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EFFECT OF LOW COMPRESSIVE STRESSES ON RADIATION-INDUCED
DIMENSIONAL CHANGES IN GRAPHITE

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EFFECT OF LOW COMPRESSIVE STRESSES ON RADIATION-INDUCED*
DIMENSIONAL CHANGES IN GRAPHITE

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ABSTRACT

When graphite is employed as a reactor moderator, the variations of temperature and neutron flux within the moderator bars cause internal stresses to be generated in the graphite. The effect of these stresses on the dimensional changes of the graphite are of immediate interest in the field of reactor design. Samples of five nuclear-grade graphites have been irradiated under constant compressive stresses of 150 and 300 psi. Length changes of the stressed samples are compared with length changes of unstressed control samples, irradiated in the same assembly. Results obtained at 625° C and for exposures up to 6500 Mwd/At in a Hanford irradiation facility are presented. Additional data on one nuclear-grade graphite, irradiated under 100 psi constant tensile stress, are included.

INTRODUCTION

The dimensional changes produced in graphite, as a result of neutron radiation, have been studied extensively. These dimensional changes are related in a complex manner to both the fast-neutron exposure and the irradiation temperature. Gradients in temperature and fast-neutron flux within the moderator bars results in differences in rates of dimensional change at different positions within the bars. These differences in dimensional change produce stresses which could lead to a self-rupturing of

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the moderator bars. It is therefore important to the design of modern graphite-moderated reactors to know how these stresses will affect the graphite bars. Specifically there are three questions of primary interest:

1. Will the stresses alter the rates of dimensional change?
2. If so, is the effect sufficient to limit the resultant stresses to a level less than the stress-rupture limit during a reactor lifetime?
3. If the stress-rupture limit is not exceeded, will the strain-rupture limit be exceeded?

The purpose of this investigation is to provide an answer to the first question of the effect of stress on the rate of dimensional change.

EQUIPMENT AND PROCEDURES

Five nuclear-grade graphites were selected for examination. Pertinent information on these graphites is shown in Table I. Other properties have been more fully described elsewhere.¹

TABLE I. GRAPHITE MATERIALS TESTED

<u>Graphite</u>	<u>Type</u>	<u>Coke</u>		<u>Graphitization temperature, °C</u>	<u>Use</u>
		<u>Particle size</u>			
TSX	Continental, Lake Charles No. 1	50% through 200 mesh	100% through 20 mesh	3000	NPR moderator
AGOT-LS	Same as TSX	Same as TSX		2800	NPR reflector
NC-8	Same as TSX	100% through 20 mesh		2800	EGCR moderator
CSF	Cleves	40% through 200 mesh	100% through 20 mesh	2800	N Reactor moderator
TSGBF	Texas Lockport	Same as CSF		2450	KE & KW Reactor moderator

All samples were cut from full-size production-run bars and machined to 0.262 ± 0.002 -in. diameter cylinders $1\frac{3}{4}$ in. long. Samples were machined with the long axis of the cylinder either parallel or transverse to the direction of extrusion of the bar. All 150-psi samples and the 300-psi TSX graphite samples were annealed at 1000° C for over 1 hr in helium or vacuum to remove any internal stresses that might have been produced by machining. All other samples were annealed at 1950° C for 15 min. in vacuum. X-ray diffraction measurements were made to obtain the crystal parameters. Length measurements were made on a Bausch and Lomb DR-25 Optical Gauge (Type 33-14-23) with which a precision of $\pm 1 \times 10^{-4}$ in. could be easily obtained on these samples.

The 150-psi samples were irradiated under constant compressive stress in graphite boats such as shown in Fig. 1. The lever arm, which is partially enclosed in the machined groove in the bottom of the graphite weight, pivots in a notch on the bottom of the compartment. The stressed sample rides freely in the central hole of the compartment divider and is supported by the end of the lever arm. The top of the sample bears against the sample spacer, which in turn is supported by the sliding lid. Two unstressed samples are placed in holes on either side of the stressed sample. Nickel and aluminum cobalt alloy flux-monitor foils, encased in quartz tubing, are placed in the smaller holes adjacent to the unstressed samples.

