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EFFECT OF TEMPERATURE HISTORY ON THE DIMENSIONAL CHANGES OF IRRADIATED NUCLEAR GRAPHITES

by

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ABSTRACT

When artificial graphites were irradiated initially to fluences of about 3×10^{21} n/cm² at T_1 of 1200° or 1000°C and then to higher fluences at T_2 of about 1000° or 800°C, respectively, the dimensional changes did not deviate significantly from the isothermal curve for T_1 , the higher temperature.

INTRODUCTION

High-temperature gas-cooled reactors utilize large graphite blocks as moderators. There are regions in the core where the graphite temperatures are initially high at T_1 but drop during the graphite block core life to lower temperatures T_2 . Therefore, the effect of temperature history on the dimensional changes of the graphite blocks is of interest.

The experimental data presented here were obtained from irradiation capsules designed to obtain isothermal data. However, it was possible to shift a number of samples from high-temperature cells to low-temperature cells in a stepwise fashion allowing irradiation at T_1 for the initial increment of fluence and at T_2 to higher fluences. Isothermal data were obtained concurrently at T_1 for comparative purposes. For some graphites isothermal data were also accumulated at T_2 , but only to low fluences.

There was insufficient space in the capsules to provide a complete set of data; therefore, it should be recognized that the experimental data reported herein are preliminary.

MATERIALS AND EXPERIMENTAL METHODS

The graphites selected for this study have been described elsewhere (Refs. 1-4). Graphites CHN, 711-TS-N, and H-327, which are anisotropic graphites, were manufactured with needle coke and were impregnated during manufacture to increase their density and strength. Grade H-315-A was prepared with Gilsocoke, and H-319 was manufactured with raw petroleum coke. The latter two are near-isotropic graphites. H-419 is a high-density high-strength isotropic graphite.

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

All specimens of each graphite, irradiated at T_1 to the initial fluence, were cut from the same parent blocks and were irradiated together in the same crucible within the capsule. During irradiation to higher fluences at T_2 , the specimens were all contained in the same capsules, those continuing at T_1 in the high-temperature cells and those at T_2 in the low-temperature cells.

EXPERIMENTAL RESULTS

Dimensional changes of the needle-coke and near-isotropic graphites are shown in Figs. 1 and 2, respectively.* The data points in Figs. 1 and 2 at T_1 and to the initial fluence F_1 are plotted and identified. Subsequent dimensional changes at higher fluences obtained isothermally at T_1 are represented by solid curves or bands, taken from Refs. 2 and 5. If a reference number is not shown, the solid lines represent the data points for that particular plot. It should be noted that the data points for T_1 at F_1 are part of the total isothermal data population that make up the solid curves or bands. The data in Figs. 1a through 1e and Figs. 2a and 2b are the same as those reported in Refs. 3 and 4, but with additional points that extend the data to higher fluences. The data in Figs. 1f through 1h and Fig. 2c are new.

The dimensional changes of the specimens irradiated initially at T_1 to F_1 and then at T_2 to F_2 are within the data population from which the isothermal curves were derived.

It is desirable to compare the rate of dimensional change of the specimens at T_2 after the initial irradiation at T_1 with isothermal data at T_2 . However, isothermal data on the graphites at T_2 (800° or 1000°C) were only obtained at low or intermediate fluences in these experiments

*Final analyses of the temperature and fluence have not been completed for data points above 7×10^{21} n/cm². Therefore, that portion of the curves should be considered preliminary. Final analyses of these data are not expected to change the results significantly.

because the low-temperature cells were also the low-fluence cells. Therefore, for the purpose of comparison, isothermal data for an anisotropic graphite designated CSF, which is similar to graphite 711-TS in orientation and thermal expansivity and in dimensional changes at 1000°C, were taken from Refs. 6 and 7. The dimensional changes of the CSF and 711-TS are almost identical in their perpendicular directions at T_1 or about 1000°C at fluences up to 8×10^{21} n/cm², and it is expected that their behavior will be similar at T_2 or about 800°C. Thus, the comparisons in Fig. 3a with respect to graphite structure appear reasonable and we can to a first approximation treat the data as if from the same material and only deal with temperature and fluence effects and ignore structural effects. It is difficult to compare the rate of change in dimensions under isothermal and nonisothermal conditions in the fluence range where turnaround is occurring because of the flatness of the curves. Therefore, a comparison was made in the perpendicular direction after turnaround and in the parallel direction before turnaround (see Fig. 3).

