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An Analysis on the TRISO Failure due to Pd-SiC Interactions in a 350-MW_{th} Prismatic HTR Fuel

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1. Introduction

A tri-structural isotropic particle (TRISO) is a coated fuel particle of a high temperature reactor (HTR) fuel which consists of a kernel, a low-density pyrocarbon layer called a buffer, an inner high-density pyrocarbon (IPyC) layer, a silicon carbide (SiC) layer, and an outer high-density pyrocarbon (OPyC) layer. During irradiation, fission product metals such as palladium and rare earths are generated in the kernel. They react chemically with the SiC layer of a TRISO and deteriorate the layer [1-5].

It has been known that, in all fuel kernel types, palladium is released from the kernel and moves to the inner surface of the SiC layer [1,4]. But rare earths such as lanthanum, cerium, praseodymium, and neodymium form oxides within a kernel with an oxygen-to-uranium (O/U) ratio of 1.1 or more [1]. At temperatures below 1400 ºC, palladium corrodes the SiC layer at rates faster than the rare earths. But the reverse is true at temperatures above 1400 ºC [1]. The normal operation temperature of an HTR is much less than 1400 ºC. Therefore, palladium is the main corroder of the SiC layer in a UC_xO_y HTR fuel, where x and y are typically 1.5 and 1.9, respectively.

This study treats the calculation of TRISO failure due to the Pd-SiC reactions under the irradiation conditions of a 350-MW_{th} prismatic HTR. The calculated failure fraction was compared with the results calculated using a USNC's computer code TRIPPER [6].

2. Pd-SiC Interactions

2.1. Mechanisms

The fission product palladium is generated in a kernel during irradiation. Palladium isotopes are produced more in low-enriched uranium kernels than in high-enriched uranium kernels because the fission yield of palladium from plutonium is much larger than that from uranium. The palladium atoms in a kernel diffuse out of the kernel, and then, they transport to the SiC layer through the buffer and IPyC layers. The Pd-SiC interaction starts to occur at the inner surface of the SiC layer. Very small nodules of a noble metal compound occur along SiC grain boundaries [3]. SiC dissociates at the leading edge of a nodule through the following possible chemical interaction [5]:

$$2Pd + SiC \rightarrow Pd_2Si + C \quad (1)$$

Significant restructuring does not appear behind the nodules. This leads to local corrosion and thinning of the SiC layer.

2.2. Modeling

The threshold amount of palladium for the Pd-SiC reaction has not been clearly observed [1,5]. It can be assumed that the SiC begins to corrode once a small amount of palladium reaches the inner surface of the SiC layer. Minato et al. [5] suggested that, in a UO_2 kernel, the release from a kernel limits the Pd-SiC interaction rate. They concluded qualitatively that the SiC corroded even in the small number of palladium atoms, and the corrosion depth of SiC is proportional to the one third power of the number of palladium atoms.

Montgomery [2,4] introduced the following corrosion rate of SiC through a data base consisting of observations of SiC attack by fission products in real time irradiation and out-of-pile heating:

$$\dot{v} = 2.613\times10^{5} e^{-\frac{252200}{RT}} , \quad (2)$$

where $\dot{v}$ = the corrosion rate (μm/h), T = the temperature (K), R = the gas constant (8.314 J mol^{-1} K^{-1}). Eq. (2) describes only the temperature dependence of the corrosion rate, but not the enrichment or heavy metal burnup dependence of the reaction rate. Since the data used to obtain Eq. (2) were scattered, it is hard to conclude that the reaction rate is explained very well only by temperature.

The rate of SiC corrosion by palladium typically depends on irradiation temperature, kernel composition, palladium concentration, other fission products, and SiC properties. A correlation describing the Pd-SiC reaction must include the effects of the above factors.

3. Calculation of the TRISO failure fraction

The information related to the design and operation of a 350-MW_{th} prismatic HTR is described in Ref. [7]. McCARD [8] was used to generate the burnup and depletion data, and COPA [9,10] was used to perform the failure fraction analysis.

3.1. Fuel burnup and depletion

Fig. 1 displays the burnup and fast fluence histories of the HTR fuel throughout 1500 effective full power days (EFPDs). The final burnups are 152 GWd/tHM at

packing fraction (PF) of 25 %. The final fast fluence approaches to 8×10^{21} n/cm^2, E_n > 0.1 MeV. Fig. 2 shows the amount of palladium generated in a kernel during irradiation. The "Pd total" graph means the total sum of palladium isotopes produced which exist in both atomic state and chemical compound in a kernel. The graphs with symbols indicate pure palladium atoms which are released from a kernel and go to the SiC layer. The palladium atoms per particle increases with the temperature and time. At 1500 EFPDs, it is about 1.4×10^{15} to 6.3×10^{15} between 800 and 1400 °C. In Fig. 3, the number of palladium atoms released from a kernel is about 3.3×10^{11} to 2.5×10^{15} between 800 and 1400 °C. The release of palladium from a kernel is negligible below 1100 °C. The release amount of palladium is about 10 % of palladium atoms in a kernel at 1200 °C, and about 40 % at 1400 °C.

