**EFFECTS OF SOME PROCESS
TEMPERATURES
ON RADIATION INDUCED CONTRACTION
OF GRAPHITE**

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SEPTEMBER, 1967

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ON RADIATION INDUCED CONTRACTION OF GRAPHITE

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EFFECTS OF SOME PROCESS TEMPERATURES
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J. H. Cox, Jr., R. W. Marek, and W. E. Parker

ABSTRACT

The dimensional change that occurs during the thermal processing of graphite and the subsequent dimensional change that occurs when graphite is irradiated has been explored. A lower rate of rise to the graphitization temperature yielded a slight improvement in dimensional stability under neutron irradiation for the transverse orientation; however, in the parallel orientation there was no appreciable effect of graphitization rate on dimensional stability.

A second series of samples was used to demonstrate the degree to which binder heat treatment influences radiation induced dimensional changes. The filler for the formulation was graphitized to 2900 °C and then mixed with a graphitizable binder. Subsequently, the samples were heat treated to temperatures appropriate for a broad range of crystallite development in the binder phase. Differences in radiation induced contraction can be attributed to the binder phase. Unlike a system in which all constituents have the same graphitization temperature, this series does not show a uniform increase in contraction rate with decreasing graphitization temperature. There is little effect of heat treatment temperature down to 2000 °C; at heat treatment temperatures below 2000 °C there is a sharp increase in contraction rate for both orientations.

INTRODUCTION

This report presents an investigation into the dimensional change that occurs during the thermal processing of graphite and the subsequent dimensional change that occurs when graphite is irradiated.

When the first nuclear reactors were built there were predictions that the graphite used would either contract, expand, or disintegrate. In fact, the graphite did exhibit both contraction and expansion under specific conditions. However, at operating temperatures greater than 300 °C graphite contraction problems were predominant. Therefore, techniques for decreasing graphite contraction were required.

The accumulation of irradiation data for different graphites also proved to be necessary so that the reactor engineers could compensate adequately for the graphite characteristics. Accordingly, many exploratory programs

were initiated to investigate graphite behavior before, during, and after irradiation.

In one of these investigations, graphites prepared with certain metallic additives exhibited greater dimensional change stability than graphites prepared without additives.⁽¹⁾ The use of additives generally results in a greater volume contraction during graphite manufacture than occurs when additives are not used. Consequently, it was proposed that if graphite undergoes greater contraction during processing, it may also exhibit increased stability when irradiated. No correlation could be made from the available data because of the possible interaction of the variety of raw materials and formulations used to prepare nuclear graphites.

In our experiment, the fabrication of a series of graphites which would have a systematic range of dimensional change during processing was desired.

Furthermore, the dimensional change was not to be influenced by raw material or formulation variations. This was successfully accomplished by preparing graphites from one formulation and controlling the dimensional change by varying the graphitizing rate. The first part of this paper describes the formulation, fabrication and irradiation data for this series of samples.

The second part deals with the degree of binder graphitization and its effect on graphite characteristics. Although other exploratory programs relating to this subject had been performed,^(2,3) there was no consistent investigation of the effect. Graphites had been prepared previously from various binders that exhibited different degrees of graphitizability. Therefore, our samples were prepared from a graphitized filler and one graphitizable binder. The formed pieces were heat-treated to temperatures between 700 °C and 2900 °C to obtain a broad range of binder graphitization.

EXPERIMENTAL

PHASE I--CONTROL OF DIMENSIONAL CHANGE BY VARIATION OF GRAPHITIZATION RATE

Raw Materials and Formulation

Sohio Lima coke from the Sohio Oil Company at Lima, Ohio was selected for the filler component of the formulation. After this material was vertically calcined to 1400 °C, it was crushed and screened to obtain 20/35 particles (U.S. Standard Screen) and No. 60 flour.

Allied Chemical Company's No. 30 medium pitch was used as the binder, and Ebony "E" oil, obtained from the Atlantic Refining Company, was employed as an extrusion aid.

