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STUDIES OF PLUTONIUM-238 FUEL FORMS AND THEIR CONTAINMENT

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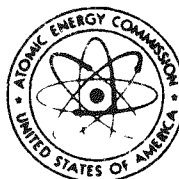
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ABSTRACT

Several forms of the plutonium-238 isotope are being considered for use as radioisotopic heat sources. Compatibility studies of the fuel forms with container materials for various applications are also being carried on. Of the forms studied, plutonium metal is the least desirable for high-temperature applications. The stability, ease of handling, and high melting point of plutonium dioxide make it the first choice among the alternate fuel forms.

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STUDIES OF PLUTONIUM-238 FUEL FORMS AND THEIR CONTAINMENT

INTRODUCTION

Plutonium-238 is one of the radioisotopes designated by the Division of Isotopes Development for use in the SNAP programs. The responsibility for research and development of plutonium-238 fuel forms and their containment has been designated to Mound Laboratory, which has had programs involving this material for more than four years.

Many of the properties of this isotope are well documented. Chemically and physically, the behavior is similar to that of plutonium-239, about which many volumes have been written. Radiochemically there is a great difference, plutonium-238 being almost 300 times more radioactive. It is this property which makes plutonium-238 attractive as a radioisotopic heat source. A half-life of 86.4 years imparts a radioactivity of 17.4 curies per gram, characterized by the 5.45 and 5.50-Mev alpha particles as shown in Figure 1. The decay scheme shows the very-low-energy gamma rays associated with the isotope. The radiation properties of plutonium-238, coupled with a spontaneous fission rate of $\sim 10^3$ neutrons per second per gram, enhance its applicability as a heat source.

PREPARATION OF PLUTONIUM-238

Plutonium-238 is produced by the neutron irradiation of neptunium-237 and subsequent beta decay of neptunium-238. Other plutonium isotopes are made during this process, so that the production grade plutonium-238 is diluted to about 80 percent of the total plutonium content. A typical isotopic analysis is shown in Table 1. The production of this isotope also increases the light element impurity level. A representative impurity analysis is shown in Table 2. Table 3 lists the thermal properties of plutonium-238 metal. It is readily seen that the diluting effect of the other plutonium isotopes reduces the specific power from 0.576 watt per gram to 0.460 watt per gram.

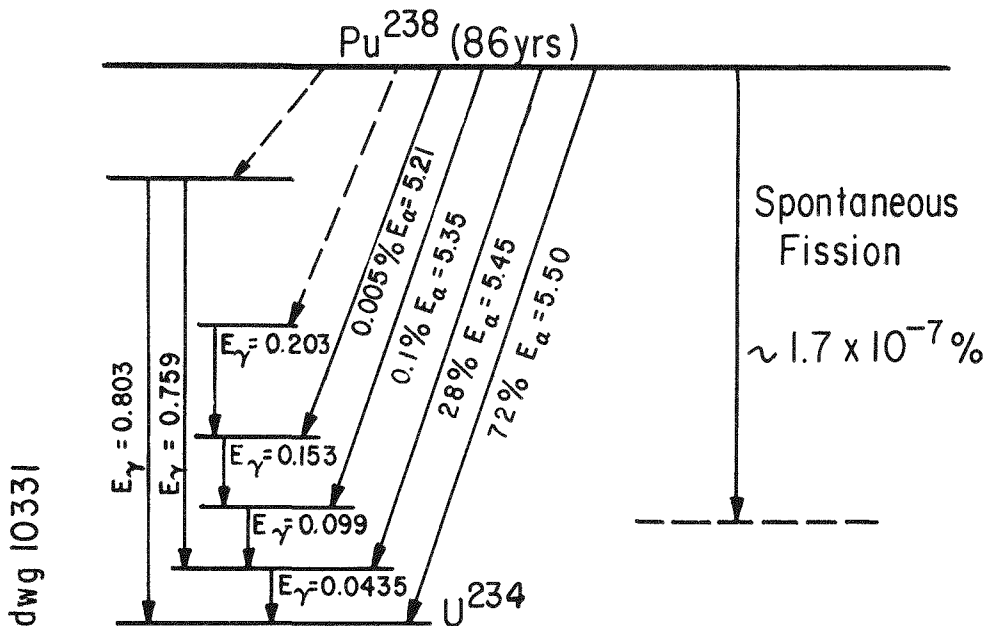


Figure 1. Plutonium-238 Decay Scheme and Energies (Mev)

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Table 1

TYPICAL ISOTOPIC ANALYSIS

Plutonium Isotope	Average Abundance (%)
238	80.0
239	16.5
240	2.5
241	0.8
242	0.08

Table 2

CHEMICAL IMPURITY ANALYSIS

Impurity	Weight (%)	Impurity	Weight (%)
Fe	0.08	Cr	0.08
B	<0.01	Ni	0.006
Si	0.002	Al	0.05
Mg	0.02	Cu	0.20

