EFFECTS OF IRRADIATION AND POST IRRADIATION ANNEALING ON THE THERMAL CONDUCTIVITY/ DIFFUSIVITY OF MONOLITHIC SIC AND SIC/SIC COMPOSITES—G. E. Youngblood, D. J. Senor, and R. H. Jones (Pacific Northwest National Laboratory)*

Extended abstract of a paper submitted to the Journal of Nuclear Materials as part of the proceedings of the 11th International Conference on Fusion Reactor Materials, Kyoto, Japan, December 7-12, 2003.

EXTENDED ABSTRACT

Laser flash thermal diffusivity measurements were made on high-purity monolithic CVD-SiC (impurity concentration <5 wppm) and 2D f-SiC/PyC/ICVI-SiC composite samples (plain weave Hi-Nicalon™ fabric layers with 0-90 layup made by the isothermal chemical vapor infiltration process and with either a "thick" 1.0 µm or a "thin" 0.11 µm PyC fiber coating) before and after irradiation in the HFIR reactor (250 to 800°C, 4-8 dpa-SiC) and after post-irradiation annealing composite samples to 1200°C.

Thermal conductivity in SiC is controlled by phonon transport. Point defects introduced into SiC during neutron irradiation are effective scattering centers for phonons, and as a consequence the thermal conductivity is sharply reduced. For irradiation temperatures below ~800°C, the accumulation of point defects (in SiC mostly single or small clusters of interstitials and isolated vacancies) saturates when the interstitial-vacancy recombination rate equals the defect production rate. For saturation conditions, the relative reduction in the SiC thermal conductivity decreases in a manner similar to its swelling reduction with increasing irradiation temperature. Examination of SiC swelling data at various irradiation temperatures and doses indicates that saturation occurs for ~2 dpa-SiC at 200°C and decreases continuously to ~0.4 dpa-SiC at 800°C. Based on a model that assumes a uniform distribution of the phonon scattering defects, the calculated defect concentration for unirradiated CVD-SiC was less than 1 appm, which is consistent with the manufacturer's value of <5 wppm impurities. The defect concentrations estimated for the irradiated CVD-SiC samples decreased continuously from ~25,000 to 940 appm as the irradiation temperature increased from 252 to 800°C. The small intrinsic defect concentration in comparison to the rather large extrinsic irradiation-induced defect concentrations illustrates why CVD-SiC makes an ideal irradiation damage monitor.

in Figure 1, the temperature dependence of K_{ir}/K_o, the ratio of the irradiated-to-unirradiated thermal conductivity values measured at the irradiation temperature, is graphically presented. The general trend that the K_{ir}/K_o-ratio increases as the irradiation temperature increases reflects the relative dominance of temperature independent point defect phonon scattering in the lower temperature range, while the temperature dependent phonon-phonon scattering becomes relatively more important with increasing temperature. The curve for high-purity CVD-SiC represents a lower limit for SiC-based materials with K_{ir}/K_o ~ 0.05 at 200°C and only gradually increasing up to ~0.12 by 800°C. For irradiation temperatures above 800°C, an apparent transition occurs where the K_{ir}/K_o-curve turns upward with an increasing slope until at 1100°C the ratio is ~0.6. Above ~800°C, the defect structure formed by the highly mobile irradiation-induced interstitials must become relatively coherent with the surrounding SiC matrix and presents less effective phonon scattering centers. Furthermore, above 800-1000°C the equivalent dose required for saturation also appears to "turn around" and starts to increase with increasing dose as evidenced by the dose dependence at ≈1 dpa observed at 1100°C.

In Figure 2, the measured thermal diffusivity $\alpha(T)$-values for the Hi-Nicalon™ composite with the "thick" PyC interface are presented for various conditions. When the PyC interfaces were removed by oxidation, the α-values dramatically decreased to about 40% of their as-received values. Such a large degradation indicates that entire fiber bundles (including the SiC matrix material contained within the bundles) were separated (thermally decoupled) from the surrounding SiC matrix. For the irradiated and annealed

*Pacific Northwest National Laboratory (PNNL) is operated for the U.S. Department of Energy by Battelle Memorial Institute under contract DE-AC06-76RLO-1830.