All raw materials were thoroughly blended in a twin shell blender. Likewise, each charge for the mixer was thoroughly blended. Presented in Table I is the description of the formulation used for loading a 2 gal capacity sigma blade mixer.

TABLE I. Phase I--Formulation

2400 g No. 60 coke flour
600 g 20/35 coke particles
960 g of -20 mesh No. 30 pitch
30 g of Ebony "E" oil

Forming and Baking

The coke-pitch blend was added to the preheated mixer and mixed for 30 min at 130 °C. Just prior to the conclusion of mixing, the Ebony "E" lubricant was added. After cooling, the mix was crushed and stored until sufficient material was accumulated for extrusion. All crushed material was again thoroughly blended before the extrusion so that each charge to the extrusion press would be representative. Three-inch-diameter rods were extruded with a vertical-loading, horizontal extrusion press.

After sizing into 8 in. lengths, the rods were packed in a sand-metallurgical coke mixture and baked in silicon carbide saggers. Baking was performed in an electric furnace according to the schedule shown in Table II.

Graphitization

To significantly affect the amount of dimensional change occurring during the thermal processing of baked carbons, seven sets of seven to eight rods each were graphitized to 2900 °C, in an inert atmosphere, according to the rates shown in Table III.

TABLE II. Phase I--Baking Schedule

5 °C/hr from 25 to 160 °C
3 °C/hr from 160 to 646 °C
5 °C/hr from 646 to 900 °C
Dwell time of 3 hr at 900 °C

TABLE III. Phase I--Graphitization Rate Schedule

Set	Inert Atmosphere	Graphitization Rate (°C/hr)
1	Argon	50
2	Argon	100
3	Argon	500
4	Nitrogen	1,000
5	Nitrogen	3,400
6	Nitrogen	10,000
7	Nitrogen	30,000

PHASE II--EFFECT OF DEGREE OF BINDER GRAPHITIZATION

Raw Materials and Formulation

Rotary calcined needle coke obtained from Great Lakes Carbon Company was used as the filler component. Although it had been calcined, a further heat-treatment to 2900 °C was performed. Consequently, subsequent heat-treatments of the formed bodies primarily affected the binder phase.

Since crushing, grinding, or sieving the filler after graphitizing would tend to mitigate its purity, the calcined coke was sized before additional thermal processing. As in the previous series, the coke was crushed and sieved to obtain 20/35 particles and No. 60 flour.

A two-step thermal processing procedure was used. The sized filler material was placed in covered graphite saggars and heat-treated in a nitrogen atmosphere to a temperature of 1700 °C at a rate of 3000 to 3500 °C/hr. The cooled material was then given the final heat-treatment to 2900 °C under similar conditions.

Effects of thermal treatment on X-ray measurements and ash levels of the filler are presented in Table IV.

TABLE IV. Phase II--G.L.C. Needle Filler--Effect of Particle Size and Heat-Treatment

Treatment Temperature	Crystallite Size (Å)		Interlayer Spacing (Å)		Ash Levels (ppm)	
	20/35 (a)	No. 60 (b)	20/35	No. 60	20/35	No. 60
Calcined	38	39	3.463	3.469	1800	1100
1700 °C	92	90	3.437	3.438	1200	1100
2900 °C	687	688	3.362	3.362	5	16

(a) 20/35 particles

(b) No. 60 - Refers to No. 60 flour.

These results demonstrate the growth in crystallite size and decrease in interlayer spacing with increasing processing temperatures. Thermal purification at 2900 °C was also outstanding and particle size effects did not appear significant. Formulating, and forming conditions were the same as those reported for Phase I.

Thermal Processing

To determine the extent that binder graphitization affects the radiation induced dimensional changes, samples were treated to the seven top temperatures listed in Table V.

