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THERMAL CONDUCTIVITY
CHANGES IN NUCLEAR
GRAPHITES AT HIGH TEMPERATURES
AND HIGH FLUENCES

by

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ABSTRACT

An anisotropic needle-coke graphite and an isotropic Gilsocoke graphite have been irradiated at 560° to 1455°C at fluences of 1.40 to 8.00×10^{21} n/cm² ($E > 0.18$ MeV) and thermal conductivity measured. The data extend thermal conductivity measurements on irradiated graphites to 1450°C. There is a strong temperature effect; thermal conductivity degradation is reduced with increasing temperature and fractional changes ranged from 20 to 25% at 1450°C, when measured at room temperature.

INTRODUCTION

High-temperature gas-cooled reactors utilize prismatic graphite fuel blocks as a neutron moderator and structural material to support the fuel. The heat generated in the fuel is transferred to the helium coolant through graphite webs. Thus, the thermal conductivity of the graphite during the life of the fuel is an important design property, and the effect of irradiation on the thermal conductivity as a function of fluence and irradiation temperature over the service conditions of the reactor must be known. It is desirable that the thermal conductivity remain high and degradation be minimized.

A program to study the effect of irradiation on thermal conductivity of an anisotropic needle-coke graphite (H-327) and an isotropic Gilsocoke graphite (H-328) has been under way for several years. Specimens were irradiated at 560° to 1455°C to fluences of 1.4 to 8×10^{21} n/cm² (E > 0.18 MeV).

EXPERIMENTAL PROCEDURE

A description of the graphites and their properties has been published elsewhere (Ref. 1). Graphite H-327 is a production-size (18-in. diameter by 34 in. long) extruded needle-coke graphite. Graphite H-328 is a production-size (17-in. diameter by 34 in. long) molded Gilsocoke graphite.

The irradiations were carried out in instrumented capsules in the Engineering Test Reactor (ETR), Idaho Falls, Idaho. A description of the capsules and dosimetry has been published previously (Refs. 2,3).*

A standard laboratory flash technique was employed to measure thermal diffusivity at room temperature or approximately 24°C. Thermal conductivity values were calculated from the thermal diffusivity data as described in Refs. 4 and 5.

The graphite specimens (approximately 0.500-in. diameter by 0.050 in. thick) were prepared prior to irradiation and irradiated within a closed graphite crucible.

*Complete analyses of the temperatures and fluences have not been completed; therefore, those given were estimated. However, final analyses are not expected to affect the temperature order of the curves or the results and conclusions.

EXPERIMENTAL RESULTS AND DISCUSSION

The thermal conductivity data for graphites H-327 and H-328, measured at room temperature, are given in Tables 1 and 2 and plotted as a function of fluence in Figs. 1 through 3. Due to the small size of the specimens, specimen numbers were not inscribed on each sample, but each was identified by position in the crucible. Several of the crucibles were spilled during unloading of the first irradiation capsule. While none of the specimens were mixed between cells, a number lost their identity for the second irradiation. Therefore, the mean thermal conductivity value and the range of values were taken as the criteria for plotting the data.

The thermal conductivity is initially reduced for all irradiation temperatures and both graphites in a manner observed previously on these graphites (Refs. 6,7) and by others on similar graphites (Refs. 8-11). A portion of the data presented here was taken above 1200°C at 1360° to 1455°C and to fluences of up to 8×10^{21} n/cm². The curves at 1040° to 1455°C are flat between about 3 and 8×10^{21} n/cm². There is a strong temperature effect; conductivity degradation is reduced with increasing irradiation temperature. More data at lower fluences are needed to determine whether saturation occurs at decreasing fluences with increasing irradiation temperature.

