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EFFECT OF TEMPERATURE HISTORY
ON THE DIMENSIONAL CHANGES OF
NUCLEAR GRAPHITES

by
G. B. Engle

MASTER

Prepared under
Contract AT(04-3)-167
Project Agreement No. 12 and 17
for the
San Francisco Operations Office
U.S. Atomic Energy Commission

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ABSTRACT

When anisotropic needle-coke graphites and near-isotropic graphites were irradiated initially to $\sim 3 \times 10^{21}$ n/cm² at $\sim 1000^\circ\text{C}$ and then to higher fluences at 760° to 800°C , the rate of dimensional change did not adjust to the slower rate expected for the lower temperature. When the initial irradiation was done at 1450°C to $\sim 3 \times 10^{21}$ n/cm² and was followed by irradiation at 1225° to 1250°C to 9×10^{21} n/cm², the dimensional change rate was higher than that for specimens irradiated to 9×10^{21} n/cm² entirely at 1225° to 1250°C .

INTRODUCTION

High-temperature gas-cooled reactors utilize large graphite blocks. There are regions in the core where the graphite temperatures are initially high ($\sim 1000^{\circ}\text{C}$) and subsequently during the lifetime of the graphite drop to lower temperatures ($\sim 700^{\circ}$ to 800°C). Therefore, the effect of the temperature history on the dimensional changes of the graphite is of interest.

In the present work a number of graphites were irradiated to intermediate fluences at $\sim 1000^{\circ}$ and 1450°C , and the irradiation was then continued to higher fluences at lower temperatures. Where data exist, the dimensional changes are compared with those of materials of the same type that were irradiated to the higher fluences at the original temperature.

MATERIALS AND EXPERIMENTAL METHODS

The graphites selected for this study have been described elsewhere (Ref. 1). Graphites CHN and 711-TS-N, which are anisotropic graphites, were manufactured with needle coke and were heavily impregnated during manufacture to increase their density and strength. Grade H-315-A was prepared with Gilsonite coke, and H-319 was manufactured with raw petroleum coke. Both of these are near-isotropic graphites.

The measuring techniques and irradiation experiments have been described previously (Refs. 1,2).

EXPERIMENTAL RESULTS

Dimensional changes are shown in Figs. 1 and 2. The curves for 950° to 1015°C were taken from Ref. 2. All specimens irradiated at 950° to 1015°C to 3.2×10^{21} n/cm² were cut from the same parent blocks and were irradiated in the same crucible within the capsule. When irradiated to higher fluences, the specimens at 760° to 800°C were contained in the same capsules as those at 950° to 1015°C, but were in low-temperature and low-flux positions.

The data points for specimens irradiated first to 3.2×10^{21} n/cm² at ~1000°C and then to 5.0×10^{21} n/cm² at 760° to 800°C reside in the same data population as those for specimens irradiated to 5.0×10^{21} n/cm² at 950° to 1015°C only. The rate of dimensional change of the specimens that were reduced in temperature remained about the same as that of the specimens irradiated only at 950° to 1015°C. The perpendicular specimens that were irradiated to 3.2×10^{21} n/cm² at 1015°C were just entering turnaround when the temperature was dropped to 760° to 800°C. Therefore, it was expected that if the irradiation were continued at ~1000°C, their rate of dimensional change would be near zero for the next 2 to 3×10^{21} n/cm². The shift to 760° to 800°C did not alter this behavior.

Data for CSF graphite (taken from Ref. 3) irradiated to $>6 \times 10^{21}$ n/cm² at 775° to 825°C and 1000° to 1075°C are compared with the 711-TS-N data in Fig. 1b. These materials are similar in structure and dimensional changes. If the CSF curve for 1000° to 1075°C were interrupted at about 3.2×10^{21} n/cm² and joined to the 775° to 825°C curve at the same fluence, the slope of the curve would change from about zero to a negative value. However, if it were joined to the 775° to 825°C curve at a comparable

Fig. 1. Dimensional changes as a function of fluence at 1000°C followed by 760° to 800°C. (a) CHN

Fig. 1. Dimensional changes as a function of fluence at 1000°C followed by 760° to 800°C. (b) 711-TS-N

Fig. 1. Dimensional changes as a function of fluence at 1000°C followed by 760° to 800°C. (c) H-315-A

Fig. 1. Dimensional changes as a function of fluence at 1000°C followed by 760° to 800°C. (d) H-319.

Fig. 2. Dimensional changes of CHN and H-315-A as a function of fluence at 1450°C followed by 1225° to 1250°C

strain ($\sim 0.9\%$), then the continuous curve for 1000° to 1075°C stepped down to 775° to 825°C would be similar to the curve for 711-TS-N under the same temperature history, except the expansion rate would be somewhat slower. Cox and Helm (Ref. 3) indicate that when the irradiation temperature was changed, the contraction rate changed to correspond to that of samples irradiated exclusively at the new temperature. However, they did not specify the temperatures in question and presented no data to substantiate their conclusion. Their observation may be correct while the specimens are in the contraction mode where maximum densification has not yet been reached, but it does not coincide with the perpendicular data presented here.

The data in Fig. 1a for CHN graphite show the same trend in the perpendicular data as that shown by the 711-TS-N data. The parallel data for CHN show a somewhat slower rate of contraction after the temperature was dropped to 760° to 800°C . However, the CHN specimen used for the temperature-drop experiment was not typical of the specimens used to construct the 950° to 1015°C curve, as it had accumulated about 50% more contraction strain at $3.2 \times 10^{21} \text{ n/cm}^2$. This particular specimen may have been ready to enter parallel turnaround, which starts at a contraction strain of about 3 to 4% (Ref. 2). Thus, the lower bulk contraction rate after dropping to 760° to 800°C may indicate that this particular specimen is entering turnaround. This can be seen more clearly by comparing volume changes in Fig. 3 where the data for the specimens that were dropped in temperature show turnaround to expansion.

More data are needed to clarify the behavior of the specimens under the conditions of temperature reduction from $\sim 1000^\circ$ to $\sim 800^\circ\text{C}$.

The data in Fig. 2 were obtained first at 1450°C and $3.2 \times 10^{21} \text{ n/cm}^2$ and then for further irradiation to $9 \times 10^{21} \text{ n/cm}^2$ at 1225° to 1250°C . The perpendicular data for CHN clearly show that the CHN expanded at a higher rate after the initial irradiation at 1450°C than the specimens irradiated only at 1175° to 1280°C . The total strains were again about

Fig. 3. Volumetric changes of CHN as a function of fluence at 1015°C followed by 760° to 800°C

the same in both cases. The situation appears to be the same for H-315-A, except that the total strain after 1450°C was higher than that for the H-315-A irradiated at 1175° to 1280°C. In both cases it again appears that the initial irradiation at the higher temperature initiates a more rapid turnaround and a higher expansion rate after turnaround.

CONCLUSIONS

When anisotropic needle-coke graphites and near-isotropic graphites were irradiated initially to $\sim 3 \times 10^{21}$ n/cm² at $\sim 1000^\circ\text{C}$ and then to higher fluences at 760° to 800°C , the rate of dimensional change did not adjust to the slower rate expected for the lower temperature. When the initial irradiation was done at 1450°C to $\sim 3 \times 10^{21}$ n/cm² and was followed by irradiation at 1225° to 1250°C to 9×10^{21} n/cm², the dimensional change rate was higher than that for specimens irradiated to 9×10^{21} n/cm² entirely at 1225° to 1250°C .

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