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**RADIATION-INDUCED DIMENSIONAL CHANGES
OF GRAPHITES CONTAINING CARBON BLACK**

J. W. HELM

FEBRUARY, 1966

RELEASED FOR ANNOUNCEMENT
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RADIATION-INDUCED DIMENSIONAL CHANGES
OF GRAPHITES CONTAINING CARBON BLACK

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March 1966

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RADIATION-INDUCED DIMENSIONAL CHANGES OF GRAPHITES CONTAINING CARBON BLACK

INTRODUCTION

Carbon black is used as an additive to the base mix for various graphites. The carbon black aids in obtaining the desired pore distribution in the final product, reduces the permeability, and increases the strength and density. However, the addition of carbon black, at least in large amounts, adversely affects the dimensional stability of the graphite under neutron irradiation.^(1,2) Irradiations of materials containing large percentages of carbon black have been conducted previously⁽¹⁾ to study the effect of accelerated contraction rates (between 10 and 100 times the rates for nuclear graphites) and to obtain large dimensional changes at relatively low neutron exposures. Samples containing 30-40% carbon black in the filler were found to contract 1 1/2 to 2 times as much as, and to reach a decreased contraction rate at a lower neutron exposure than, nuclear graphite.⁽²⁾

To investigate the effect of carbon black (in amounts varying up to 50% of the mix) on the subsequent dimensional stability under irradiation, a series of samples were irradiated in the Engineering Test Reactor (ETR) in two consecutive sets of capsules. A conventional nuclear-grade graphite, CSF, was used as a comparison material.⁽³⁻⁵⁾ The five carbon-black graphites were tested to provide a range for a possible use of carbon-black addition as a means of controlling permeability and yet minimizing the effect on dimensional stability.

The reference CSF graphite was obtained from a full-size extrusion, and the carbon-black samples from experimental cylindrical extrusions. The graphites are described in Table I. The carbon-black graphites were made by mixing the

Thermax carbon black with the graphite powder, adding the pitch as binder, and molding into cylinders. Then all were heat treated in one batch at 3000 °C.

TABLE I

GRAPHITE DESCRIPTION

<u>Sample Designation</u>	<u>Block Number</u>	<u>Block Size, in.</u>	<u>Remarks (a)</u>		
CSF ^(b)	142	4 x 4 x 50	Reactor grade; Cleves coke (conventional type). F-purified; graphitized at 2800 °C; density, 1.66 g/cm ³		
			<u>Composition of the Mix % of Filler</u>		
			<u>Artificial Graphite Powder</u>	<u>Thermax Carbon Black</u>	<u>Pitch Binder Content</u>
0% black ^(c)	61-75	2 diam x 4.2	100	0	440
20% black ^(c)	61-76	2 diam x 3.0	80	20	285
25% black ^(c)	61-77	2 diam x 3.0	75	25	285
33% black ^(c)	61-78	2 diam x 3.0	67	33	265
50% black ^(c)	61-79	2 diam x 3.0	50	50	310

(a) Further details on the CSF graphite may be found in Nuclear Graphite.⁽⁶⁾

(b) Manufactured by Carbon Products Division, Union Carbide Corporation.

(c) Manufactured by Pechiney Company, France and supplied by L. W. Graham, Dragon Project, Winfrith, England.

The sampling schemes for the CSF and carbon-black graphites are shown in Figures 1 and 2, respectively. After machining to a quarter-round shape, the individual samples were annealed at 1800 °C for 1 hr at a pressure of 3×10^{-4} Torr.

The basic noninstrumented capsule used for the irradiations has been discussed previously.⁽⁷⁾ Lengths were measured physically from end-to-end on all samples by a Bausch and Lomb DR-25 Optical Gage (Type 33-14-23). The six capsules were installed in the ETR. A complete capsule history is given in Table II.

FIGURE 1

SAMPLING SCHEME - CSF GRAPHITE

FIGURE 2

SAMPLING SCHEME - CARBON-BLACK GRAPHITE

TABLE II
CAPSULE HISTORY

<u>Capsule Number</u>	<u>ETR Position</u>	<u>ETR Elevation, in.</u>	<u>Installation Date</u>	<u>Removal Date</u>	<u>Effective Days*</u>
GEH-13-147	E5 NE	12 to 18	4-11-62	88.5	9-17-62
GEH-13-148	"	18 to 24	"	"	"
GEH-13-149	"	24 to 30	"	"	"
GEH-13-173	"	12 to 18	12-5-62	85.7	5-13-63
GEH-13-174	"	18 to 24	"	"	"
GEH-13-175	"	24 to 30	"	"	"

*Full-power operation at 174 MW

The end-to-end length measurements (described above) were repeated on all samples. Sample irradiation temperatures, estimated to be ± 100 °C, are determined by heat-transfer calculations⁽⁸⁾ and confirmed by comparison of reference CSF length data with data obtained from instrumented capsule irradiations.⁽³⁻⁵⁾

Neutron exposures are based on the activities of nickel and iron foils contained in each capsule. These activities are used in conjunction with the activation cross sections for each material and neutron-spectra calculations⁽³⁻⁵⁾ for the appropriate capsule position.