Table 3

PLUTONIUM-238 METAL CHARACTERISTICS

Characteristic	Value
Isotopic Purity	80%
Specific Power	0.460 watt/g
Density	15-16 g/cc
Thermal Density	7.13 watts/cc
Thermal Conductivity (Plutonium-239)	0.0098 cal/°C/cm/sec
Specific Heat (Plutonium-239)	
C _p	0.036 ± 0.002 cal/g/°C
C _v	0.032 ± 0.002 cal/g/°C
Melting Point Range	630 ± 10°C

The importance of the impurity level is reflected in the melting point of the metal. Plutonium forms low-melting eutectics with a number of elements. Among these are iron and nickel. A few-tenths of an atom percent of these impurities lowers the initial melting of the plutonium to as low as 410°C. Thus, it is quite imperative to keep the amounts of these impurities to a minimum.

For design purposes, plutonium metal is ideal for applications where the operating temperature of the fuel does not exceed 550°C. Past applications have been in the range of 500°C or less. For safety reasons,

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operating temperatures near and above the melting point of plutonium have been avoided since liquid plutonium becomes most difficult to contain for any extended period of time.

ALTERNATE FUEL FORMS

In seeking an alternate fuel, there are a number of factors to consider. High thermal density, elevated temperature stability, and low radiation levels are among the characteristics for which one looks. Three compounds - PuC, PuN, and PuO₂ - look most attractive in these respects.

The properties of PuO₂ (plutonium dioxide) are summarized in Table 4. Even though this compound does not offer the highest power density, its stability and ease of handling make it first choice among the alternate fuel forms. It is prepared by precipitation of plutonium oxalate from plutonium nitrate solution. Heating of the oxalate will convert this compound to PuO₂. The highest-density PuO₂ is prepared by passing small, rough-shaped particles through an induction-coupled plasma torch. Figures 2, 3, and 4 show these particles in the stages of rough, partially spheroidized and spheroidized particles, respectively. Multigram quantities of these particles have been made. X-ray investigation shows a stoichiometry of PuO_{1.98}. A study is presently underway to determine the variation of efficiency of conversion with particle size. Initial investigations show that below 35μ the efficiency is quite low and control of the radioactive dust becomes a serious problem.

Table 5 lists the properties of PuN (plutonium nitride). The thermal density of this compound is increased over that of the dioxide due to the lower diluting effect of nitrogen and the higher compound density. It is formed by the reaction of nitrogen with plutonium hydride at 600°C. Mound Laboratory personnel have prepared small amounts of this compound. There is a very serious handling problem with the nitride, however. It is pyrophoric, and upon exposure to very small amounts of air or moisture it rapidly converts to the dioxide. Plutonium nitride, with its extremely high melting point, offers unique application possibilities, nevertheless.



Figure 2. *Rough Plutonium Dioxide Crystals*

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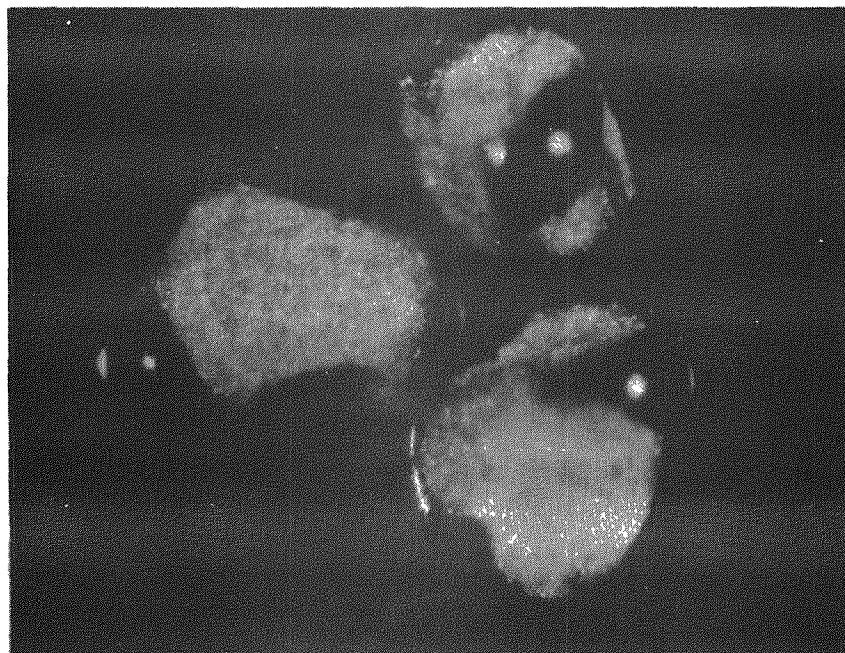


