. Oxygen decomposition pressures are being measured to 1600°C using a thermogravimetric system and the $\text{H}_2/\text{H}_2\text{O}$ equilibrium to establish partial oxygen pressures. Thermodynamic quantities for the nonstoichiometric oxide will be calculated. Phase identification is being carried out using high temperature x-ray diffraction.

• Compatibility of Irradiated Mixed Oxide Fuels with Sodium and Stainless Steel

The objective is to evaluate the compatibility of irradiated mixed oxides with sodium and stainless steel as a function of burnup, stoichiometry, time, and temperature. Diffusion couples between irradiated mixed oxide fuel, previously characterized by shielded microprobe analyses, and stainless steel will be prepared. Isothermal and isochronal anneals are carried out and the redistribution of fission products then evaluated using microprobe analysis. Static compatibility tests between high burnup mixed oxide and sodium are also being considered and the rate of dissolution of various fission products into sodium determined.

- b) 17 M.Y.
- c) Nonmetallic
- d) J. L. Bates, T. D. Chikalla
- e) Reactor Fuels and Materials Development Programs for Fuels and Materials Branch of USAEC Division of Reactor Development and Technology (BNWL-1223, November 1969)

Thermal Conductivity and Electrical Resistivity of Uranium Oxycarbides

J. L. Bates

BNWL-989, March 1969

The Thermoelectric Power of Uranium Dioxide at High Temperatures

J. L. Bates and G. J. Dau

Submitted for journal publication

Determination of Oxygen/Metal Ratios of Nonstoichiometric Uranium, Plutonium, and Mixed Uranium-Plutonium Oxides

T. D. Chikalla and C. E. McNeilly

BNWL-SA-2493, submitted to J. Nucl. Matls.

- e) Microstructure and Lattice Defects in Polycrystalline Nuclear
Ceramics
J. L. Daniel and S. J. Mayhan
Proceedings of Electron Microscopy Society of America
Twenty-Sixth Annual Meeting, Claitors' Publishing Division,
Baton Rouge, La. (1968), 270

The Effect of Oxygen Stoichiometry on the Thermal Diffusivity
and Conductivity of $(U_{0.75}Pu_{0.25})O_{2-x}$
R. L. Gibby
BNWL-927, January 1969

Hot Hardness, Enthalpy, Heat Capacity, Thermal Conductivity,
and Electrical Resistivity of Uranium Oxycarbides
J. L. Henry, R. Blickensderfer, D. Paulson, and J. L. Bates
Submitted for publication in J. Am. Cer. Soc.

Chemical Etchants for Plutonium Ceramics
S. J. Mayhan and M. P. Bauleke
BNWL-SA-2176, submitted to J. Nucl. Matls.

The Metallographic Preparation of Some Plutonium-Uranium Fuel
Materials
F. M. Smith, S. J. Mayhan, and R. V. Olson
Inter. J. of Metallography, 2 (2/3) (1969), 179

Examination of Radioactive Materials by Scanning Electron
Microscopy
J. L. Daniel
BNWL-SA-2385, presented at the 2nd Annual Scanning Electron
Microscopy Symposium, sponsored by IIT Research Institute,
Chicago, Illinois, April 29-May 1, 1969

Powder Morphology of Nuclear Ceramics by Scanning Electron
Microscopy
J. L. Daniel, S. J. Mayhan, E. T. Weber, and M. J. Barr
BNWL-SA-2507, presented at the 27th Annual Meeting of the
Electron Microscopy Society of America, St. Paul, Minn.,
August 26-29, 1969

Techniques for Measurement of Density, Surface Tension,
Viscosity, and Compressibility of Liquid Oxides
R. P. Nelson, J. A. Christensen, O. D. Slagle,
J. J. Rasmussen, and P. L. Farnsworth
BNWL-SA-2629 A, presented at the 22nd Pacific Coast
Regional Meeting of the American Ceramic Society, Seattle,
Washington, October 15-17, 1969

- e) Viscosity, Surface Tension, and Compressibility of Molten UO_2
R. P. Nelson, J. J. Rasmussen, and O. D. Slagle
BNWL-SA-2667 A, presented at the 22nd Pacific Coast
Regional Meeting of the American Ceramic Society, Seattle,
Washington, October 15-17, 1969

Phase Relationships in the Americium-Oxygen System

T. D. Chikalla and L. Eyring
J. Inorg. Nucl. Chem., 30 (1968), 133

Nonstoichiometry Effects on the Melting Heat Rating for UO_2 -
25 w/o PuO_2 Fast Fuels

