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EFFECT OF TEMPERATURE ON RADIATION-INDUCED CONTRACTION OF REACTOR GRAPHITE*

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Radiation-induced contraction as a function of temperature was studied in needle-like and conventional coke graphites from 450 to 1200° C. Fast neutron exposures ranged up to 3.2×10^{21} nvt, $E > 0.18$ Mev, corresponding to approximately 20,000 MWD/AT in a Hanford graphite-moderated reactor. The results significantly extend information on the effect of high temperature irradiation on needle-like coke graphites manufactured with particle size, impregnation, and graphitization temperature as variables. In general, the transverse contraction rate goes through a broad minimum between the temperatures of 650 and 950 C with the least contraction occurring at approximately 800 C. Needle coke and CSF graphites show the same effect but the contraction of needle coke graphites is approximately 0.6 that of CSF at all temperatures. In the parallel direction the rates of contraction are the same for both CSF and needle-coke graphites.

I. INTRODUCTION

The limited state of knowledge of radiation-induced dimensional changes in graphite was described in a paper presented at the Fourth Carbon Conference.⁽¹⁾ Since that earlier paper, the contraction of needle-coke graphite and CSF graphite has been studied more extensively and it is now possible to describe the effects of temperature on contraction over the range 450 to 1200° C and up to exposures of 3.2×10^{21} nvt ($E > 0.18$ Mev).

Even though the contraction observed at high temperatures is small, it has important applications to reactor design. In the case of graphite used as a structural element in a reactor, dimensional instability can gradually affect the mechanical configuration and cause operational problems over the lifetime of the reactor. These problems, although serious, are not as extreme as the problems arising from expansion caused by radiation damage at low temperatures. The expansions are much greater in both rate and magnitude.

* Work performed for the Atomic Energy Commission under Contract AT(45-1)-1350.

(1) DAVIDSON, J.M., WOODRUFF, E.M., and H.H. YOSHIKAWA, "High Temperature Radiation Contraction in Graphite," Proc. Fourth Conf. on Carbon, 599-605, (1960).

At low temperatures ($< 300^{\circ} \text{C}$) the expansion rate is dependent on exposure, temperature of exposure and type of graphite. Figure 1 shows the effect of neutron exposure at 30°C on several grades of reactor graphite. The graphite expands in the direction transverse to the extrusion axis and contracts in the direction parallel to the extrusion axis. At high temperatures it contracts in both directions.

For the purpose of this paper, exposures are given in either MWD/AT of fuel or in nvt of neutrons with energies in excess of 0.18 Mev. The conversion between MWD/AT and nvt is not a constant but depends on the spectra of the reactor positions considered. As an example, for the Experimental Gas-Cooled Reactor (EGCR), $1 \text{ MWD/AT} = 1.3 \times 10^{17} \text{ nvt}$, $E > 0.18 \text{ Mev}$, for exposures generated in core positions of high-flux, light water-moderated, testing reactors, such as the E-7 position of the General Electric Test Reactor. Since the conversion constant varies depending on the reactors being compared, it should be applied carefully.

The graphites referred to are:

- KC - Kendall coke graphite, graphitized at 2800°C , density 1.7 g/cc .
- TSCBF - Texas coke graphite, graphitized and F-purified in one step at 2450°C , density 1.65 g/cc .
- CSF - Cleves coke graphite, reactor grade, F-purified at 2700°C , density 1.66 g/cc .
- VC - Continental needle coke graphite. Approximately 40% of particle mix through 200 mesh screen, all through 20 mesh screen, one impregnation. Refinery process altered to produce needle-like coke, graphitized at 2800°C , density 1.71 g/cc .
- NC-7 - Similar to VC but smaller percent of particle mix through 200 mesh screen, extrusion size $16\text{-}3/8 \times 16\text{-}3/8$, density 1.50 g/cc .

II. EXPERIMENTAL

Figure 2 shows the internal view of a typical irradiation capsule. The samples were made into quarter-round shapes, assembled four in a group, and then arranged longitudinally in the capsule. Each group contained one sample of CSF graphite to reference the test results to prior irradiation experience. Graphite holders were designed to allow motion of the samples longitudinally due to thermal expansion but were themselves constrained by 0.005 inch thick molybdenum disks transverse to the capsule to prevent lateral motion. The disks were attached to aluminum cooling rings which thermally grounded the assembly to the outside aluminum jacket and also provided holders for flux monitoring foils.

The fast neutron exposures of the samples were determined both experimentally and theoretically. Nickel foils utilizing the high energy (n,p) reaction were used to verify theoretical calculations of the flux made using the PDQ code on an IBM 704. All experimentally derived fluxes were corrected for thermal neutron burnout of Co^{58} and Co^{58m} . The testing was performed at high specific flux intensities (average longitudinal flux was 3.2×10^{14} nv) such that the equivalent of 15,000-20,000 EGCR MWD/AT was accumulated in only a few months.