Figure 3. *Partially Spheroidized Plutonium Dioxide Crystals*

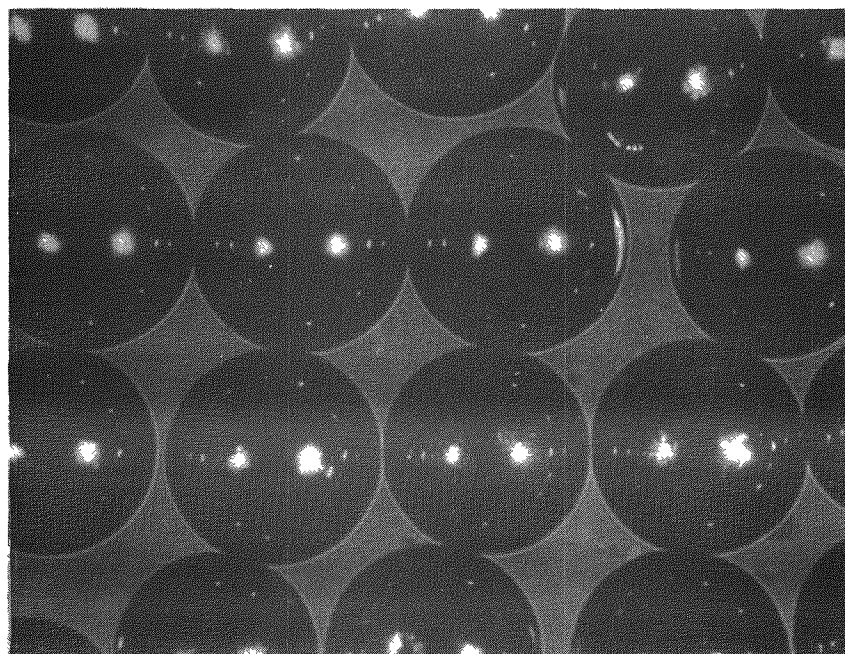


Figure 4. *Spheroidized Plutonium Dioxide Crystals*

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Table 4

PROPERTIES OF PLUTONIUM DIOXIDE

Property	Value
Specific Power	0.406 watt/g ^a
Density	11.4 g/cc
Thermal Density	4.63 watts/cc
Thermal Density (80% Packing Fraction)	3.60 watts/cc
Thermal Conductivity	--
Specific Heat	--
Melting Point	~ 2300°C

^aBased on theoretical density PuO₂: Pu²³⁸/Pu total = 80.00%

Table 5

PROPERTIES OF PLUTONIUM NITRIDE

Property	Value
Specific Power	0.434 watt/g
Density (X-ray)	14.23 g/cc
Thermal Density	6.17 watts/cc
Thermal Density (80% Packing Fraction)	4.93 watts/cc
Thermal Conductivity	--
Specific Heat	--
Melting Point	~ 2600°C

The properties of PuC (plutonium carbide) are listed in Table 6. At the present time this compound of plutonium-238 has not been prepared at Mound Laboratory. However, it has been reported¹ that graphite and plutonium hydride react at 800°C to give a good yield of Pu²³⁸C. This same report indicates that the handling problems would be similar to those of PuN.

Table 6

PROPERTIES OF PLUTONIUM CARBIDE

Property	Value
Specific Power	0.438 watt/g
Density (X-ray)	13.6 g/cc
Thermal Density	5.95 watts/cc
Thermal Density (80% Packing Fraction)	4.76 watts/cc
Thermal Conductivity	--
Specific Heat	--
Melting Point	~ 1850°C

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A comparison of the four fuel forms discussed is made in Table 7. The neutrons from the metal come from spontaneous fission. Of the alternate fuel forms, PuN has the lowest cross section for α, n reactions, as well as the highest thermal density and the highest melting point. It should be reiterated, however, that, considering all factors, plutonium dioxide would be the recommended fuel form for temperature applications above the melting point of plutonium.

Table 7

COMPARISON OF FUEL FORMS

Fuel Form	Thermal Density (watts/cc)	Melting Point (°C)	Neutron Activity (n/sec/g)
Pu-238 Metal	7.13	630	10 ³
PuO ₂	3.60	2300	10 ⁴
PuN	4.93	2600	10 ³
PuC	4.76	1850	10 ⁴

From a safety standpoint, the dioxide is insoluble. Indications are that in sea water this compound would be essentially inert. PuO₂ which has been sintered above 1000°C can be dissolved by sulfate fusion or by refluxing in HNO₃-HF.