TABLE V. Phase II--Thermal Processing

Set	Top Temperature
8	700 °C
9	900 °C
10	1400 °C
11	2000 °C
12	2350 °C
13	2750 °C
14	2930 °C

Heating Schedule: 25 to 160 °C at 5°/hr; 160 to 646 °C at 3°/hr; 646 to 700 or 900 °C at 5°/hr; 900 °C to top temperature at 100°/hr; 3 hr dwell at top temperature.

Green rods were baked in silicon carbide saggers according to the procedure described in Phase I. Sets 11 to 14 were baked to 900 °C and subsequently processed to the representative temperature in an induction furnace. An argon atmosphere was maintained for both the heatup and cooling cycle. The reheating of these samples to 900 °C in the induction furnace was at the rate of 200 °C/hr.

IRRADIATION STUDIES

Sample Preparation

Each 3 in. extrusion was sampled as shown in Figure 1. The samples were then machined to 0.43 in. diam cylinders; the transverse samples averaged 2.8 in. long, and the parallel samples averaged 3.9 in. long. The sample ends were polished for length measurements. All samples from both sample series, except those processed at 700 and 900 °C, were annealed at 1000 °C for one hour to relieve machining stresses.

FIGURE 1. Sampling Diagram for Irradiation Samples

Property Measurements

Pre- and postirradiation length measurements were made with a Baush and Lomb DR-25 Optical Gage (Type 33-14-23), checked against Lufkin standard test gages. Length measurements on individual samples had a standard deviation of 2×10^{-5} in.

Coefficient of thermal expansion values were determined for the 25 to 425 °C range. A quartz correction of 0.549×10^{-6} in./in. °C was applied to the results.

Irradiation

The samples were enclosed in graphite containers during irradiation in the Hanford reactor hot test holes. Irradiation temperature varied in the range 550 to 700 °C, but was the same for all samples. The atmosphere was a mixture of helium and carbon dioxide.

Neutron exposures were measured using nickel, iron, and cobalt flux monitor wires. All exposures are for neutron energies greater than 0.18 MeV.^(4,5) Integral cross-sections for the fast-neutron monitors were determined from the cross-sections for the materials and the calculated neutron spectra for the irradiation facility. The experimental details of determination of neutron exposures are described by Morgan,⁽⁶⁾ who also described the computer code (FOILS) used.

RESULTS AND DISCUSSION

PHASE I--CONTROL OF DIMENSIONAL CHANGE VIA GRAPHITIZATION RATE

Percent change in volume is allied to graphitization rate as shown by the data in Table VI. The change in volume for the "Green to Graphite" condition and "Baked to Graphite" state demonstrate the excellent correlation with graphitization rate. Although "Green to Graphite" values indicate the overall change, the "Baked to Graphite" values

are also of interest since they result from changes in processing conditions.

TABLE VI. Phase I--Percent Change in Volume

Item	Graphitization Rate	% Change in Volume	
		Green to Graphite	Baked to Graphite
1	50 °C/hr	-5.6	-2.8
2	100 °C/hr	-5.2	-2.5
3	500 °C/hr	-3.1	-0.5
4	1,000 °C/hr	-2.4	-0.2
5	3,400 °C/hr	-0.9	+1.5
6	10,000 °C/hr	+0.3	+3.4
7	30,000 °C/hr	+4.9	+8.5

At a heating rate of 50 °C/hr the shrinkage is at, or very near, a limiting value. However, the expansion that results from graphitization rates greater than 1500 °C/hr does not approach a limiting value even at the rate of 30,000 °C/hr. This relationship is linear between 3000 and 30,000 °C/hr; an average expansion of 0.26% occurs for every 1000 °C/hr increase in graphitization rate. These results are graphically displayed in Figure 2.

Changes in apparent density also correlate well with volume change data and the graphitization rate. Figure 3 represents the plot of the final graphite density versus graphitization rate. With increasing graphitization rate there is a consistent and predictable decrease in density.