J. A. Christensen
BNWL-CC-1490, January 1968

Cinemicrography for Dynamic Electron Microscopy

J. L. Daniel
Laboratory Management, 6 (1968), 12

Scientific Exchange Assignment to Japan; Review and Evaluation

J. L. Daniel
BNWL-724, March 1968

The Thermal Diffusivity and Thermal Conductivity of Stoichio-
metric $(\text{U}_{0.80}\text{Pu}_{0.20})\text{O}_2$

R. L. Gibby
BNWL-704, May 1968

Dynamic Electron Microscopy of High Temperature Reactions

S. J. Mayhan and J. L. Daniel
Advanced Techniques for Material Investigation and
Fabrication, Science of Advanced Materials and Process
Engineering Series, Volume 14 (November 1968)

High Pressure Effects on Single Crystals, Polycrystals, and
Powders

S. Takahashi and J. L. Daniel
Supplement to Transactions of the Japan Institute of
Metals 9 (1968), 343

Thin Section Microstructures of Nuclear Ceramics

J. L. Daniel, S. J. Mayhan, R. J. Baker, and R. H. Beauchamp
Presented at the American Ceramic Society Annual Meeting,
April 20-25, 1968

e) Ceramic Fuels for Fast Reactors

S. Goldsmith, R. J. Jackson, and E. T. Weber
BNWL-SA-1550, presented at the American Nuclear Society
Topical Meeting on Fast Reactors, Cincinnati, Ohio,
April 1-4, 1968

Development of Mixed-Nitride Fuel Materials Obtained by
Carbothermic Reduction of the Oxides
E. T. Weber, W. O. Greenhalgh, and R. L. Gibby
Presented at the Annual Meeting, American Nuclear Society,
Toronto, Canada, June 9-13, 1968

Electrical Conductivity of Uranium Dioxide
J. L. Bates, C. A. Hinman, and T. Kawada
J. Am. Cer. Soc., 50 (1967), 652

Electrical Conductivity of UO_2 : Part II, Final Report
J. L. Bates
BNWL-296, November 1967

Chemical Thinning of Pneumatically Impacted UO_2 for Electron
Transmission Microscopy
M. P. Bauleke
BNWL-416, May 1967

Dissociation Pressures and Partial Thermodynamic Quantities
for Americium Oxides
T. D. Chikalla and L. Eyring
J. Inorg. Nucl. Chem., 29 (1967), 2281

A Review of Heat Transfer in Mixed Oxide Fuels
J. A. Christensen
BNWL-CC-1390, March 1967

Stoichiometry Effects on Oxide Nuclear Fuels - I. Power Rating
Required for Melting and Oxygen Redistribution in Molten
Center UO_{2+x} Fuels
J. A. Christensen
BNWL-536, December 1967

High Pressure Effects on Single Crystals, Polycrystals, and
Powders
S. Takahashi, J. L. Daniel, N. Asami, B. Ishii, and Y. Honda
Japan Institute of Metals: Proceedings of the International
Conference on the Strength of Metals and Alloys, Tokyo,
Japan, September 4-8, 1967

e) Thermal Diffusivity of Uranium Oxycarbide

J. L. Bates

Presented at the Fall Meeting of the Nuclear Division,
American Ceramic Society, October 20-22, 1967, Urbana, Ill.

Cooling-Rate Effects in Molten-Center Oxide Fuels

J. A. Christensen

Presented at the American Ceramic Society Regional Meeting,
San Francisco, Calif., October 1967

58. Irradiation Behavior of Fast Reactor Fuel Materials

a) The purpose of this investigation is to achieve means for prediction of the irradiation behavior of fast reactor fuels. The tasks within the program are organized so that each attacks a major area of concern in the irradiation behavior of fuels. The overall program is directed toward the development and experimental testing of analytical models for predicting the response of fast reactor fuels to irradiation. In the immediate future, efforts will be focused on developing models describing fuel swelling and migration of chemical constituents (original plus fission product) during irradiation. Some of the data needed to test the models and evaluate their parameters will be supplied by critical irradiation experiments conducted in this program while other needed data will be obtained from laboratory research programs or by analysis of appropriate samples obtained from other irradiation programs. The plutonium research portion of the program is divided into the following tasks:

• In-Reactor Thermal Performance of $\text{UO}_2\text{-PuO}_2$ Fuel

The objective of this task is to determine the effects of design and fabrication parameters on those thermal properties of fast reactor oxide fuel materials which are critical for design of near-term LMFBR fuels. Measurements to be made include the following: (1) fuel-sheath gap ΔT as a function of heat rating and irradiation time; (2) effects of xenon on fuel-sheath gap ΔT ; (3) high burnup effects on melting heat rating; (4) kinetics of fuel restructuring and effects of principal methods of void deployment on temperature distribution within the fuel; and (5) O/M effects on fuel melting heat rating. In addition, the above efforts will be integrated with concurrent fast flux fuels irradiations to provide support in optimizing EBR-II test design and interpretation.

