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INTERACTION OF NOBLE-METAL FISSION PRODUCTS WITH PYROLYTIC SILICON CARBIDE

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Fuel particles for the High-Temperature Gas-Cooled Reactor (HTGR) contain layers of pyrolytic carbon and silicon carbide, which act as a miniature pressure vessel and form the primary fission product barrier. Of the many fission products formed during irradiation, the noble metals are of particular interest because they interact significantly with the SiC layer and their concentrations are somewhat higher in the low-enriched uranium fuels currently under consideration. To study fission product-SiC interactions, particles of UO_2 or UC_2 are doped with fisision product elements before coating and are then held in a thermal gradient up to several thousand hours.[1] Examination of the SiC coatings by TEM-AEM after annealing shows that silver behaves differently from the palladium group.

Palladium Group

Figure 1 shows the typical SiC microstructure as deposited at 1500°C and 0.70 μm/min. The SiC coating from a UC_2-Mo-Ru-Rh-Pd particle is shown in Fig. 2 after annealing 260 h at 1597°C in a thermal gradient of 275°C/cm. Small (⩽1 μm) nodules of a noble metal compound occur along SiC grain boundaries. No significant restructuring of the SiC behind the nodules is evident, suggesting that they move by dissolving SiC at the leading edge and by forming SiC at the trailing edge. Analysis of the nodules by EDX indicated Pd, Rh, U, and Si, which is consistent with electron microprobe results, except that prior observations could not differentiate Pd and Rh because of overlapping $L\alpha$ lines. Our analysis with the JEM-100CX resolved Pd and Rh $K\alpha$ lines. A few larger nodules observed in this specimen had a more equiaxed shape and appeared to contain only U and Si. Migration of U is seen only in carbide-based fuel particles.

Silver

In concentrations typical of end-of-life fuel, silver neither attacks the SiC layer nor forms second phases as does the Pd group. However, silver is sometimes released from particles even when the SiC layer is intact.[2] Figure 3 shows the microstructure of the SiC coating from a UO_2-Ag particle after annealing for 2000 h at 1450°C in a thermal gradient of 275°C/cm. Note the similarity to the as-deposited structure. No second phases or grain boundary films are observed in this coating. Analysis by EDX detected a large concentration of Ag accumulating at the inner surface of the SiC, but no Ag was detected along several grain boundaries probed with a 100-Å spot. The kinetics of Ag transport through SiC suggest a grain boundary mechanism.[2] Our results suggest that, if a grain boundary layer of Ag (or an Ag-Si alloy) exists in this sample, it is exceedingly thin. We plan to use lattice fringe imaging or higher resolution EDX to try to resolve this layer.

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[1]T. B. Lindemer and R. L. Pearson, *J. Am. Cer. Soc.* 60(1–2): 5–17 (1977).
[2]H. Nabielek, P. E. Brown, and P. Offermann, *Nucl. Technol.* 35: 483–93 (1977).

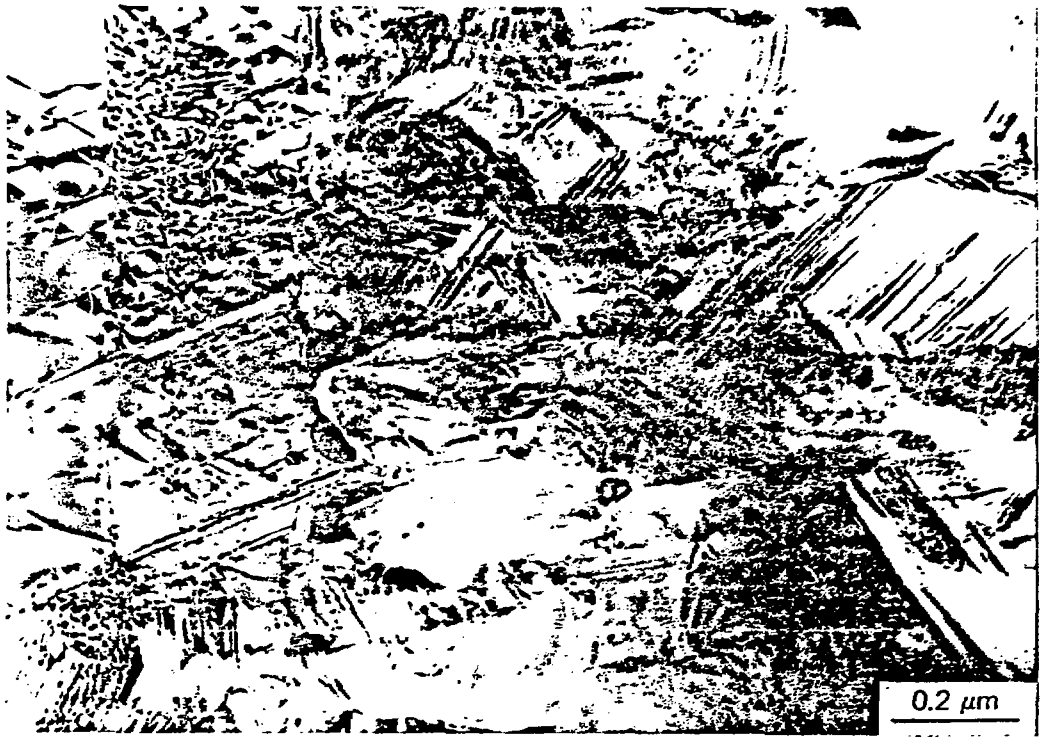

Fig. 1. Pyrolytic SiC as deposited. Growth direction is right to left.

Fig. 2. Nodules of Pd-Rh-Si alloy at SiC grain boundaries after annealing UC_2-Mo-Rh-Ru-Pd particle.

Fig. 3. SiC coating on Ag-doped UO_2 particle after annealing.