It should be noted that the initial rate of contraction in both graphites in Fig. 3 is highest at T_1 or 1000°C. In the perpendicular direction, the contraction in the 711-TS specimen had just reached a maximum at 3 to 4×10^{21} n/cm² (see Ref. 2) when the irradiation at 1000°C was interrupted and the temperature for subsequent fluences was dropped to 800°C. Thus, the needle-coke graphite was in a condition to begin expansion if continued to higher fluences at T_1 (~1000°C) or perhaps at any T_2 below 1000°C. The CSF specimens irradiated at T_2 (~800°C) did not reach maximum contraction until about 9.5×10^{21} n/cm² $\pm$ $\sim 2 \times 10^{21}$ n/cm². Thus, comparison of the rate of expansion at T_2 (~800°C) of the graphites from the point of maximum contraction after irradiation at T_1 may be made by transposing the T_2 curve AB to A'B'. Three observations are immediately obvious: (1) the initial rate of expansion of curve A'B' is low compared with curve A'C (rate at T_2 after initial irradiation at T_1), (2) the crossover to net expansion of curve A'C occurs at a lower fluence than does A'B', and (3) the expansion rate of curve A'B' appears to be rapidly approaching that of A'C after a net expansion of about 1%.

A similar comparison can be made with CSF and H-327 graphites in their parallel directions (see Fig. 3b). In this situation, the graphite specimens are still contracting and turnaround has not been reached. Curve A'B' in this case shows essentially the same slope as AC. The initial contraction rate at T_1 is greater than at T_2 and this difference accounts for the difference in the magnitude of the contraction at point C vs B'. It may be expected that the curves in Fig. 3b will show the same characteristics as those in Fig. 3a after turnaround occurs and expansion begins.

DISCUSSION

The dimensional changes of graphites irradiated first at T_1 of $\sim 1200^\circ$ or 1000°C followed by further irradiation at T_2 of $\sim 1000^\circ$ or 800°C , respectively, have been shown, by the data presented here, to follow the same curve for irradiation at T_1 only. A similar effect has been reported for pyrolytic carbons (Refs. 8,9). The experiments on pyrolytic carbons were not quite the same as those described here for graphites, but were similar enough to warrant comparisons. Dimensional change data were reported for pyrolytic specimens irradiated at 950°C after prior irradiation at 1650°C (Ref. 8) and at 630°C after prior irradiation at 950°C (Ref. 9). A higher expansion rate by about a factor of 2 was observed in perpendicular specimens irradiated at 950°C after prior irradiation at 1650°C than in specimens irradiated at only 950°C over the same fluence range (Ref. 8). It should be noted that the specimen irradiated at 950°C had not accumulated the same total strain as the $1650^\circ \rightarrow 950^\circ\text{C}$ specimen and, therefore, the rate comparison was not made at equivalent strains. The dimensional changes during irradiation at 630°C of specimens having prior irradiation at 950°C were substantially greater than the changes caused by continued irradiation at a constant temperature of 630°C to the same total fluence (Ref. 9). However, the total volume change at a constant temperature of 950°C was greater than that after 950° and 630°C .

Another experiment was reported in Ref. 9 where the temperature of the capsule was reported to have been continuously programmed down from 1000° to 750°C rather than coming to the higher fluence at T_2 (750°C) by a step irradiation (T_1 to F_1 followed by T_2 to F_2). Volumetric changes of specimens irradiated at 1000° to 750°C superimpose on isothermal data obtained at 900° to 1000°C or 1000°C when taken to the same total fluence.

One additional observation was reported in Ref. 9. In this experiment, two series of pyrolytic carbons were first irradiated at 510°C to 3.8×10^{21} n/cm² and at 1000°C to 5.6×10^{21} n/cm², respectively. The two series of specimens were then irradiated again starting at 1050°C and programmed down to 630°C for an additional fluence increment of 6.4×10^{21} n/cm² to total fluences of 10.2 and 12.0 $\times 10^{21}$ n/cm², respectively. The irradiation-induced changes which occurred at 510°C were small and were ignored, whereas the subsequent dimensional changes of the specimens irradiated at 1050°C and programmed down to 630°C were the same as the isothermal curve at ~1000°C.

Additional data are needed where irradiation temperatures are programmed down from T_1 to T_2 at a number of fluences prior to maximum contraction, with subsequent irradiation through turnaround to net expansion. Isothermal data at T_1 and T_2 are also required. Such experiments would provide design data for regions of the reactor where temperature is continually dropping as a function of fluence.