IRRADIATION RESULTS

Length changes for the carbon-black samples are presented in Table III (Figures 3-5). For all capsules, the transverse CSF reference data agreed very well with the CSF data from instrumented capsules.⁽³⁻⁵⁾ The solid lines connect the results from the same sample reirradiated. The results from the individual capsules are plotted on separate figures to provide a direct comparison of the types in the given capsule.

TABLE III

RESULTS OF CARBON-BLACK GRAPHITE IRRADIATIONS

Capsule	Sample Code	Sample Number	Temperature, °C	Exposure- ²¹ nvt x 10 ⁻²¹ E > 0.18 MeV	$\Delta L/L$, %	Exposure- ²¹ nvt x 10 ⁻²¹ E > 0.18 MeV (Total)	$\Delta L/L$, % (Total)
Number	CSF 1	142-192	600-700	1.20	-0.051	2.31	-0.212
	0% black 1	61-75-1			-0.088		-0.286
	0% black II	61-75-201			-0.158		-0.397
	20% black 1	61-76-1			-0.076		-0.241
	20% black II	61-76-201			-0.157		-0.330
	50% black 1	61-79-1			-0.073		-0.148
	50% black II	61-79-205			-0.140		-0.330
Number	CSF 1	142-191	500-600	1.21	-0.083	2.30	-0.302
	20% black 1	61-76-2			-0.116		-0.313
	20% black II	61-76-202			-0.192		-0.502
	33% black 1	61-78-1			-0.122		-0.310
	33% black II	61-78-201			-0.178		-0.462
	50% black 1	61-79-2			-0.114		-0.268
	50% black II	61-79-206			-0.178		-0.391
Number	CSF 1	142-194	500-600	0.90	-0.070	1.84	-0.226
	0% black 1	61-75-2			-0.094		-0.260
	25% black 1	61-77-1			-0.096		-0.253
	25% black II	61-77-201			-0.158		-0.373
	33% black 1	61-78-2			-0.100		-0.226
	33% black II	61-78-202			-0.142		-0.372

FIGURE 3

Physical Distortion of Pechiney Carbon-Black Graphite
as a Function of Exposure

FIGURE 4

Physical Distortion of Pechiney Carbon-Black Graphite
as a Function of Exposure

FIGURE 5

Physical Distortion of Pechiney Carbon-Black Graphite
as a Function of Exposure

For all the carbon-black graphites, the parallel samples contracted more than the transverse. This behavior is typical of reactor-grade graphites.⁽³⁻⁵⁾ Among the transverse samples, CSF showed the least contraction after the first irradiation period. However, after the second irradiation, the graphite containing the largest percentage of carbon black (50%) showed the least contraction. After the second irradiation, the 33% black material contracted about the same as the CSF; the 25%, 20%, and 0% material showed successively more contraction.

In the parallel direction, the same behavior was observed. The 50% black graphite showed the least contraction at the end of the second irradiation, the 0% black graphite the most, and the others about the same amount between the two extremes.

DISCUSSION

The samples containing progressively smaller amounts of carbon black, 50% down to 0%, showed progressively more contraction in both the transverse and parallel directions at the highest exposures. This is in exact contrast to data⁽⁹⁾ at much lower exposures, approximately 6×10^{20} nvt, which show the most contraction for the graphite containing 10% carbon black. Since at the lower exposures reported here the carbon-black graphites all showed about the same behavior, it appears that in the transverse direction graphites containing the higher percentages of carbon black contract at a very rapid rate initially and then at a slower rate. This agrees with trends seen for less-graphite materials, such as TSGBF which is graphitized at 2450 °C, that reach turnaround⁽³⁻⁵⁾ more rapidly than conventional nuclear graphites.⁽⁴⁾ It also tends to agree with data⁽²⁾ on other materials containing carbon black which showed a tendency toward turnaround sooner than the conventional CSF graphite. In those materials the percentage of binder was much less than for the carbon-black graphites reported herein. Thus, it appears the carbon black causes the contraction to

proceed so rapidly that the c expansion of the crystallites becomes controlling at quite low exposures, which leads to the decreased contraction rate. In reactor applications this may mean that the probable expansion at the high neutron exposures will be the critical item in the transverse direction. At the exposures obtained, it appears that carbon black percentages between 20 and 33% do not adversely affect the contraction behavior; however, at higher exposures this may not be true.

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