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MOUND LABORATORY-MONSANTO
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MONSANTO RESEARCH CORPORATION

MOUND LABORATORY MIAMISBURG, OHIO

U. S. GOVERNMENT CONTRACT NO. AT-33-1-GEN-53

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February 6, 1964

Mr. W. B. Creamer, Manager
Dayton Area Office
U. S. Atomic Energy Commission
Post Office Box 66
Miamisburg, Ohio

Dear Mr. Creamer:

Enclosed are three copies of the "Summary of the Division of Reactor Development Programs at Mound Laboratory for January, 1964."

Please arrange for the transmittal of two copies to the Division of Reactor Development, Attention Mr. J. M. Simmons, Chief of Fuels and Materials Development Branch, U. S. Atomic Energy Commission, Washington 25, D. C.

Very truly yours,

D. L. Scott

David L. Scott
Vice President,
Plant Manager

DLS:wd

Distribution: Copy 1A - 3A - W. B. Creamer, DAO
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SUMMARY OF THE DIVISION OF REACTOR DEVELOPMENT PROGRAMS AT MOUND LABORATORY FOR JANUARY, 1964

Submitted by: Dr. G. Richard Grove

Prepared by: Dr. L. J. Wittenberg

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MONSANTO RESEARCH CORPORATION

A SUBSIDIARY OF MONSANTO CHEMICAL COMPANY

Monsanto

ATOMIC ENERGY COMMISSION · UNITED STATES OF AMERICA

MOUND LABORATORY

MIAMISBURG, OHIO

OPERATED FOR

UNITED STATES ATOMIC ENERGY COMMISSION

U.S. GOVERNMENT CONTRACT NO. AT-33-1-GEN-53

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February 6, 1964

SUMMARY OF THE DIVISION OF REACTOR DEVELOPMENT PROGRAMS AT MOUND LABORATORY FOR JANUARY, 1964

Submitted by: Dr. G. Richard Grove
Prepared by: Dr. L. J. Wittenberg

Plutonium-Cerium-Nickel Phase Diagram

Metallographic examination of the low nickel concentration region of the plutonium-cerium-nickel phase diagram indicated that the two eutectic valleys originating on the Ce-Ni binary at 22.5 and 34.5 atom per cent cerium and extending into the ternary system form an extensive primary solid solution region of plutonium in Ce_5Ni_2. A ternary eutectic has been identified at approximately 55 Pu - 22 Ce - 23 atom per cent Ni (440°C). The equilibrium phases at the eutectic, all of which contain extensive solid solubility are Pu, Ce, and Ce_5Ni_2.

Plutonium-Cadmium Binary System

The results for six compositions in the plutonium-cadmium binary system analyzed by differential thermal analysis, indicate the probability of at least one high melting congruent compound in the system. Two isotherms for either eutectic or peritectic reactions were noted, one at 910°C for compositions of 10 to 50 atom per cent cadmium, and the other at 730°C for 82 to 95 atom per cent cadmium alloys. Liquidus temperatures of compositions from 10 to 80 atom per cent cadmium are above 1000°C.

Plutonium-Cerium-Iron Phase Diagram

The investigation of eight ternary compositions in the plutonium-cerium-iron ternary system by differential thermal analysis supports the proposed eutectic valley which extends from the cerium-iron binary phase diagram at 90 Ce - 10 atom per cent iron to the plutonium-iron binary phase diagram at 90 Pu - 10 atom per cent iron. A ternary eutectic is apparently located

in the plutonium-rich portion of the ternary diagram at 430°C.

Since disagreement was noted between the present data and the literature diagram of the cerium-iron system, the binary system was reinvestigated up to 50 atom per cent iron.

Plutonium-Gold Binary System

Analysis with the electron microprobe x-ray analyzer indicates that there are 5 compounds in the plutonium-gold binary system, with probable formulas of AuPu, Au_5Pu_4, Au_5Pu_2, Au_3Pu, and Au_4Pu. Apparently the AuPu is a congruent melting compound, and Au_3Pu and Au_4Pu are incongruent compounds. A eutectic exists at about 86.0 atom per cent gold.

Development of High Temperature Differential Thermal Analysis

Previous difficulties in the construction of a high temperature differential thermal analysis apparatus have been overcome in a newly designed apparatus which is capable of operation to 1500°C. Platinum-platinum 13 per cent rhodium thermocouples which are insulated with magnesia and beryllia and sheathed with platinum 10 per cent rhodium are utilized. High-purity magnesia and beryllia are utilized as the thermocouple insulator materials because of their high electrical resistivities at elevated temperatures.