PHASE II--EFFECT OF GRAPHITIC CHARACTER OF THE BINDER

de Halas and Yoshikawa proposed that the mechanism of radiation damage is dependent on poorly graphitized regions

FIGURE 2. Phase I--Percent Volume Change (Bake to Graphite) versus Graphitizing Rate, Sets 1 to 7

FIGURE 3. Graphite Density versus Graphitization Rate, Sets 1 to 7

of artificial graphite.⁽⁷⁾ They indicated that regions of lower graphitic character were located at the crystallite and particle boundaries, i.e. in the binder phase.

The effect of degree of binder graphitization on graphite properties has been studied by substituting binders of various degrees of graphitizability.^(2,3) For example, samples prepared with an "easily graphitized" binder such as No. 30 medium coal-tar pitch were compared to those prepared with a "not easily graphitized" binder like phenolformaldehyde resin. Although samples prepared from the more graphitizable binders appeared to be more dimensionally stable when irradiated, additional information was required to establish the validity of the conclusion, since "graphitizability" was not the only characteristic that differentiated the binders.

"Graphitizability" may be considered as a measure of the potential for crystallite growth and an approach to the theoretical interlayer spacings for graphite. Crystallite growth is very dependent on temperature, but the limitation for growth is a characteristic of the raw material.

For this investigation, the filler was graphitized to 2900 °C before it was mixed with the "graphitizable binder". Samples were subsequently heat-treated to temperatures considered to be suitable for the development of a broad range of crystallite growth in the binder phase. Since the development of crystallite size in the filler was set by its previous heat treatment, changes in radiation induced contraction may be attributed to the degree of graphitization of the binder.

Although the characteristics of apparent density and/or volume change during fabrication are not of primary interest here, the effects are significantly different from those obtained in Phase I. In Phase II, a net volume expansion was observed after thermal processing.

The data in Table VII indicate that the largest net change in volume was measured after samples were heated to the lowest temperature (700 °C). Although shrinkage of the binder phase occurred on heating to temperatures greater than 900 °C, the net binder shrinkage was not sufficient to counteract the earlier low temperature expansion.

TABLE VII. Phase II--Apparent Density and Volume

Top Temperature (°C)	Apparent Density (g/cm ³) Green - Top Temperature		% Δ in Volume Green - Top Temperature
700	1.82	1.46	+23.8
900	1.81	1.51	+16.2
1400	1.81	1.48	+16.2
2000	1.81	1.48	+11.5
2350	1.83	1.46	+15.0
2750	1.81	1.47	+10.9
2900	1.81	1.50	+12.0

The effect of top temperature on electrical resistivity is displayed in Figure 4. Since the filler had already been graphitized, the change in resistivity with treatment temperature may be assumed to be indicative of the developing graphitic character of the pitch (binder) coke. An expected trend to decreasing resistivity values is observed.

In Figure 5, the efficacy of thermal purification is graphically demonstrated. The filler initially was thermally purified to an ash level of less than 16 ppm. However, when the filler was mixed with pitch and processed normally, the ash level of the samples was increased to more than 500 ppm. At 2350 °C the ash level had been decreased to 32 ppm; at 2900 °C the ash level was less than 5 ppm.

FIGURE 4. Effect of Temperature (Top) on Resistivity

FIGURE 5. Phase II--Ash Level versus Top Temperature

IRRADIATION STUDIESCoefficient of Thermal Expansion

Coefficient of thermal expansion (CTE) values for both sample series are given in Table VIII.

Although decreasing the graphitization rate during processing increased

the apparent density of the samples, no similar effect was observed in the CTE or in the change in CTE after irradiation. The density change indicates that the pore volume of the graphite increases with graphitization rate, but the lack of change in CTE implies that the fine crack volume or crystallinity does not vary.

