REDUCTION IN THERMAL CONDUCTIVITY DUE TO NEUTRON IRRADIATION

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INTRODUCTION

Graphite has been used in every major tokamak machine as a plasma facing material primarily due to it good strength and the low atomic number of carbon. Recently, graphite composites have been considered for the International Thermonuclear Experimental Reactor (ITER) because of the good thermal shock resistance and the excellent thermal conductivity of some advanced composites. One concern regarding the use of high conductivity composites in the ITER environment is how much neutron-induced degradation in thermal conductivity will occur during reactor operation.

The physical processes governing the thermal conductivity of graphites, as well as the mechanisms responsible for the radiation-induced degradation in conductivity, have been well established [1]. For all but the poorest grades of carbon, the thermal conductivity is dominated by phonon transport along the graphite basal planes and is reduced by scattering "obstacles" such as grain boundaries and lattice defects. For graphites with the largest crystallites, i.e. pyrolitic graphite or natural flake, the in-plane room temperature thermal conductivity approaches 2000 W/m-K [2].

The thermal conductivity of graphite-based materials can be written as a summation of the thermal resistance due to scattering obstacles:

$$(1)\quad K(x,T) = \beta(x)\left(\frac{1}{K_u} + \frac{1}{K_{GB}} + \frac{1}{K_i}\right)^{-1}$$

where the term $\beta(x)$ is a coefficient which includes terms due to orientation (with respect to the basal plane), porosity and some other minor contributors. This coefficient is in most cases assumed to be constant with temperature, with a value of around 0.6 . The first two terms inside the parentheses are the contributions to the thermal conductivity due to the umklapp scattering (K_u) and the grain boundary scattering (K_{gb}). The grain boundary phonon scattering ($1/K_{gb}$) dominates the thermal resistance at low temperatures and is insignificant above a few hundred degrees Celsius, depending on the perfection of the graphite. The umklapp scattering, which defines the phonon-phonon scattering effect on the thermal conductivity, dominates at higher temperatures and scales nearly as T^2 [1]. The umklapp scattering therefore defines the upper limit to the thermal conductivity for a "perfect" graphite. Following Taylor's analysis,[3] the umklapp-limited thermal conductivity of the graphite crystal would be ~ 2200 W/m K at room temperature, which is not far removed from the best pyrolitic graphites or the vapor grown carbon fibers mentioned earlier.

The third term in equation (1), K_i, is the contribution to the thermal resistance in the basal plane due to defect scattering. Following neutron irradiation, various types of stable defects will be produced depending on the irradiation temperature. These defects can be very effective in scattering phonons at flux levels which would be considered modest for fusion power reactors, and would quickly dominate the other terms in equation (1). Several types of defects have been identified in graphite, though at irradiation temperatures less than ~650°C, only simple defects are found in significant quantities. It has been further shown[4] that at an irradiation temperature near 150°C, the defect which dominates the thermal resistance is the lattice vacancy. For temperatures from ~150-1000°C small vacancy loops become important.

RESULTS

The following will give a semi-empirical relationship for the degradation of the Mitsubishi-Kasei MKC-1PH composite material as a function of irradiation temperature and neutron displacement dose. In this analysis the temperature dependence of the terms for the umklapp and grain boundary scattering in equation (1) follow Kelly's analysis [5]. Using many references [6-10] on the thermal conductivity taken at the irradiation temperature of intermediate thermal conductivity graphites and composites, the defect scattering term in equation one has been replaced with a empirical radiation damage term which is a function of irradiation temperature and displacements per atom (dpa) :

$$(2)\quad R(dpa,T) = K(dpa,T)/K_{unirr}(T) = -M(T)\log(dpa) + S(T)$$

$$M = 0.25 - 0.00017\,T(°C) \text{ and } S(T) = 0.000683\,T(°C)$$

which then yields:

$$(3)\quad K(x) = R(dpa,T) \bullet \beta(x)\left(\frac{1}{K_u} + \frac{1}{K_{GB}}\right)^{-1}$$

where the thermal conductivity as limited by umklapp scattering has been calculated [5] for basal planes in graphite and can be adequately fitted by the following:

$$(4)\quad K_u(T) = 91.285 \bullet T(°C)^{-0.4279}$$

Range from 0 to 1200 °C

and:

$$(5)\quad K_{GB}(T) = \left[\frac{La(Å)}{21,000}\right] f(T)$$

Range -272 to 1200 °C

where La is the grain size taken to be 6000Å and f(T) is related to the basal conductivity of a graphite crystal. The term f(T: °C) can be fitted from Kelly's data [5] as:

$$(6)\quad f(T) = 126.06 + 0.6007\,T - 2.474\times10^{-4}\,T^2 - 1.6741\times10^{-6}\,T^3 + 2.8591\times10^{-9}\,T^4 - 1.3252\times10^{-12}\,T^5$$

Given in figures 1 and 2 are the results from this algorithm. Figure 1 gives the normalized thermal conductivity at the irradiation temperature from the radiation damage term of equation (2). Figure 2 gives the absolute thermal conductivity in the high conductivity direction for the MKC-1PH material as calculated from equation (3).

