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PREPARATION AND PROPERTIES
OF PLUTONIUM CARBONITRIDE

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Certain plutonium compounds offer significant advantages over the metal for reactor use in that they may possess a higher melting point, greater corrosion resistance, or increased thermal stability. Plutonium compounds possessing some of these advantages include oxides, nitrides, and carbides. It is possible that an improved material could be obtained by a combination of two or more of these compounds.

The purpose of this work was to prepare and characterize plutonium carbonitride in hopes of obtaining a material with a high melting point and improved corrosion resistance.

Three possible methods of preparing the carbonitride were considered:

The plutonium-plutonium carbide cermet was prepared by the direct reaction of plutonium with different relative amounts of graphite at 1700°C . The concentration of the plutonium in the cermet varied from 28 to 67 v/o as determined by metallographic point counting. The composition of the final carbonitride was established by the carbon content of the cermet.

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The plutonium carbide was prepared by reacting stoichiometric amounts of plutonium and carbon at 1700°C.

Powdered plutonium carbide was prepared by grinding massive carbide under liquid Freon-113. The powdered mixture with a carbowax binder was cold pressed at 40-50 tsi prior to sintering.

The most promising of these methods appeared to be the reaction of plutonium carbide and ammonia. A still better procedure may be arc melting the carbide in a nitrogen atmosphere as briefly described by workers at Battelle¹. Because of the reactivity of the carbide with oxygen or additional carbon at high temperatures, close control of the atmosphere is necessary regardless of the method.

In most cases a pure single phase plutonium carbonitride did not result. The product generally contained small amounts of the unreacted reactants; therefore, the properties reported are not exact. They do, however, represent a close approxi-

¹ W. M. Pardue, et. al., Synthesis, Fabrication, and Chemical Reactivity of Plutonium Mononitride, USAEC Report BMI-1693 (September 1964).

mation. The values represent average or best values of the products made primarily by reaction of plutonium-plutonium carbide cermet with ammonia.

Melting Point

The carbonitride does not melt congruently; so, the values reported are actually decomposition temperatures and are a function of nitrogen pressure. Figures 1 and 2 show these results.

Density

			<u>Theoretical</u>
50 PuN-50 PuC	12.12 g/cc		13.9 g/cc
67 PuN-33 PuC	13.79 g/cc		14.0 g/cc

Solubility

<u>Composition</u>	<u>Solubility (g Pu/l)</u>			
	<u>6M HCl</u>	<u>3M HNO₃</u>	<u>6M H₂SO₄</u>	<u>5M H₃PO₄</u>
28 PuN-72 PuC	158	39.1	7.9	6.8
50 PuN-50 PuC	249	19.9	9.2	4.7
67 PuN-33 PuC	173	5.0	6.9	3.5

Corrosion

1. Air - The temperature at which appreciable oxidation starts:

28 PuN-72 PuC - 320°C

50 PuN-50 PuC - 220°C

67 PuN-33 PuC - 200°C

Figure 3 shows the corrosion rates at room temperature.

2. Water - Rapid corrosion was observed for all carbonitride compositions in water.

Plutonium carbonitride appears to be an interesting and potentially useful material because of its possible broad range of stoichiometry, its high melting point and the resistance to air oxidation of certain compositions. Some unexplained inconsistencies are observed in these preliminary results such as the apparent lack of any relation between the corrosion behavior and the composition.

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FIGURE 1

MELTING POINTS OF PLUTONIUM CARBONITRIDES AT 1.0 ATMOSPHERE NITROGEN PRESSURE.

FIGURE 2
MELTING POINTS OF PLUTONIUM CARBONITRIDES

FIGURE 3
CORROSION IN AIR OF PLUTONIUM CARBONITRIDES