TABLE VIII. *Coefficient of Thermal Expansion Values^(a)*

Graphitization Rate, °C/hr	Orientation	Exposure: 2.56×10^{20} nvt		
		CTE (25 to 400 °C) $\times 10^6$		
		Initial	Irradiated	% Change
50	Transverse	3.31	4.89	+47.7
	Parallel	1.70	2.09	+22.9
100	Transverse	3.34	5.00	+49.7
	Parallel	1.70	2.10	+23.5
500	Transverse	3.12	4.48	+43.6
	Parallel	1.67	2.10	+25.7
1,000	Transverse	3.41	4.83	+41.6
	Parallel	1.63	2.06	+26.4
3,400	Transverse	3.11	4.46	+43.4
	Parallel	1.78	2.10	+18.0
10,000	Transverse	3.34	4.67	+39.4
	Parallel	1.64	2.11	+28.7
30,000	Transverse	3.03	4.63	+52.8
	Parallel	1.77	2.14	+20.9
Binder Temperature, °C				
700	Transverse	4.77	7.45	+56.2
	Parallel	1.35	1.83	+35.6
900	Transverse	4.11	6.96	+69.3
	Parallel	1.68	2.12	+26.2
1,400	Transverse	3.41	5.77	+69.2
	Parallel	1.15	1.59	+38.3
2,000	Transverse	3.32	5.20	+56.6
	Parallel	1.24	1.39	+12.1
2,350	Transverse	3.75	5.70	+52.0
	Parallel	1.16	1.20	+ 3.4
2,750	Transverse	3.23	4.49	+39.0
	Parallel	1.01	1.19	+17.8
2,900	Transverse	3.17	4.58	+44.5
	Parallel	1.04	0.98	- 5.7

(a) All values represent the average of two samples.

After an exposure of 2.56×10^{20} nvt, CTE values increased 40 to 50% for all transverse samples and 18 to 28% for all parallel samples. This increase may indicate possible crack closure with irradiation. Since the cracks that affect CTE are believed to be primarily between planes and hence concentrated in the parallel orientation, the largest increase should be expected in the transverse samples as the data show.

In general, samples with lower binder

graphitization temperatures displayed higher initial coefficients of thermal expansion and also experienced the largest increase due to irradiation. Thus it is quite apparent that the binder material makes a significant contribution to the bulk behavior.

Length Changes

Radiation induced sample length changes are listed in Table IX for both sample series.

TABLE IX. Length Changes

Graphitization Rate, °C/hr	Exposure, nvt x 10^{-20}	Percent Length Change ^(a) and Standard Deviation	
		Transverse	Parallel
50	2.56	+0.021 ± 0.001	+0.012 ± 0.001
	9.32	-0.028 ± 0.007	-0.091 ± 0.003
100	2.56	+0.020 ± 0.001	+0.014 ± 0.001
	9.32	-0.029 ± 0.006	-0.087 ± 0.004
500	2.56	+0.018 ± 0.001	+0.012 ± 0.003
	9.32	-0.047 ± 0.002	-0.096 ± 0.012
1,000	2.56	+0.024 ± 0.002	+0.017 ± 0.001
	9.32	-0.051 ± 0.003	-0.102 ± 0.001
3,400	2.56	+0.027 ± 0.008	+0.020 ± 0.003
	9.32	-0.051 ± 0.005	-0.093 ± 0.003
10,000	2.56	+0.032 ± 0.001	+0.026 ± 0.001
	9.32	-0.050 ± 0.002	-0.100 ± 0.003
30,000	2.56	+0.036 ± 0.002	+0.025 ± 0.001
	9.32	-0.051 ± 0.003	-0.096 ± 0.001
Binder Temperature, °C			
700	2.56	-0.046 ± 0.123	-0.856 ± 0.036
	9.32	-1.370 ± 0.228	-1.694 ± 0.032
900	2.56	-0.836 ± 0.112	-0.858 ± 0.034
	9.32	-0.996 ± 0.171	-1.941 ± 0.050
1,400	2.56	-0.280 ± 0.051	-0.215 ± 0.014
	9.32	-0.871 ± 0.162	-0.984 ± 0.043
2,000	2.56	-0.022 ± 0.003	-0.003 ± 0.002
	9.32	-0.168 ± 0.002	-0.205 ± 0.010
2,350	2.56	-0.029 ± 0.004	+0.003 ± 0.000
	9.32	-0.181 ± 0.011	-0.145 ± 0.001
2,750	2.56	+0.018 ± 0.006	+0.009 ± 0.002
	9.32	-0.056 ± 0.006	-0.101 ± 0.004
2,900	2.56	+0.019 ± 0.002	+0.007 ± 0.001
	9.32	-0.036 ± 0.005	-0.098 ± 0.015