Taylor et al. (Ref. 10) have analyzed the behavior of the thermal conductivity changes in highly annealed pyrolytic carbons in the range 30° to 450°C to 5×10^{20} n/cm² and reactor graphites at 200° to 1350°C to 5×10^{21} n/cm² in terms of the lattice defects that cause dimensional changes. These authors concluded that thermal conductivity changes are due to submicroscopic clusters, vacancies in the lattice, and vacancy loops, and that interstitial loops and collapsed vacancy lines, which are

responsible for dimensional changes, are insignificant. They indicate thermal conductivity changes of reactor graphites are larger at irradiation temperatures above 650°C than can be accounted for by vacancy content estimated from lattice parameter changes. Estimates of room temperature thermal conductivity changes by the analysis of crystallite dimensional changes in reactor graphites at 650° to 1350°C to 4×10^{21} n/cm² showed the vacancy loops responsible for thermal conductivity reduction increase both in size and separation with increasing irradiation temperature. This is in agreement with the data presented here and may explain the smaller fractional changes in conductivity with increasing irradiation temperature.

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TABLE 1
THERMAL CONDUCTIVITY CHANGES FOR H-327 GRAPHITE

Orientation	Unirradiated K (cal/cm-sec-°C)	Fluence 1 $\times 10^{-21}$ (n/cm ²) (E > 0.18 MeV)	Temperature 1 (°C)	K (cal/cm-sec-°C)	Fluence 2 $\times 10^{-21}$ (n/cm ²) (E > 0.18 MeV)	Temperature 2 (°C)	Total Fluence $\times 10^{-21}$ (n/cm ²)	Temp. Range (°C)	K (cal/cm-sec-°C)
H	0.525 0.563	0.80	560	0.198	0.60	605	1.40	560-605	0.162
				0.177					0.152
				0.200					0.197
				0.216					0.100
				0.218					0.175
				0.200					0.207
				0.169					0.148
				--					0.179(a)
				--					0.172(a)
				--					0.210(a)
I	0.364 0.365	0.80	560	0.106	0.60	605	1.40	560-605	0.110
				0.096					0.080
				0.129					0.105
				0.096					0.080
				0.086(b)					--
				0.118(b)					--
				0.103(b)					--
				0.120					0.088
				0.109					0.093
				0.125					0.106
				0.122					0.134
				0.115					0.140
				0.142					0.102
				0.124					0.097
				0.129					0.110
				0.129					0.106
				0.116					0.123
				--					0.123
				--					0.145
				--					0.151
H	-- --	1.40	830	0.157	1.20	870	2.60	830-870	0.123
				0.134					0.145
				0.134					0.151
				0.127					0.126
				0.171					0.150
				0.157(b)					--
				--					0.219(a)
				--					0.119
				--					0.093
				--					0.102
I	0.369 0.351	1.40	830	0.094	1.20	870	2.60	830-870	0.088
				0.093					0.100
				0.092					0.109
				0.089					--
				0.104					--
				0.127					0.093(a)
				0.098(b)					--
				0.089(b)					--
				--					0.093(a)
				--					0.093(a)
H	0.541 0.549 0.525	3.30	1040	0.275	2.10	1205	5.40	1040-1205	0.281
				0.182					0.292
				0.317					0.281
				0.305					--
				0.303					0.303
				0.389					0.287
				0.300(b)					--
				--					0.296(a)
				--					0.291(a)
				--					0.291(a)
I	0.371 0.372 0.363 0.365	3.30	1040	0.185	2.10	1205	5.40	1040-1205	0.188
				0.176					0.197
				0.205					0.197
				0.182					0.203
				0.201					0.177
				0.182					0.198
				0.176					0.234
				0.209					0.202
				0.177					0.271
				0.207					0.220
				0.212					0.212
				0.159					0.197
				0.174					0.200
				0.199					0.204
				0.162(b)					--
				0.202(b)					--
				--					0.215(a)
				--					0.203(a)
				--					0.203(a)

(a) Added for 2nd irradiation.
(b) Removed after 1st irradiation.