CONCLUSIONS

When artificial graphites were irradiated initially to fluences of about 3×10^{21} n/cm² at T_1 of about 1000° or 1200°C and then to higher fluences at T_2 of about 800° or 1000°C, respectively, the dimensional changes did not deviate significantly from the isothermal curve for T_1 , the higher temperature.

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Fig. 1. Dimensional changes of needle-coke graphites irradiated initially at T_1 and subsequently at reduced temperature. (a) CHN

Fig. 1. Dimensional changes of needle-coke graphites irradiated initially at T_1 and subsequently at reduced temperature T_2 . (b) CHN

Fig. 1. Dimensional changes of needle-coke graphites irradiated initially at T_1 and subsequently at reduced temperature T_2 . (c) 711-TS

Fig. 1. Dimensional changes of needle-coke graphites irradiated initially at T_1 and subsequently at reduced temperature T_2 . (d) H-327 (pre-production)

Fig. 1. Dimensional changes of needle-coke graphites irradiated initially at T_1 and subsequently at reduced temperature T_2 . (e) H-327 (pre-production)

Fig. 1. Dimensional changes of needle-coke graphites irradiated initially at T_1 and subsequently at reduced temperature T_2 . (f) H-327 (Fort St. Vrain production from center of log)

Fig. 1. Dimensional changes of needle-coke graphites irradiated initially at T_1 and subsequently at reduced temperature T_2 . (g) H-327 (Fort St. Vrain production from edge of log)

Fig. 1. Dimensional changes of needle-coke graphites irradiated initially at T_1 and subsequently at reduced temperature T_2 . (h) H-327 (Fort St. Vrain production from edge and center of log)

Fig. 2. Dimensional changes of near-isotropic graphites irradiated initially at T_1 and subsequently at reduced temperature T_2 . (a) H-315-A (Gilsocoke graphite)

Fig. 2. Dimensional changes of near-isotropic graphites irradiated initially at T_1 and subsequently at reduced temperature T_2 . (b) H-319 (raw-coke graphite)

Fig. 2. Dimensional changes of near-isotropic graphites irradiated initially at T_1 and subsequently at reduced temperature T_2 . (c) H-419

Fig. 3. Dimensional changes of needle-coke graphites irradiated at T_1 to F_1 followed by irradiation at T_2 to F_2 vs CSF graphite irradiated at T_2 to $15 \times 10^{21} \text{ n/cm}^2$. (a) 711-TS (1)

Fig. 3. Dimensional changes of needle-coke graphites irradiated at T_1 to F_1 followed by irradiation at T_2 to F_2 vs CSF graphite irradiated at T_2 to $15 \times 10^{21} \text{ n/cm}^2$. (b) H-327 (||)

REFERENCES

1. Engle, G. B., and K. Koyama, "Dimensional and Property Changes of Graphites Irradiated at High Temperatures," Carbon 6, 455 (1968).
2. Engle, G. B., "Irradiation Behavior of Nuclear Graphites at Elevated Temperatures," Carbon 9, 539 (1971).
3. Engle, G. B., "Effect of Temperature History on the Dimensional Changes of Nuclear Graphites," USAEC Report GA-10001, Gulf General Atomic, March 23, 1970.
4. Engle, G. B., J. C. Bokros, and R. J. Price, "Research on Graphite, Annual Report May 15, 1970, through June 30, 1971." USAEC Report Gulf-GA-A10694, Gulf General Atomic, November 15, 1971.
5. Engle, G. B. and A. L. Pitner, "High-Temperature Irradiation Behavior of Production-Grade Nuclear Graphites," USAEC Report GA-9973, Gulf General Atomic, July 10, 1970.
6. Cox, J. H., and J. W. Helm, "Graphite Irradiations 300°-1200°C, Carbon 7, 319 (1969).
7. Helm, J. W., "Irradiation of Graphite at Temperatures of 300 to 1200°C," USAEC Report BNWL-1056B, Battelle Northwest Laboratories, August 1969.
8. Bokros, J. C., G. L. Guthrie, R. W. Dunlap, and A. S. Schwartz, "Radiation-Induced Dimensional Changes and Creep in Carbonaceous Materials," USAEC Report GA-8700, Gulf General Atomic, August 1968.
9. Bokros, J. C., and D. W. Stevens, "Irradiation Behavior of Isotropic Carbons," Carbon 9, 19 (1971).