(a) Each value represents the average of four samples.

Apparently the graphitization heating rate makes little difference in the radiation induced contraction rate for either orientation. The length change data for this series are presented in Figure 6. The transverse samples show a slightly lower contraction rate for the lower graphitization rate, while the parallel samples show no difference at this exposure level. Certainly, the dimensional change that occurred during thermal processing does not correlate with the changes observed during irradiation.

Both orientations of the binder graphitization series show a definite effect of binder processing temperature

on the contraction rate, as shown in Figure 7 and 8. An increase in contraction rate is apparent with decreasing processing temperature. The sample sets heat-treated from 2000 to 2900 °C show a slight variation, while the contraction rate is greatly enhanced for temperatures below 2000 °C. This contrasts to the behavior of materials with both the binder and filler heat-treated to various temperatures. A sample series with both binder and filler heated to various temperatures from 1400 to 2700 °C has shown⁽⁸⁾ a contraction rate uniformly decreasing with increasing processing temperature.

FIGURE 6. Graphitization Rate Series--
Length Changes

FIGURE 7. Binder Graphitization
Series--Transverse Sample Length Changes

FIGURE 8. Binder Graphitization
Series--Parallel Sample Length Changes

These observations imply that the binder materials significantly contribute to the graphite irradiation dimensional change when extreme binder variations are encountered, e.g., widely differing processing temperatures or degrees of graphitization of the binder material. Likewise, other investigations⁽⁸⁾ have shown that only large increases in binder content have a significant effect on dimensional behavior when exposed to a neutron field.

Based on the length change data, it appears that the 700 and 900 °C binder temperature sets are entering the turnaround portion of the dimensional change curve. However, both of these sample sets warped during the last exposure period. An example is shown in Figure 9. As a result, the increase in contraction rate compared to the higher processing temperature samples is real, but the absolute magnitude is questionable.

The binder graphitization series has been included in irradiations, at a

test reactor, which will reach exposures in excess of 2×10^{21} nvt. The result of these tests will verify the above data and possibly clarify the indicated turnaround behavior.

CONCLUSIONS

By varying the graphitization rate the dimensional change resulting from the thermal processing of baked carbon bodies may be closely controlled. However, dimensional change that occurs during processing is not indicative of the dimensional change that occurs when graphite is irradiated.

Samples were processed to obtain a consistent crystallite development in the binder phase. When these samples were irradiated, they exhibited dimensional changes that correlate with the processing "heat treatment" temperatures. This program validates the theory that the "graphitizability" of filler and binder is an important

Warped Parallel Samples
900 °C Binder Temperature
Exposure = 9.32×10^{20} nvt

FIGURE 9. Binder Graphitization Series

characteristic in determining the dimensional stability of irradiated graphite. If the binder graphitization temperature is less than 2000 °C, the contraction rate is greatly enhanced. However, between 2000 and 2900 °C there is little effect on the dimensional stability. In contrast, materials with both the filler and binder graphitized to the same temperature show a uniform increase in contraction rate with decreasing graphitization temperature. Hence, there is incentive to use a pregraphitized filler for materials which require a low final heat treatment, e.g., boronated graphite or possibly fueled graphite.

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