TABLE 1 (continued)

Orientation	Unirradiated K (cal/cm-sec-°C)	Fluence 1 $\times 10^{-21}$ (n/cm ²) (E > 0.18 MeV)	Temperature 1 (°C)	K (cal/cm-sec-°C)	Fluence 2 $\times 10^{-21}$ (n/cm ²) (E > 0.18 MeV)	Temperature 2 (°C)	Total Fluence $\times 10^{-21}$ (n/cm ²)	Temp. Range (°C)	K (cal/cm-sec-°C)
II	0.531 0.502 0.525 0.505	4.20	1360	0.266 0.292 0.326 0.336 0.333 0.296 0.359 0.337 ^(b)	3.10	1360	7.30	1360	0.285 0.251 0.260 0.243 0.219 0.221 0.224 --
				--	3.10	1360	3.10	1360	0.252 ^(a)
I	0.374 0.361 0.360 0.374	4.20	1360	0.225 0.213 0.175 0.225 0.213 0.231 0.240 0.224 0.262 0.193 0.248 0.212 0.221 0.238 0.251 ^(b) 0.222 ^(b) 0.232 ^(b)	3.10	1360	7.30	1360	0.227 0.229 0.235 0.255 0.265 0.239 0.199 0.252 0.227 0.242 0.218 0.257 0.224 0.251 -- -- --
				--	3.10	1360	3.10	1360	0.215 ^(a) 0.253 ^(a)
II	-- --	4.55	1445	0.310 0.290 0.383 0.364 ^(b) 0.339 ^(b) 0.206 0.194 0.203 0.202 0.223 ^(b)	3.50	1455	8.05	1445-1455	0.371 0.408 0.431 -- -- 0.273 0.263 0.256 0.257 --
				-- --	3.50	1455	3.50	1455	0.406 ^(a) 0.239 ^(a)
I	-- --	4.55	1445	0.252 0.288 0.270 0.252 ^(b) 0.244 ^(b)	3.50	1455	8.05	1445-1455	0.359 0.272 0.274 -- --
				--	3.50	1455	3.50	1455	0.243 ^(a)

(a) Added for 2nd irradiation.

(b) Removed after 1st irradiation.

TABLE 2
THERMAL CONDUCTIVITY CHANGES FOR H-328 GRAPHITE
(PERPENDICULAR SPECIMENS)

Unirradiated K (cal/cm-sec-°C)	Fluence 1 $\times 10^{-21}$ (n/cm ²) (E > 0.18 MeV)	Temperature 1 (°C)	K (cal/cm-sec-°C)	Fluence 2 $\times 10^{-21}$ (n/cm ²) (E > 0.18 MeV)	Temperature 2 (°C)	Total Fluence $\times 10^{-21}$ (n/cm ²) (E > 0.18 MeV)	Temperature Range (°C)	K (cal/cm-sec-°C)
0.285 0.280	0.80	560	0.138 0.141 0.139 0.141 0.142 0.136 0.136 0.132 ^(a) 0.126 ^(a) --	0.60 0.60	605 605	1.40 0.60	560-605 605	0.097 0.133 0.121 0.128 0.114 0.134 0.134 -- -- 0.156 ^(b)
0.283 0.276 0.293 0.277	3.30	1040	0.184 0.239 0.187 0.188 0.188 0.189 0.160 0.190 --	2.10 2.10	1205 1205	5.40 2.10	1040-1205 1205	0.257 0.190 0.181 0.182 0.183 0.200 0.179 0.182 0.188 ^(b)
0.285 0.280 0.283 0.281 0.273	4.20	1360	0.175 0.177 0.199 0.186 0.176 0.192 0.196 0.186 ^(a) 0.182 ^(a) --	3.10 3.10	1360 1360	7.30 3.10	1360 1360	0.226 0.193 0.213 0.207 0.194 0.211 0.207 -- -- 0.192 ^(b)

(a) Removed after 1st irradiation.

(b) Added for 2nd irradiation.

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Fig. 1. Thermal conductivity versus fluence at 560° to 1455°C for H-327 needle-coke graphite: (a) parallel

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Fig. 1. Thermal conductivity versus fluence at 560° to 1455°C for H-327 needle-coke graphite: (b) perpendicular

LC95496

Fig. 2. Thermal conductivity versus fluence at 560° to 1455°C for H-328 graphite (perpendicular)

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