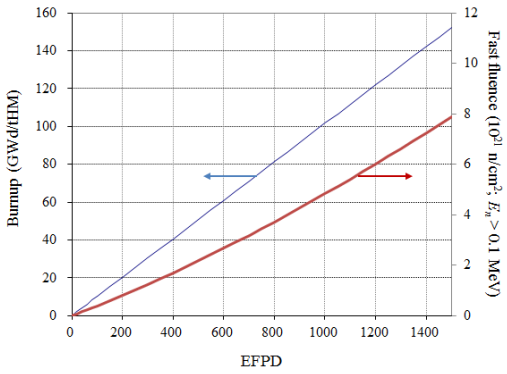

Fig. 1. Fuel burnup and fluence.

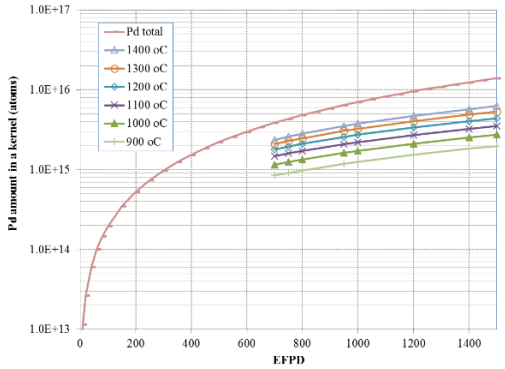

Fig. 2. Generation of palladium in a kernel during irradiation.

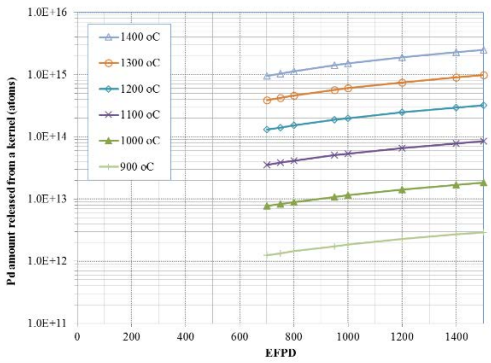

Fig. 3. Release of palladium from a kernel during irradiation.

3.2. Failure Fraction of TRISOs

Figs. 4 and 5 show the failure fractions of TRISOs at various temperatures, which were calculated using COPA and TRIPPER. The COPA and TRIPPER results are in good agreement. The TRISO failure occurs at an earlier point of time at the elevated temperatures, and it is not significant at temperatures below 1200 °C. According to COPA results in Fig. 5, significant failure is delayed for about one hundred days when the temperature is being maintained at 1200 °C for the first two hundred days and then jumps to 1300 °C.

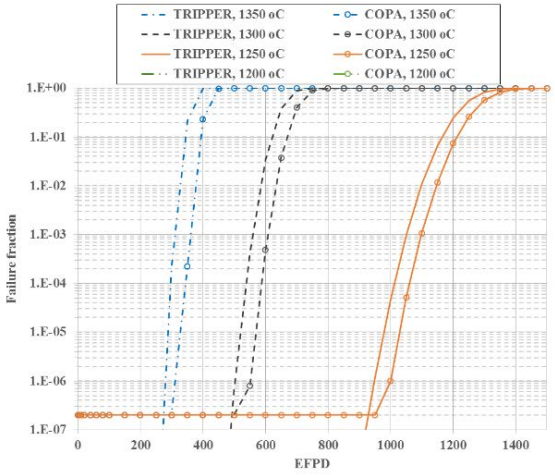

Fig. 4. Failure fraction of TRISOs under constant irradiation temperatures.

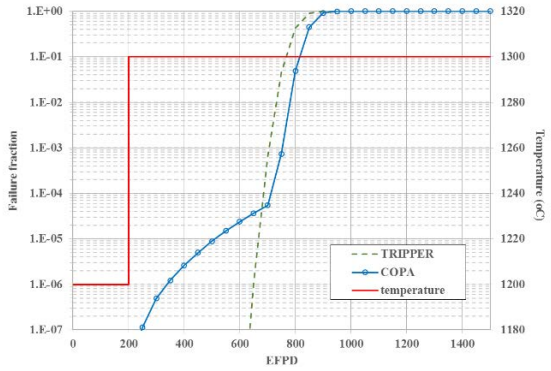

Fig. 5. Failure fraction of TRISOs under jumped irradiation temperature.

4. Summary

The mechanism and modeling on the reaction of palladium with the SiC layer of an HTR TRISO were reviewed. An analysis of TRISO failure due to Pd-SiC interactions has been done for a 350-MW_{th} prismatic HTR, whose fuel burnup and fast fluence are about 152 GWd/tHM and 8×10^{21} n/cm^2, E_n > 0.1 MeV at 1500 EFPD and packing fraction of 25 %, respectively.

- Kernel composition influences the birth of palladium during irradiation because the fission yield of palladium depends on fissile materials. The diffusion from a kernel limits the Pd-SiC interaction rate in a UO_2 kernel.
- The currently developed correlation for the Pd-SiC reaction rate describes only the temperature dependence of the rate. It must be modified to include the effects of other factors such as kernel composition, palladium concentration, other fission products, and SiC properties.
- The release of palladium from a kernel increases with temperature. The number of palladium atoms released is negligible below 1100 ^oC, about 10 % at 1200 ^oC, and about 40 % at 1400 ^oC.
- The TRISO failure due to the chemical attack of palladium is negligible at temperatures below 1200 ^oC. The failure fractions calculated using COPA and TRIPPER are in good agreement.

ACKNOWLEDGEMENTS

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