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Lawrence Radiation Laboratory

UNIVERSITY OF CALIFORNIA

LIVERMORE

**Materials Aspects of U^{233} , U^{235} and Pu^{239}
for the Space Power Reactor**

J. S. Kane

November 1, 1966

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SPACE POWER NOTE NO. 158 - November 1, 1966

TO: Distribution

FROM: J. S. Kane

SUBJECT: Materials Aspects of U^{233} , U^{235} and Pu^{239} for the Space Power Reactor

Introduction

In this memorandum I would like to discuss the relative merits of the fissile isotopes U^{233} , U^{235} and Pu^{239} . The considerations here will be from the aspect of materials properties, rather than suitability for a specific nuclear design. In turn, the materials questions reduce to: 1) the comparative performance of the compounds of these isotopes when used in high temperature nuclear reactors, and 2) the difficulties encountered in handling them for research, development and reactor fabrication.

Properties of Uranium Compounds Compared with Analogous Plutonium Compounds

To make an assessment of possible relative performance of a compound containing a fissile isotope, it is necessary to postulate failure mechanisms. In high temperature nuclear reactors, the fuel can cause problems by one or all of the following mechanisms. (Plus others I have no doubt overlooked.)

1. Vaporization of fuel, causing either excessive pressures or transport and loss of fuel.
2. Melting. The resulting liquid then reacts with other components of the system.
3. Poor physical properties -- poor thermal transport, low density of fissile atoms, low strength, poor creep resistance, etc.
4. Excessive dimensional changes with burn up.
5. Rapid diffusion of fuel components through structure, etc.

Plutonium and uranium are chemically quite similar, and especially their refractory compounds with other elements are quite alike. The most noteworthy exception to this is that U is more easily oxidized to the +6 state, and therefore the oxide systems differ, especially at high oxygen pressures. But under more reducing conditions, as would be typical of space power applications, the compounds of the two metals are quite similar, viz:

Metal	Compounds found with		
	C	N	S
Pu	Pu_3C_2 , PuC, PuC_2 , Pu_2C_3	PuN	PuS, Pu_2S_3
U	UC, UC_2 , U_2C_3	UN, U_2N_3 , UN_2	US, U_2S_3

Indeed, most of the refractory compounds are soluble in their analogues; that is UC and PuC can be mixed in any proportion, and this is probably true of the others. Thus in these compounds a continuous spectrum of nuclear properties could be obtained by appropriate mixtures. The associated handling problems, however, would be horrendous, both from the aspect of safety and of bookkeeping.

In general, the compounds of Pu are not as "refractory" as those of U. In the interstitial type compounds, the stability comes at least in part from metal-metal bonding, and this is weaker for Pu than for U. (The heat of sublimation of Pu is ~ 80 kcal/mole; that of U ~ 125 kcal/mole.) Both the melting points and the sublimation temperatures are lower for the compounds of Pu than for U. It would therefore seem that one would pay a penalty in temperature if Pu is substituted for U in the space power reactor. Just what the temperature decrement would be would have to be learned by experiment.

A comparison of the five postulated failure mechanisms stated earlier can be guessed at for the carbides and nitrides of U vs. Pu.

1. PuC and PuN will certainly volatilize at a lower temperature than the U compounds.
2. The melting points of Pu compounds are lower.

PuC decomposes peritectically at $\sim 1650^{\circ}\text{C}$, UC melts congruently (?) at 2560°C .

Pu_2C_3 decomposes peritectically at $\sim 2025^{\circ}\text{C}$, U_2C_3 is unstable above $\sim 1800^{\circ}\text{C}$.

PuN forms liquid metal at a given N_2 pressure at a lower temperature than UN. The dissociation pressure of PuN is about three times that of UN at $\sim 2500^{\circ}\text{C}$.

3. Physical properties would be similar for U and Pu compounds. Mechanical properties would be poorer for Pu compounds at elevated temperatures.
4. Deformation on burn-up would be greater for Pu compounds, since the strength and creep resistance are lower at high temperatures.
5. I would guess that Pu would diffuse through metals, etc., faster than U.

Handling Problems*

The cost of research and development of radioactive reactor fuels is highly dependent on the ease with which they can be handled. Some comments on this point seem appropriate.

* Information courtesy of W. J. Ramsey.

For uranium, most of the research and development can be done on U^{238} , which presents very modest problems; thus many of the mechanical, physical and chemical questions can be studied easily for reactor concepts based on U^{233} or U^{235} . This is not true of the nuclear experimentation, of course. For plutonium, which is mostly Pu^{239} but which also contains other isotopes, there is no similar "stand-in", and all experimentation must be done on Pu^{239} . This isotope is ~ 100 times more toxic than pure U^{235} , and handling problems would greatly increase research, development and fabrication costs.

U^{233} is the most expensive of the fissile isotopes, and its handling costs would also be the greatest. As produced, U^{233} is often contaminated with a small amount of U^{232} , whose daughters emit hard gamma rays. This penetrating radiation increases as the daughters grow in. To handle U^{233} safely, facilities similar to those used for Pu are necessary, with the added expense and complication of shielding or perhaps even remote handling equipment.

JSK:mal

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