Letters to the Editors

Radiation Effects on Pyrolytic Graphite*

Pyrolytic graphite, either as free-standing pieces or as a coating, appears to be a promising material for high-temperature nuclear reactor applications. For its proper utilization, however, data on radiation effects are necessary.

A standard commercial grade of pyrolytic graphite**, which was deposited at 2200 C and at a pressure of a few mm Hg leading to an initial density of 2.20 g/cm³, was irradiated while heated to approximately 650 C for two successive exposures. Irradiation exposures in units of neutrons/cm², $E > 0.18$ MeV, were determined from nickel activation and the calculated neutron spectrum in the irradiation facility.

Length changes on four samples as a function of exposure are plotted in Figure 1. Large variations are usually observed during the initial irradiation, and therefore the differential changes during the second irradiation are considered more significant than total deformations. Rates during the second irradiation were -0.085 ± 0.004 per cent in the a direction and 0.22 ± 0.04 per cent in the c direction per 10^{20} neutrons/cm².

The coefficient of thermal expansion (CTE) in the a direction was changed slightly in individual samples. However, there was no significant change in the average CTE. In the c direction there was a steady increase in CTE with exposure, as shown in Table I. Apparently changes in both dimensions and CTE should be anticipated in pyrolytic graphite during service in high-temperature reactor environments.

Determinations of the lattice parameters at an exposure of 3.2×10^{20} nvt showed an increase of 0.5 ± 0.2 per cent in the c spacing from the original value of 6.845 Å and an increase of 0.11 ± 0.05 per cent in the a spacing from an initial value of 2.452 Å.
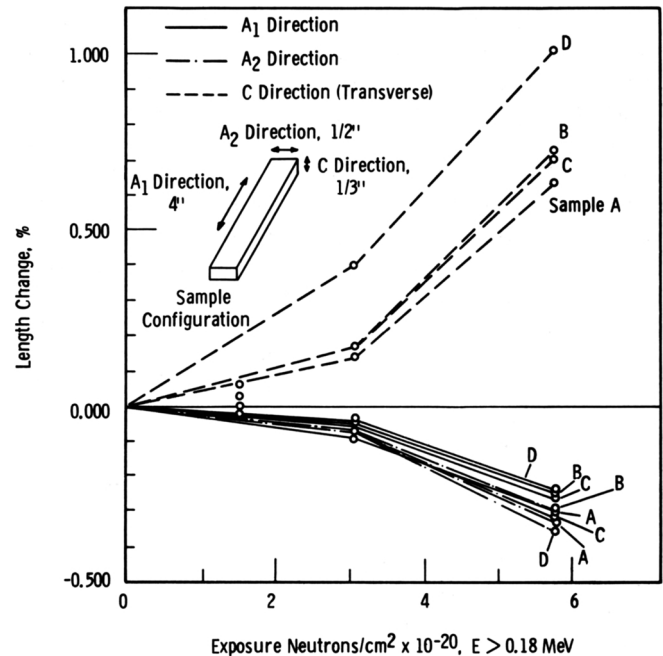
Fig. 1. Radiation-induced length changes in pyrolytic graphite. Irradiation temperature was approximately 650 C.

The irradiation-induced dimensional changes make it appear that the effect of irradiation is exactly the reverse of the effect of heat treatment on bulk dimensions[1]. Apparently further disorder is introduced into the originally turbostratic structure of the as-deposited material. Taking into

TABLE I

Radiation Effect on the Coefficients of Thermal Expansion[a]

Exposure, neutrons/cm² × 10^{-20}, $E \geq 0.18$ MeV		
0	3.2	6.0
α_c, per 10^6 C +20.5 ± 0.1	23.3 ± 0.1	24.6 ± 0.1
α_a, per 10^6 C +0.06 ± 0.04	0.05 ± 0.04	+0.07 ± 0.04

[a]Average CTE determined between 25 and 425 C.

[1]E. R. STOVER, "Effects of Annealing on The Structure of Pyrolytic Graphite," General Electric Report No. 60-RL-2564 M, (November 1960).

account observations on polycrystalline graphite, it appears that the effect of irradiation in pyrolytic graphite is the intercalation of carbon complexes, probably larger than 3 atoms, between the layer planes.