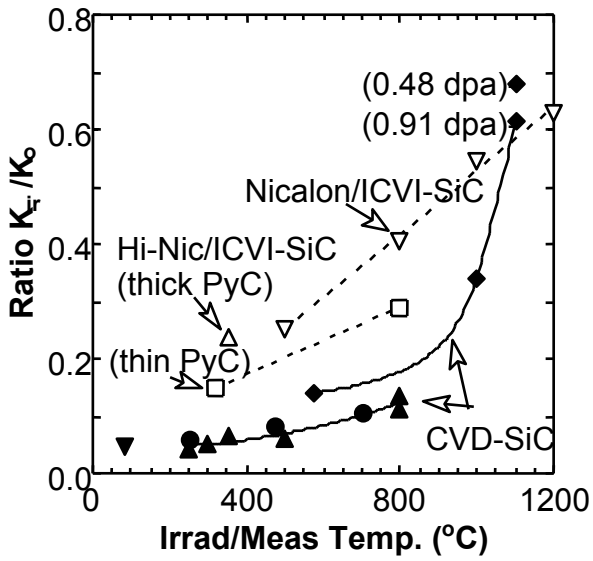

Figure 1. Ratio K_{ir}/K_o for high-purity CVD-SiC and SiC and for 2D f-SiC/SiC composites.

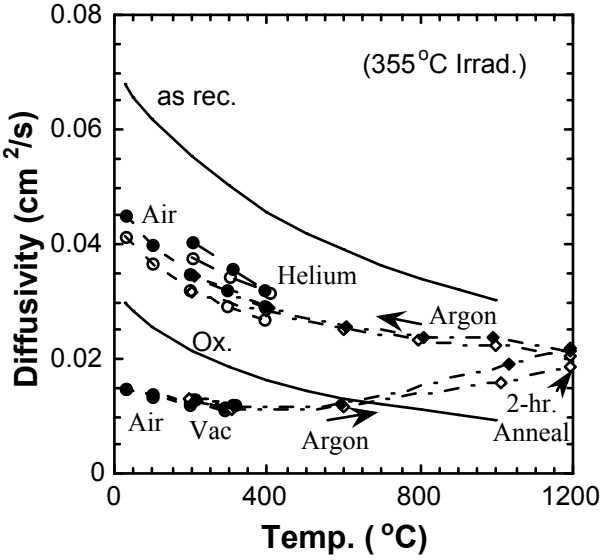

Figure 2. Thermal diffusivity of 2D Nicalon/ICVI-SiC composites with a "thick" PyC interface for various unirradiated (solid lines) and irradiated and/or annealed (dashed lines) conditions.

conditions, close agreement of the α(T)-values made in vacuum, argon or air just after irradiation indicate that at this point physical fiber/matrix (f/m) separations did not exist even though the matrix was expected to swell ~0.8% and the Hi-Nicalon™ fiber was expected to shrink ~1.7%. Apparently, the radial swelling in the irradiated PyC fiber coating compensated somewhat for the differential shrinkage-swelling between the f/m components, and significant f/m thermal decoupling didn't occur. In contrast, when the Hi-Nicalon™ composite with a "thin" PyC interface was irradiated at 330 or at 800°C and then taken through the same measurement and annealing sequence, the α(T)-values were substantially different in different atmospheres just after irradiation (data not shown). For this case, radial swelling in the irradiated "thin" PYC interface was insufficient to close the f/m separations and at least partial f/m thermal decoupling occurred. However, after annealing to 1200°C and cooling to the irradiation temperature, substantial f/m debonding (or decoupling) occurred for both cases due to permanent net shrinkage in the fiber and the PyC interface components.

A thermal conductivity model based on composite constituent properties as well as microstructural and architectural variables was used to predict α(T)-values for the irradiated composites. The predictions closely matched the observed magnitudes and slopes of the measured α(T)-values after irradiation. Input to the model required that the major contribution to K_{eff} for the irradiated and annealed Hi-Nicalon composites was the conductivity of a still-continuous SiC matrix, which indicates that the major cause of the degradation in K_{eff} was the accumulation of the irradiation-induced point defects in the matrix constituent.

For the unirradiated Hi-Nicalon/PyC/ICVI-SiC composites, the K_{eff}-values calculated from the α(T)-data ranged from a maximum 15 W/mK at 200°C down to 8-10 W/mK at 1000°C. Then from estimated K_{ir}/K_o-values for these composites (also shown in Figure 1), K_{ir}(T) values can also be estimated.

Acknowledgements

The irradiation tests were performed as part of the U.S./Japan JUPITER fusion materials collaboration.