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DIMENSIONAL AND PROPERTY CHANGES OF IMPREGNATED AND ISOTROPIC GRAPHITES IRRADIATED AT 300°-1450°C*

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ABSTRACT

The effect of fast-neutron radiation on the dimensions, thermal expansion coefficients and apparent crystallite heights of a number of anisotropic and isotropic nuclear graphites was studied. The anisotropic graphites were prepared with needle-coke or Texas coke fillers and were impregnated with coal-tar pitch or a non-graphitic carbon. Two isotropic graphites which were prepared with Gilsonite coke and impregnated with coal-tar pitch and an isotropic graphite which was prepared with uncalcined petroleum coke were also studied. The bulk densities of these materials ranged from 1.72 to 1.89 g/cm^3, thermal expansion coefficients from 1.5 to 6.1 x 10^{-6}, $^oC^{-1}$, apparent crystallite heights, L_c, from 340 to 900Å, electrical resistivity from 5.2 to 13 x 10^{-4} ohm-cm and crystallite orientation ratios as measured by an x-ray method from .64 to .98.

The graphites were irradiated in the General Electric Test Reactor or the Engineering Test Reactor in instrumented capsules in the range 300° to 1450°C and 1.20 to 3.25 x 10^{21}n/cm^2 (E > 0.18 Mev). The temperatures were measured with W/W-Re or chromel/alumel thermocouples and fast-neutron exposures were measured by the activation of Ni and Fe dosimeter wires.

The presence of an impregnant carbon tends to increase the thermal expansion coefficients and closed pore volume and to reduce the apparent crystallite height of the anisotropic graphites. The isotropic graphites were characterized as less crystalline than the anisotropic materials.

When the irradiation was carried out in the range 880° to 1250°C and 2 to 3 x 10^{21}n/cm^2 (E > 0.18 Mev) saturation of the linear contraction was observed in samples of several anisotropic graphites in which the majority of the layer planes were positioned normal to the measured length of the sample. In some cases expansion had apparently begun after the initial contraction. The isotropic graphites and a needle-coke graphite which had been impregnated with a non-graphitizing coke did not show this behavior, but continued to contract in both directions when irradiated at the same temperatures and exposures. One of the isotropic graphites which was prepared with Gilsonite coke and pitch showed a pronounced anisotropic behavior in its linear contraction.

The apparent crystallite height, L_c, saturated at a common value of 150-250Å for all graphites studied without regard to their unirradiated values or to the irradiation temperature. An exception to this was the samples irradiated at 615°C which were found to have L_c values in the range 300-400Å.

The changes in the bulk thermal expansion coefficients were small or non-existent except for a slight increase in the graphites which were made with Texas coke or for samples which were irradiated at 300° or 400°C. The graphites studied here all exhibited a common linear relationship between their unirradiated thermal expansion coefficients and their bulk linear

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dimensional changes when irradiation was carried out in the range 320° to 615°C. When irradiation was done in the range 840° to 1490°C, a separate linear relationship was observed for anisotropic or isotropic graphites. In this temperature range, the relationship was shifted toward higher contraction for the isotropic graphites. The nature of these relationships is discussed in terms of the densification of the crystallites as a function of their size and of the pore structure of the various materials.

The presence of a non-graphitizing impregnant carbon in the pores of an anisotropic needle-coke graphite produced an increase in the volume contraction and substantially reduced the anisotropy of its linear contraction when irradiated in the range 880° to 1015°C and to 3.25×10^{21} n/cm^2 ($E > 0.18$ Mev). This effect was not observed at lower doses of about $2 \times 10^{21} n/cm^2$ ($E > 0.18$ Mev). These results are ascribed to an enhanced densification of the small, imperfect crystallites of the impregnant carbon within the pores of the base material.