The 300-psi boats are of similar construction except that the lids and lever arms are strengthened, and the weights are milled out to contain high density, high purity, Al_2O_3 blocks. The stresses were adjusted by changing the pivot points to within $\pm 5\%$, $- 10\%$ of the stated stresses prior

to the first irradiation period. All irradiations were performed in test facilities of Hanford KE and KW Reactors at a temperature of 625° C. The maximum temperature variations were probably within + 50°, - 25° C. Additional samples were heated in an oven at 635° C to determine whether dimensional changes occurred. The unstressed "oven-samples" showed a small amount of growth; however, this is thought to be due to a small amount of oxidation occurring during the test. The length changes of the stressed "oven-samples" showed large fluctuations, but there does not appear to be any consistent change of length with time.

DATA

Fig. 2 to 9 show the length changes of the stressed and unstressed samples. The limits of one standard deviation, indicated by the vertical bars through the mean values, are of normal magnitude for standard size (0.426-in. diameter x 4 in.) graphite samples used in most other Hanford irradiations. The principle reason for the deviations is a small, but real difference between individual samples. Additionally, the variance is increased by a normal uncertainty of $\pm 10\%$ in actual exposure for the same nominal power generation. After the initial period of growth, the samples appear to contract at approximately a constant rate with respect to exposure. It is apparent from these graphs that the contraction of the stressed samples is greater than that of the unstressed samples.

Table II contains the contraction rates as obtained by least squares analysis of the data. In the analysis the rates were assumed to be a linear function of exposure after about 1200 Mwd/At and all stressed samples were treated as if from one sample population.

TABLE II. SAMPLE CONTRACTION RATES

Graphite	Orientation	Sample contraction rates*		P.L. (%) ⁺	Stressed-to- unstressed ratio
		Unstressed samples, %/Mwd/At x 10 ⁵	Stressed samples, %/Mwd/At x 10 ⁵		
TSX	Transverse	0.97 ± 0.13	1.58 ± 0.25	95	1.6
AGOT-LS	Transverse	0.49 ± 0.08	1.23 ± 0.21	99	2.5
NC-8	Transverse	0.89 ± 0.34	2.52 ± 0.43	99	2.8
	Parallel	0.90 ± 0.12	2.92 ± 1.31	95	3.2
CSF	Transverse	1.07 ± 0.13	2.11 ± 0.33	99	2.0
	Parallel	1.43 ± 0.18	1.90 ± 0.27	85	1.3
TSGBF	Transverse	0.98 ± 0.08	1.37 ± 0.25	90	1.4
	Parallel	1.40 ± 0.10	1.59 ± 0.11	75	1.1

* Mean value ± one standard deviation.

+ Probability level at which the stressed and unstressed rates are significantly different.

A point of interest is that the contraction rates of the unstressed samples are less than the contraction rates of standard-size graphite samples (0.426 in. diameter by 4 in. length) irradiated under similar conditions. This apparent sample-size effect has also been noted² when the contraction of standard-size samples were compared with the contraction of large (4 x 4 x 24 in.) graphite bars.

Two of the stressed and four of the unstressed samples of AGOT-LS from boat P-5 that had been irradiated to 6485 Mwd/At were heated to determine whether the dimensional changes could be annealed. All of the samples contracted an additional 0.067 ± 0.005 per cent when heated between 1000° C and 1500° C. No significant length change occurred during further heating to 1940° C. An interesting feature was that the samples which had been stressed during irradiation contracted equally as much as those which had been irradiated unstressed.

X-ray measurements were made to obtain the changes of the crystallite dimensions during the irradiation and annealing treatments.* During irradiation the c-spacing increased by 0.55 ± 0.13 per cent and L_c decreased by about 30 per cent. The a-spacing apparently increased about 0.02 per cent and there appeared to be a small increase in L_a . After annealing to 1940° C, the c-spacing had returned to within one standard deviation of the unirradiated value, and the a-spacing decreased to about 0.12 per cent less than its unirradiated value. L_c appeared to have increased slightly, but still remained much below the unirradiated value. The range of values obtained for L_a encompassed all previous L_a values determined both prior to and after irradiation; it was therefore impossible to obtain a meaningful average after annealing.