The radiation damage function (figure 2) was generated using available data on irradiated intermediate thermal conductivity materials and pyrolitic graphite [6-10]. The linearity assumed in this algorithm is accurate within the temperature range of figure 2 (300-1200 °C), but seriously overestimates the residual thermal conductivity for temperatures below 300°C. Below this temperature the production of stable defects increases dramatically, presumably due to the lack of both interstitial and vacancy mobility. It should also be noted that for lower thermal conductivity materials (less than about 150 W/m-K) at room temperature, the existing database varies quite widely and the radiation damage function yields large errors.

It is apparent that data on the thermal conductivity of these high thermal conductivity C/C composites is needed to accurately develop a model such as presented here. This data currently unavailable. The only source of data [11] on the MKC-1PH (or similar high conductivity composites) is for a temperature too low to be used to verify this algorithm. In that work, a normalized thermal conductivity of ~10 % for a 150°C irradiation to 0.25 dpa.

CONCLUSIONS

A semi-empirical algorithm for the degradation in thermal conductivity in the high thermal conductivity MKC-1PH material has been developed over the temperature range of 300-1200 °C and 0 to 1 dpa. Such an algorithm should prove useful to designers of plasma facing components for fusion reactors.

REFERENCES

[1] B. T. Kelly, Physics of Graphite . Applied Science Publishers. 1981.

[2] A. De Combarieu, *J. Phys.* (France), [28] 931(1968).

[3] R. Taylor, *Phil. Mag.*, [13] 157(1966) .

[4] R. Taylor, B. T. Kelly and K. E. Gilchrist, *J. Phys. Chem. Solids* [30] 2251(1969).

[5] B. T. Kelly, Chemistry and Physics of Carbon. [5] 119(1969) , ed. P. L. Walker. Marcel Dekker, Inc. New York.

[6] T, Maruyama and M. Harayama, J. Nucl. Mat. [195] 44(1992)

[7] C. H. Wu, J. P. Bonal and B. Thiele. J. Nucl. Mat. [212-215] 1168 (1994)

[8] L. Binkele. High Temp. High Press. [4] 401 (1972)

[9] B. A. Thiele, et al. Proc. 16th Int. Symp. on Effects of Radiation on Materials. ASTM (1992).

[10] R. Taylor, B. T. Kelly and K. E. Gilchrist. J. Phys. Chem. Solids. [30] 2251 (1969.

[11] L. L. Snead and T. D Burchell. J. Nucl. Mat. accepted (1995).

Figure 1
Normalized Reduction in Conductivity : R(D)

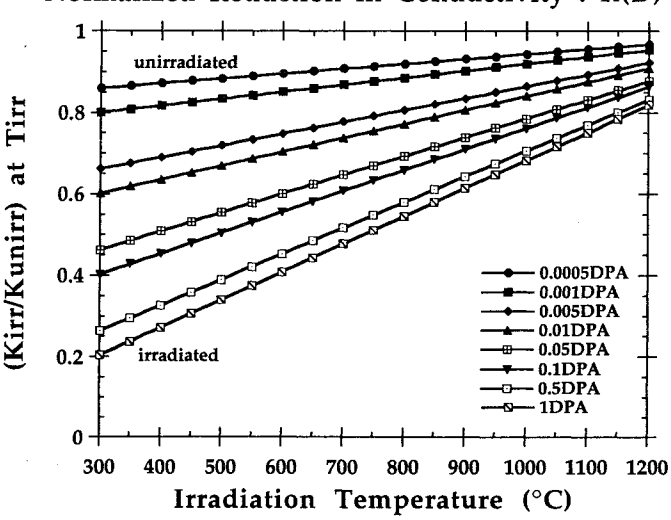
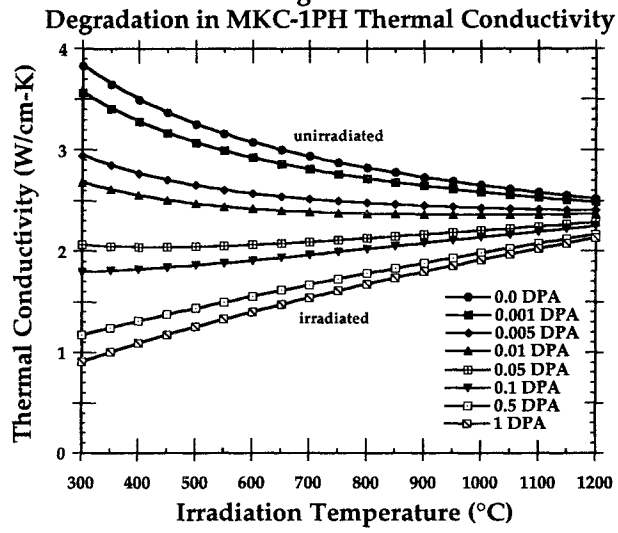
Figure 2
Degradation in MKC-1PH Thermal Conductivity