COMPATIBILITY STUDIES

Compatibility studies of plutonium and its compounds have been directed towards the safe containment of these materials for many years at a time. The temperature of containment would be the operating temperature of a device such as a thermoelectric generator. The materials under study for the containment of the fuel forms have been the refractory metals and their alloys. Some of the super alloys (Haynes Alloy 25, Rene 41, Waspalloy, and Udimet 700) have been investigated for the containment of PuO₂. The phase diagrams of plutonium and some of the refractory metals are shown in Figure 5.² The predicted solubility of refractory metal in plutonium is low and appeared most promising from this standpoint as well as that of high-temperature strength for the containment of the alternate fuel forms. Table 8 shows representative results of the compatibility studies. The fact that molten plutonium cannot be contained in metal is not new. However, the fact that these 0.020-inch containers survived for two weeks gives reassurance that in the event of a short-term high-temperature excursion the metal would remain contained. At 1000°C there is no evidence of the plutonium wetting the cermet liners, which is encouraging for higher temperature applications. The containment of the metal at 550°C in a variety of materials has posed no problems up to this time.

Plutonium dioxide is compatible with the refractory metals, their alloys, and the super alloys at temperatures up to 1000°C. Long-term studies above this temperature are planned. Short-term compatibility tests indicate a very rapid reaction between plutonium dioxide and tantalum at 1400°C. Molybdenum and tungsten are unaffected at temperatures approaching 2000°C for short terms.

At the present time, no compatibility data on the nitride or carbide have been obtained. Future plans include compatibility studies of these compounds as well as higher temperature capabilities, and investigations of other fuel forms and materials of containment.

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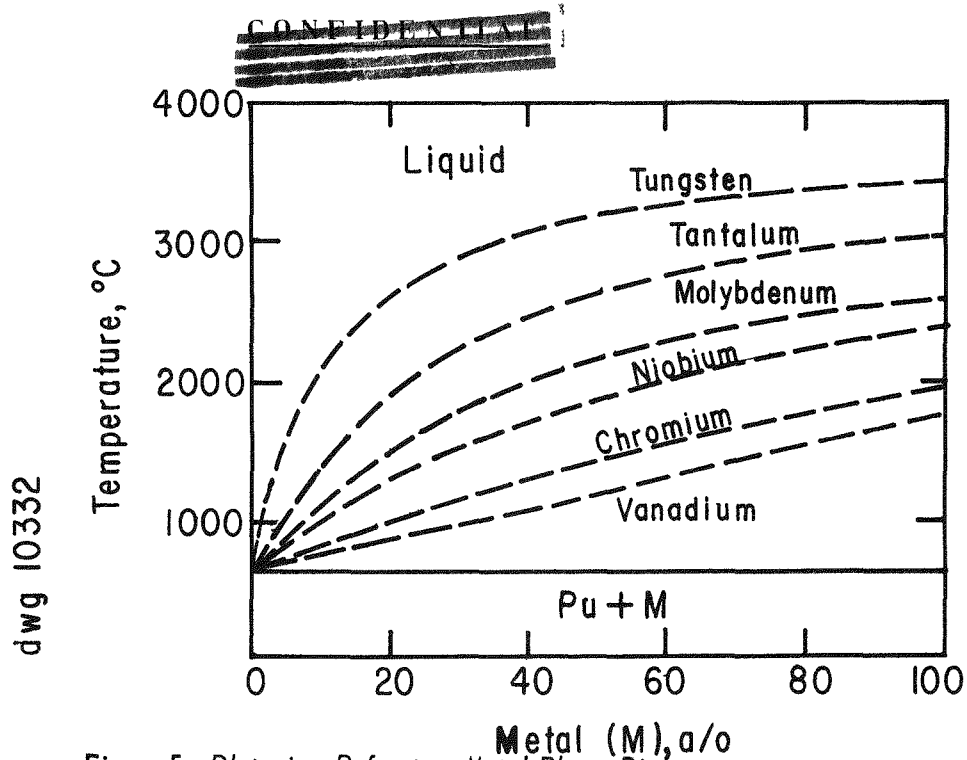


Figure 5. Plutonium-Refractory Metal Phase Diagram

Table 8

COMPATIBILITY RESULTS

Fuel Form	Container Material	Test Temperature (°C)	Test Duration (days)	Results
Pu-238 (Metal)	Ta	1000	12	Failure
	Re	1000	15	Continuing
	Cermets	1000	90	Continuing
	Ta	550	150	Continuing
PuO ₂	Ta	1000	143	Continuing
	Haynes Alloy 25	1000	143	Continuing

CONCLUSION

Past, present, and proposed future uses of the plutonium-238 isotope speak well in its behalf. With the wide limits of temperature applications of the isotope, there is little doubt but that its potential has only begun to be realized.

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2. F. W. Schonfeld, "Plutonium Phase Diagrams Studied at Los Alamos", *The Metal Plutonium*, A. S. Coffinberry and W. N. Miner (eds.), Chicago, University of Chicago Press, 1961, p. 247.