Physical Properties

The viscosity of a plutonium-13 atom per cent iron hypereutectic alloy was measured at 11 temperatures from its liquidus temperature of 460 to 788°C. The measurement of this hypereutectic alloy extends the study of the relationship between viscosity and composition in the plutonium-iron binary alloy system. The study has shown that the viscosity rises with iron additions to plutonium to a maximum at the eutectic composition, 9.5 atom per cent iron. Further additions of iron cause a decrease in viscosity. The liquid apparently responds to some ordering or "structuring" influence at the eutectic composition. Additionally, measure-

ments extended below the liquidus temperature revealed significant characteristics of the liquidus surface of this portion of the plutonium-iron phase diagram.

Physical Properties

The viscosity of a plutonium - one weight per cent gallium alloy was measured at 12 temperatures from its liquidus temperature to 816°C, in the oscillating cup viscosimeter. The liquidus temperature at 669°C was precisely determined during this experiment. The viscosity decreased linearly with increasing temperatures on a plot of log (η/T), versus $(1/T)$, where (η) is expressed in poise and (T) is in degrees Kelvin. The viscosity at 672°C was 8.74 centipoise and decreased to 6.58 centipoise at 816°C. The activation energy as calculated from the slope of the line on this plot was 6044 cal/g - atom. The one weight per cent addition of gallium to plutonium resulted in a 50 per cent increase in viscosity as compared to unalloyed plutonium over the same temperature interval.

Cerium Purity Studies

Small additions of oxygen in the form of cerous oxide to cerium metal markedly reduced the melting point of the cerium. These results indicate a eutectic reaction in the cerium-oxygen system at high cerium concentration.

Thermal Property Studies

Several inquiries have been received concerning the diatomaceous earth castings which are used for thermal insulation in the high temperature calorimeter. Since the thermal properties of this material have not been measured directly, the thermal conductivity of this material was calculated from the sensitivity data from the previous calorimeter. A value of 4.1×10^{-4} cal/°C-sec-cm, approximately four times the conductivity of the unfired, loosely packed powder, was obtained.

Supplementary

F. D. Lonadier and L. J. Wittenberg visited the General Electric, Nuclear Material and Propulsion Operation on January 6 and Battelle Memorial Institute on January 7. Programs and instrumentation useful for the Division of Isotope Development studies at Mound Laboratory were reviewed and observed. Subsequently, L. J. Wittenberg and T. K. Engel, visited Dr. Herbert Deem, at Battelle Memorial Institute, January 31, for the purpose of observing high temperature thermal property apparatus which would be appropriate for the investigations at Mound.

The article "Plutonium Dioxide Microspheres for Nuclear Fuel Produced with Induction-Coupled Plasma", was summarized in the Industrial Publication of the Lepel High Frequency Heating Review, which was published January 2, 1964.

L. J. Wittenberg gave a seminar on "Properties of Molten Plutonium Alloys" for the Chemical, Metallurgical and Nuclear Engineering Department, University of Cincinnati, on January 16.

Mr. Geoffrey Craig Wall, of the Australian Atomic Energy Commission, visited the Research Department on January 24, 1964. His chief interest concerned the preparation of plutonium and thorium dioxide microspheres.

Donald Ofte attended the American Chemical Society Meeting in Denver, Colorado, the week of January 20, 1964 and presented the paper "Viscosity-Composition Relationships of Molten Hypoeutectic Plutonium-Iron Alloys." During the week he visited areas of interest at Dow Chemical Company, Rocky Flats Division, and the Ames Laboratory, Ames, Iowa.

A paper entitled "Spheroidization of Plutonium Bearing Ceramics for Reactor Fuels Applications", by P. A. Tucker, K. D. Phipps, D. Ofte, and L. J. Wittenberg, has been submitted and accepted for presentation at the American Ceramic Society Annual Meeting for April 20, 1964.

Mr. James Gebhart, a recent Metallurgical Engineering graduate from Missouri School of Mines, was added to the group in January.

Dr. J. F. Willging and Mr. Warren Hammond of Dow Chemical Company, Rocky Flats Division visited the Reactor Fuels Laboratory on January 30.

Mr. T. K. Engel observed the testing of the furnace being purchased for the new high temperature calorimeter at Marshall Products Company, Columbus, Ohio, January 30.