• Irradiation of Hypostoichiometric $\text{UO}_2\text{-PuO}_2$ Fuel

The objective of this work is to determine the combined effects of stoichiometry, burnup, and heat rating on dimensional stability, fuel-sheath compatibility, and constituent redistribution in $\text{UO}_2\text{-PuO}_2$ fuels operating under conditions at least as severe as anticipated in FTR. Thermal performance, swelling, and gas release characteristics of $\text{UO}_2\text{-PuO}_2$ fuel of various stoichiometries will be determined. The degree of axial and radial plutonium and oxygen redistribution, fission product species location, and changes in cladding chemistry will be investigated using extensive microprobe and microchemical analyses.

• Migration Behavior of $\text{UO}_2\text{-PuO}_2$ Fuel

The objective of this task is to measure and interpret thermal-gradient activated migration of fuel constituents and fission products and assess effects of such migration on properties which limit fuel performance. Studies are directed toward measuring the oxygen, plutonium, and fission product concentration gradients in oxide fuels irradiated at conditions yielding large thermal gradients. Computer programs are being developed which assess the relative importance of potential migration mechanisms from concentration distribution data and associated measurements of basic diffusion quantities. These programs will be particularly useful in assigning values to stochastic modeling parameters. Much of the raw data for required analyses will be obtained from irradiations performed under other portions of this program.

- b) 14 M.Y.
- c) Nonmetallic
- d) J. A. Christensen
- e) Reactor Fuels and Materials Development Programs for Fuels and Materials Branch of USAEC Division of Reactor Development and Technology (BNWL-1223, November 1969)

Research at Battelle-Northwest on Transport Processes in Oxide Nuclear Fuels

J. A. Christensen
BNWL-1202, October 1969

The Effects of Xenon on Fuel-Clad Gap Conductance

G. R. Horn
ANS Trans., 12 (2) (1969)

Fission Gas Induced Swelling in Uranium at High Temperatures and Pressures

G. L. Kulcinski, R. D. Leggett, C. R. Hann, and B. Mastel
J. Nucl. Matls., 30 (1969), 303

e) Reversibility of Fission Gas Swelling in Uranium

G. L. Kulcinski and R. D. Leggett

J. Nucl. Matls., 31 (1969), 279

Dissolution of UC Particles in Uranium During Postirradiation Annealing

G. L. Kulcinski and R. D. Leggett

Trans. AIME, 245 (1969), 929

High Pressure - High Temperature System for Postirradiation Annealing of Uranium

G. L. Kulcinski, R. A. Thiede, and C. R. Hann

Rev. Sci. Inst., 40 (1969), 214

Melting Heat Rating of UO_2 - 25 w/o PuO_2 Fuels

L. A. Lawrence, J. A. Christensen, and G. R. Horn

ANS Trans., 12 (2) (1969)

Evaluation and Interpretation of Stochastic Models for Nucleation, Growth, and Migration

K. R. Merckx and W. L. Nicholson

To be published in proceedings of the Battelle Colloquium, Inelastic Behavior in Solids, September 1969

Models for Reactor Fuel Element Which Consider Void Deployment

K. R. Merckx

Nucl. Engrg. and Design, 9 (1969), 15

Unfolding Particle Size Distributions

K. R. Merckx and W. L. Nicholson

Technometrics, 11 (4) (1969), 707

Nonstoichiometry Effects on the Melting Heat Rating for UO_2 - 25% PuO_2 Fuels

J. A. Christensen

ANS Trans., 11 (1) (1968)

Nonstoichiometry Effects on the Microstructure of Irradiated UO_2 - 25 wt. % PuO_2 Fuels

J. A. Christensen

ANS Trans., 11 (2) (1968)

High Burnup Plutonium Oxide Irradiations

J. A. Christensen

ANS Trans., 11 (2) (1968)

In-Reactor Sintered Fuel

G. R. Horn and R. L. Gulley

ANS Trans., 11 (1) (1968)

59. Transplutonium Ceramics Research

a) This is a new program (FY 1970) which involves research studies on the solid-state properties and behavior of transplutonium ceramic materials. The initial program emphasis will be on transplutonium oxides and will include a study of the oxygen decomposition pressures over nonstoichiometric berkelium oxide and a systematic study of the effects of self-radiation damage due to alpha decay and recoil atom bombardment in selected actinide dioxides.