In addition to this series of samples, five parallel NC-8 samples were irradiated under a constant tensile stress of 100 psi. Only one irradiation period has been completed to date, and the exposure (938 Mwd/At) is insufficient to draw any final conclusions. The results show that the unstressed cylindrical samples (0.250-in. diameter by 1.50-in. long) have undergone no net length change. Unstressed "dogbone" samples (0.250-in. diameter by 1.50-in. gauge sections, 2.25-in. overall length, as in Fig. 10), decreased in length by 0.036 ± 0.019 in. This decrease may be attributed to contraction of the end sections. Stressed "dogbone" samples increased by 0.024 ± 0.009 in. overall length.

* L_c and L_a are the apparent crystallite lengths in the c and a directions, respectively.

DISCUSSION

The increase in contraction rate under applied stress is in good agreement with recent results⁴ obtained from "restrained growth experiments" if the increase is proportional to the applied stress. It is noted that the uncertainty in the contraction rates, listed in Table II, is of sufficient magnitude to encompass such a proportionality. However, it appears unlikely that the increase in rate is dependent upon stress under the conditions reported herein.

The data on the unstressed samples are consistent with previous interpretations³ in which radiation-induced contraction in graphite at high temperatures is attributed to two separable processes: (1) a vestige of low-temperature crystallite damage which produces an expansion transverse to the extrusion direction and contraction parallel to the extrusion direction; and (2) a permanent, non-annealable contraction that is nearly independent of the irradiation temperature. This latter effect, which is the dominant, long-term process at high temperatures, has been attributed in part to the relief of stresses at particle-binder junctions.⁵ The annealing results in this study confirm that the radiation-induced transverse contraction is non-annealable. In fact the samples underwent an additional contraction which is attributed to annealing of the radiation-induced crystallite expansion.

The results of this investigation appear to support the following interpretation. Within the crystallites the same series of disorder-reorder process occur for stressed samples as for unstressed samples. The principal

difference is that the compressive stress, which is transmitted to the particle-binder junctions, accelerates the non-annealable contraction in the direction of imposed stress. The mechanism which controls the contraction rate may be as follows:

- (1) Internal stresses, generated in a piece of graphite during cooling from graphitization temperature and during irradiation, are partially relieved by microcracks⁶ at external surfaces.
- (2) These microcracks are more effective in relieving stresses on small samples, where the volume-to-surface ratio is decreased, thereby reducing the contraction rate.
- (3) An externally applied compressive stress may inhibit the displacement at these microcracks and the associated stress relief at the sample edges.

If this interpretation is valid, then the results cannot be directly applied to larger samples, nor to other stress ranges, until a better understanding of sample-size effects are obtained.

CONCLUSIONS

1. Compressive stresses of 150 and 300 psi increase the rate of contraction of the irradiated graphite samples.
2. There does not appear to be a difference between the effect of 150 and 300 psi.
3. An effect of sample size on the rate of contraction of unstressed samples is observed and is in agreement with previous observations.

4. Results reported herein are in substantial agreement with the radiation-damage theory proposed earlier.³
5. An extension of the theory has been proposed which could account for the observed results. However, these data are not conclusive proof of its validity and additional work is needed to determine its acceptability.

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CAPTIONS TO FIGURES

Figure 1. Cutaway drawing of a 150 psi compression test boat.

Fig. 2 to 9 incl. Length changes of graphite samples irradiated at 625° C, with and without an externally applied compressive stress.

Figure 10. Sectional drawing of a graphite "dogbone" tensile sample.

Figure 1

Figure 2

Figure 3

Figure 4

Figure 5

Figure 6

Figure 7

Figure 8

Figure 9

$0.125''$ Radius

Section A-A

Section B-B

Figure 10