To measure temperatures of the samples, the sample groups were instrumented with Geminol N-P thermocouples insulated with vitreous aluminum oxide.

Changes in length were measured both physically and optically. Physical measurements were made end-to-end with a Bausch and Lomb DR-25 optical gage incorporating reproducible anvil pressure. Optical measurements were made of the distance between holes near the ends of each sample with an optical comparator. Standard deviations of measurement observed were 5×10^{-5} and 16×10^{-5} inch for physical and optical measurements, respectively.

III. HIGH TEMPERATURE CONTRACTION

Two experimental capsules were irradiated at temperatures of 450-850° C and 800-1200° C, respectively. Individual samples received different exposures for the term of the irradiation achieved different temperatures because of unavoidable longitudinal variations of gamma and neutron flux. Since the neutron flux and the gamma flux were proportional, the sample with the highest exposure achieved the highest irradiation temperature. Since the temperature ranges of the two capsules overlapped, the data from one could be joined to the data from the other at different neutron exposures. Upon normalization of contraction to a single exposure, 10^{21} nvt, the data from samples at the same temperature in the two capsules were in good agreement. This coincidence adequately justifies the normalization to a single exposure, without consideration of dose rate, as performed in the following figures.

In Figures 3 and 4, the results of the needle-coke graphite irradiations are presented for normalized exposures of 10^{21} nvt. The needle-coke graphite is relatively more stable than the CSF and contracts less in the transverse direction than in the parallel direction. In Figure 3, the effect of differences in sampling locations for parallel samples from the 16 x 16 inch extrusion is shown to be negligible. Samples were taken at the center, midway, and at the outside edge of the bar and are designated by different symbols. The data in Figure 4 include results from needle-coke graphites that were produced by different vendors to the same specification. The contraction rates were found to be substantially the same.

In Figure 5, data from the preceding two figures and covering the temperature range 450-1200° C are plotted with a logarithmic ordinate. The ratio of contraction of CSF to needle-coke graphite in the transverse direction is nearly constant which indicates that the ratio is insensitive to the temperature. The rate of contraction per 10^{21} nvt is greater in the parallel direction than in the transverse. Both CSF and needle-coke graphite show the same rate in the parallel direction. The rates in both directions decrease as the temperature increases until the region of minimum contraction rate for both graphites is reached at around 800° C.

IV. SUMMARY

The foregoing data show the characteristic temperature sensitive radiation damage to artificial graphite. While the irradiation always produced contraction in the direction parallel with extrusion, it produces either expansion or contraction in the direction transverse to extrusion, depending on the temperature of irradiation. Irradiation at temperatures less than 300° C results in large expansions in the transverse direction, and at higher temperatures, a contraction although smaller in magnitude. The generalized behavior of reactor graphite as a function of temperature is shown in Figure 6. In this figure the data for CSF graphite are shown as a function of temperature from 30-1200° C. The expansion rate in the transverse direction is very high at low temperatures; it lessens and becomes a contraction rate in the vicinity of 300° C. Thereafter the contraction rate lessens and goes through a minimum in the vicinity of 800° C. The contraction rate in the parallel direction is greatest at low temperatures; it lessens at high temperatures and approaches the rate for the transverse direction.

Certain limitations must be placed on the data presented. They are:

- (1) A distortion rate linear with exposure has been assumed when the data are normalized to a unit exposure of 10^{21} nvt, and then plotted as a function of temperature. Should the rate not be linear, erroneous conclusions on the effect of temperature could result. Limited data from other experiments indicate that, to the exposure reported in this paper, the rate is constant.
- (2) Dependence on neutron intensity has been studied at low temperatures, but work remains to be done at elevated temperatures. A strong dependence on intensity could modify conclusions made from these data.
- (3) Although the exposures for high temperature data were moderately high, no knowledge presently exists of the continuing rate of distortion at higher exposures.

FIGURE CAPTIONS

Figure 1 - Physical Distortion of Reactor Grade Graphite at 30° C

Figure 2 - Internal View - High Temperature Graphite Irradiation Capsule

Figure 3 - Effect of Temperature of Irradiation on Physical Distortion

Figure 4 - Effect of Temperature of Irradiation on Physical Distortion

Figure 5 - Physical Distortion of Reactor Grade Graphites at Elevated Temperatures

Figure 6 - Effect of Irradiation Temperature on Graphite Physical Distortion

FIGURE 1
Physical Distortion of Reactor Grade Graphite Irradiated at 30° C

FIGURE 2
Internal View - High Temperature Graphite Irradiation Capsule

FIGURE 3
Effect of Temperature of Irradiation on Physical Distortion

FIGURE 4
Effect of Temperature of Irradiation on Physical Distortion

FIGURE 5

Physical Distortion of Reactor Grade Graphites at Elevated Temperatures

FIGURE 6

Effect of Irradiation Temperature on Graphite Physical Distortion