The objective in the berkelium oxide study is to evaluate the stability of BkO_{2-x} with respect to dissociation and derive the equilibrium oxygen dissociation pressures over nonstoichiometric compositions between Bk_2O_3 and BkO_2 . This will lead to the pressure - temperature - composition relationships in the system and allow evaluation of the thermodynamic stability of compositions in the interval $1.50 < (\text{O/Bk}) < 2.00$. The objective of the self-radiation damage study is to understand the self-damage mechanism and to determine differences in damage rate and saturation values as a function of disintegration rate. Diffraction-line profile analyses will be carried out and will allow evaluation of the lattice strain accompanying the damage process. Annealing experiments will be conducted and the kinetics of damage annealing will be studied in detail to determine whether definite recovery stages occur.

The recoil of the heavy atoms of actinide dioxides will be simulated using a computer. This involves several extensions of existing cascade simulation work, such as modifying the computer code, CASCADE-CLUSTER. With an appropriately modified code, computer experiments will be run to determine the configurations resulting from the collision processes. Of particular concern will be the investigation of possible strong dependencies on certain crystallographic directions, the approach to saturation, the effect of nonstoichiometry, and whether defect clustering should be expected.

Ultimately, studies will be aimed at understanding the occurrence of nonstoichiometric intermediate phases in actinide oxide systems and the influence of self-irradiation on their stability.

- b) 2 M.Y.
- c) Nonmetallic
- d) T. D. Chikalla

60. Isotopic Fuels Program

a) There is an increasing interest in the use of the longer-lived isotopes such as ^{241}Am , ^{238}Pu , and ^{244}Cm for heat source applications. However, many of the basic high temperature properties required in the design and safety analyses of these heat source materials are unknown. The objective of this program is the determination of some of the thermophysical and thermochemical properties of heat source oxides, with emphasis on measurements at temperatures close to and above the melting point. The thermal diffusivity and thermal conductivity of $^{238}\text{PuO}_2$ and $^{238}\text{PuO}_2\text{-ThO}_2$ solid solution fuel forms have been measured to 1600°C and these experiments will be extended to provide data to the melting point. The surface tension of molten PuO_2 will be determined through analyses of pendant drops of a cylindrical meniscus. The viscosity of molten PuO_2 will be measured using an oscillating crucible technique at several temperatures above the melting point. The effects of self-radiation damage in $^{238}\text{PuO}_2$ at room temperature have been studied using x-ray powder diffraction techniques. Another investigation in this program involves the determination of the vapor pressure of molten plutonium oxide.

Though the current emphasis in this program is on $^{238}\text{PuO}_2$ and PuO_2 -based solid solutions, past work has included research on curium oxides and several papers dealing with the thermodynamics and phase equilibria in the curium-oxygen system as well as physical properties of Cm_2O_3 have been published.

- b) 3 M.Y.
- c) Nonmetallic
- d) T. D. Chikalla
- e) The Curium-Oxygen System
T. D. Chikalla and L. Eyring
J. Inorg. Nucl. Chem., 31 (1969), 85

The Thermal Diffusivity, Thermal Conductivity, and Transformation Temperatures of Curium Sesquioxide

R. L. Gibby, C. E. McNeilly, and T. D. Chikalla
Submitted to J. Nucl. Matls.

Crystallographic Transformations in Curium and Promethium Oxides by DTA

C. E. McNeilly

Presented at the Annual Meeting of the North American Thermal Analysis Society, Columbus, Ohio, November 17-18, 1969

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Pure

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ENVIRONMENT

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Gas

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23
25
26
53

Magnetic field

3
4
24
33

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9 15
10 16
11 57
14 58

Pressure

Above atmospheric

1
46
55

Temperature

Above 1000°K

2	10	27	45	54
6	12	28	47	57
7	14	36	48	58
8	15	37	49	59
9	16	39	53	60

Below room temperature

3
4
20
33
43

(indexed by program number)