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HIGH-TEMPERATURE COMPATIBILITY TESTING OF MOLTEN PLUTONIUM FUEL-CONTAINER SYSTEMS*

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One of the major problems in the development of a fast breeder reactor fueled with molten plutonium is compatibility of the fuel and its container. An approach to this problem is to test the fuel-container systems at relatively high temperatures (900-1150°C) and to extrapolate the results to lower temperatures (600-700°C) more practical for reactor operation. The objective is to greatly reduce the time required to obtain corrosion information; a factor of 100 or more will be demonstrated. The present summary describes the results of high-temperature studies on two fuel systems: plutonium contained in tantalum capsules and 97.5 w/o plutonium - 2.5 w/o iron - gallium stabilized fuel alloy contained in tantalum capsules. The tests were conducted in vacua by a technique which has been described earlier.^{1,2}

The experimental results are presented in Fig. 1, a plot of capsule lifetime as a function of reciprocal temperature. Data for the low-temperature portion of the Pu-Fe curve were taken from capsule tests³ conducted in isothermal sodium loops. Capsule lifetimes for the Pu-Ta system are much shorter than those for the Pu-Fe - Ta system.

The linear relationship obtained is strong evidence for a constant capsule-failure mechanism over the temperature range, and demonstrates the validity, for the Pu-Fe - Ta system, of extrapolating high-temperature test data to lower temperatures. The data also imply no observable difference between high-purity sodium (<1 ppm oxygen) and high vacuum (1×10^{-7} torr) as an external capsule environment during testing.

From a combination of metallographic and microautoradiographic techniques, the failure mechanism was found to be intergranular penetration of plutonium through the capsule wall. Assuming a constant penetration rate, overall rates through the capsule wall were calculated at several temperatures for the Pu-Fe curve of Fig. 1. From an Arrhenius plot of

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Fig. 1. Temperature Dependence of Capsule Lifetime.

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these data the temperature dependence of the penetration rate, P , may be expressed as

$$P = 0.064 e^{-30,400/RT} \text{ cm/sec.}$$

The activation energy (30.4 kcal) in the above expression is not unreasonable for a grain-boundary diffusion process;⁴ if anything, the activation energy may be a little low, indicating enhanced diffusion. Other processes, such as the high reactivity of Pu with grain boundary interstitials,⁵ grain boundary wetting,³ and the formation of a Ta-Fe reaction layer which has been observed on the internal capsule wall in some tests, may also affect Pu penetration.

A capsule treatment which prevents or greatly retards plutonium penetration through the capsule wall is carburization of the tantalum capsule under conditions which result in diffusion of carbon partially through the tantalum grain boundaries. Although the mechanism is not fully understood, the interaction of carbon, tantalum and plutonium in the grain boundaries blocks plutonium penetration. As an example, in two tests (Fig. 1), capsules containing Pu-Fe fuel were externally carburized and tested at 1000°C and 1100°C. The runs were stopped, before failure, after 660 and 380 hours at temperature, respectively. These numbers represent improvements over the average lifetimes (Fig. 1) by factors of at least 13 and 16. If the capsules had been tested to failure, the factors might have been much larger. Work is currently in progress to characterize optimum conditions for carburization of Ta, and to determine the physical properties of the final product. It is already known⁶ that Ta can contain considerable Ta_2C in the grain boundaries without detrimental effects in tensile strength or ductility.

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