These data also aid in understanding high-temperature radiation effects in polycrystalline graphite[2]. The dimensional changes observed in pyrolytic graphite could be predicted qualitatively on the basis of the analysis of high-temperature neutron damage to polycrystalline graphite[3], which predicts an expansion of the crystallites in the c direction and a contraction in the a direction. Because pyrolytic carbon is very nearly theoretical density and possesses a high degree of anisotropy (properties also common to crystallites) it was expected that radiation effects in pyrolytic carbon would exhibit the behavior inferred for the crystallites in polycrystalline graphite. These results for pyrolytic material thus imply that high-temperature radiation-induced contraction in polycrystalline reactor graphites must occur either by decreasing the porosity or by densifying any carbon that was initially less than theoretical density, but not by a volume contraction in the graphite cyrstallites.

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[2] R. E. NIGHTINGALE, Ed., *Nuclear Graphite*, Ch. 9, Academic Press, N. Y. (1962).

[3] D. R. de HALAS and H. H. YOSHIKAWA, *Proc. Fifth Carbon Conf.*, Pgs. 249-255, Pergamon Press, New York (1962).

Isotopic Hafnium Resonance-Absorption Data

The extensive use of hafnium as a control material in nuclear reactors has made necessary an accurate knowledge of its nuclear absorption properties. Although knowledge of the hafnium differential absorption cross section is fairly well detailed, available resonance data for the even-even isotopes is limited, and significant discrepancies still exist between calculated and experimental resonance integrals for natural hafnium. In this regard, I. Itkin[1] has pointed out that the upper and lower bounds on the calculated resonance integral are 1720 and 2150 barns while reported

[1] WAPD-TM-324, "Analysis of the Neutron Capture Cross Section and Resonance Integral of Hafnium," I. ITKIN, (August, 1962).

experimental values range from 1300 to 2800 barns.

As part of the analytical and experimental investigations of the use of hafnium as a burnable poison in intermediate spectrum reactors[2], a sample of natural hafnium mixed with fuel was prepared for irradiation in such a system. The text specimen was a fuel element with 2 wt-% natural hafnium added to the fuel. After irradiation, radio-chemical and mass spectrometric analyses were performed at KAPL by F. M. Rourke and F. W. Walker to yield hafnium isotopic ratios and fuel-depletion data for several sections of the specimen.

This letter summarizes the results of the study of this isotopic-depletion data made to assess the present knowledge of the isotopic hafnium absorption cross sections in the resonance energy region. Isotopic-depletion data obtained in an intermediate-spectrum reactor emphasizes this energy region of interest.

For the system[a] of $Hf^{176,177,178,179,180}$ and U^{235}, the experiment provides a value for the fuel depletion and for four ratios of the five isotopic number densities for each section of the specimen. Two interrelated procedures for comparison of the experimental data with analytical predictions are employed. The first procedure uses the solution to the system of sequentially coupled linear differential equations describing the time dependence of the isotopic hafnium number densities to calculate the isotopic ratios on the basis of the BNL-325, 2nd Ed. cross-section data and knowledge of the intermediate spectrum as obtained from experiment and analysis. Comparison of the calculated and measured isotopic ratios then provides a measure of the adequacy of the cross-section data. The second procedure consists of inverting these solutions, which describe the time dependence of the hafnium system, to obtain the ratios (α_i) of the effective absorption cross section of the ith hafnium isotope to the effective absorption cross section of fuel[b] in terms of the experimental isotopic-

[a] Examination of the isotopic depletion of natural hafnium shows that the contribution to the number density of Hf^{176} by neutron captures in Hf^{174} is negligible for the values of fuel depletion of interest.

$$[b] \quad \alpha_i = \frac{\int_R dr \int_0^\infty dE \ \sigma_i(E) \ \phi(r,E)}{\int_R dr \int_0^\infty dE \ \sigma_u(E) \ \phi(r,E)}$$

where the spatial integration is taken over the specimen, and σ_i and σ_u are respectively the microscopic absorption cross sections for the ith isotope of hafnium and for fuel.

[2] The Physics of Intermediate Spectrum Reactors, UC-81, (September 1958).