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RADIATION DAMAGE IN DIAMOND AND
SILICON CARBIDE, II

by

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CHEMISTRY DIVISION

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RADIATION DAMAGE IN DIAMOND AND SILICON CARBIDE, II*

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ABSTRACT

Further observations of changes in lattice constants and densities of diamond and silicon carbide on exposure in various nuclear reactors are given and discussed.

Studies of radiation damage in diamond and silicon carbide have been previously reported by W. Primak, L. H. Fuchs, and P. Day.¹ These studies have been continued.

EXPERIMENTAL

Samples

Diamond - X-ray lattice constant samples were prepared from 300-320 mesh white Bort diamond dust purchased through commercial channels. Samples for density determinations were chips about 1/2 mm. Several stones about 2 mm across were irradiated and one was used for a density determination.

α -Silicon Carbide - All of the samples were prepared from several samples of masses of transparent, light green crystals, some of which were platelets about 2 mm thick and 1 cm across furnished by Dr. G. R. Finlay of the Norton Company. The density samples were fragments obtained by crushing, and the x-ray samples were prepared by crushing these to a fine dust in Plattner steel mortars. Some of the x-ray data were obtained on samples crushed prior to irradiation, while other data were obtained on samples crushed after irradiation.

Irradiations

A number of irradiations were performed in Hanford water-cooled test holes (type B facilities). The samples were wrapped in aluminum foil, and the packages were placed in cylindrical aluminum containers of 1/16" wall thickness, approximately 1/2" o.d. and 6" long, whose ends were closed

* Part I issued as ANL-5250.

¹W. Primak, L. H. Fuchs, and P. Day, ANL-5250.

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with solid aluminum plugs about 1" long. These are referred to as single casings. In irradiations IW 40 and IW 41, the samples were placed in small holes which had been drilled into cylindrical graphite blocks whose diameter was 3/8". The latter were then placed in single casings. The irradiation data are given in Table I.

Table I
IRRADIATION DATA FOR HEW IRRADIATIONS

<u>Irradiation (Ref. No.)</u>	<u>Location</u>	<u>Exposure (Mwd/aT)</u>
IW 32	Bth-H	292
IW 40	Dth-DR	656
IW 41	Dth-DR	660
IW 50	Dth-DR	289
IW 51	Dth-DR	296
IW 52	Dth-DR	638
IW 53	Dth-DR	1060

One irradiation (IW 90) was performed in the EBR. The samples were sealed in a vitreous silica capsule, 0.345" diameter with 1-mm wall, containing helium at 0.9 atmosphere pressure. The samples were separated from each other with aluminum discs and silica wool. The silica capsule was placed in a thin-wall stainless steel capsule, 0.389" diameter by 0.015", and was crimped shut. The stainless steel capsule was put into a standard fuel container (no fuel in this tube) and sealed shut by welding. The samples were located in the outer rod circle of the core, position 217, about 3" below rod center. They were exposed for a period in which about 1745 Mwh was conveyed to NaK (the reactor coolant). The integrated neutron flux (all fast) was estimated to have been 1.1-1.2 (10^{21}) within a factor of 2. For 2% of the exposure, the temperature was between 70°C and 190°C. About half this occurred when 71% of the total nvt had been received and the rest when 98% of the total nvt had been received. There were 122 start-ups in the irradiation period and, assuming these required 1/2 hour each, this would give an additional 3-1/2 per cent of the total nvt at low temperature. The rest of the irradiation was conducted at about 270°C.

Two irradiations were performed in the MTR. In the first (IW 95) the samples were wrapped in aluminum foil and inserted into an aluminum can containing graphite samples. The can was in position L-45 at centerline

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for an accumulated thermal flux of about $2.5(10^{21})$. Graphite samples placed in this can were reported by A. Whatley to have been damaged the equivalent of an exposure of 5000 Mwd/aT (extrapolated) in a type B facility at HEW. In the second (IW 106) the samples were packed with aluminum chips in a perforated aluminum can which permitted process water to circulate in contact with the samples. The can was irradiated in position L-43 about 2" below centerline, surrounded by fuel on three sides for an estimated accumulated thermal flux of $2(10^{20})$.

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RADIATION DAMAGE RESULTS

Lattice Constants of Diamond - Diamond powder (300 mesh) was sealed into vitreous silica x-ray capillaries and some of these were irradiated. The lattice constants were determined using unfiltered copper radiation (to give additional lines in the back-reflection region) and a powder camera. The results are given in Table II.

Table II

LATTICE CONSTANTS OF DIAMOND

<u>Irradiation (Ref. No.)</u>	<u>Sample^a (Ref. No.)</u>	<u>Lattice Constant (kX units)</u>	<u>Changes (Per cent)</u>
Unirradiated	WP 14	3.5596	0
IW 50	WP 331	3.5837	0.68
IW 51	WP 335	3.5836	0.67
IW 52	WP 367	3.5928	0.91
IW 40	WP 322	3.5930	0.94
IW 41	WP 317	3.5935	0.95
IW 53	WP 390	3.6004	1.15
IW 95	WP 305	3.5925	0.92

^aThese reference numbers are not to be confused with a set of reference numbers used by T. J. Neubert which also bore the prefix WP.

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Lattice Constants of Silicon Carbide - Three results are reported here. The first was obtained on a fragment of silicon carbide which was crushed subsequent to irradiation. The remaining two were obtained on x-ray capillaries which were filled with crushed silicon carbide and then irradiated. The results are reported in Table III.

Table III

LATTICE CONSTANTS OF SILICON CARBIDE

<u>Irradiation</u> (Ref. No.)	<u>Sample</u> (Ref. No.)	<u>Lattice Constant</u> (kX units)		<u>Changes</u> (Per cent)	
		"a"	"c"	"a"	"c"
Unirradiated	WP 365	3.080	15.060		
Unirradiated	WP 25	3.079	15.065		
IW 32	WP 366	3.092	15.140	0.41	0.51
IW 40	WP 321	3.101	15.170	0.70	0.71
IW 41	WP 316	3.100	15.164	0.67	0.67
IW 95	WP 346	3.102	15.178	0.73	0.77
IW 90	WP 348	3.107	15.227	0.89	1.09

Density of Irradiated Diamond - The densities of diamond chips, 1/2 mm across, were determined by floating them in a thallium malonate-formate solution (held in a thermostat) whose density was determined with an Ostwald pycnometer. The results are given in Table IV.

Table IV

DENSITY OF DIAMOND CHIPS

<u>Irradiation</u> (Ref. No.)	<u>Sample</u> (Ref. No.)	<u>Density</u> (g/ml)	<u>Change</u> (Per cent)
Unirradiated	WP 440	3.5147	
IW 90	WP 450	3.4272	2.49
IW 106	WP 391	3.3910	3.52
	WP 370		

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Density of Irradiated Silicon Carbide - The densities of some silicon carbide chips were determined in a manner similar to that used for the diamond chips except that a carbon tetrachloride-methylene iodide solution was used. The densities of other chips were obtained by the method of Hutchison and Johnston² using a solution of methylene iodide and o-phenylene chloride. The results are given in Table V.

Table V

DENSITY OF SILICON CARBIDE CHIPS

<u>Irradiation</u> (Ref. No.)	<u>Sample</u> (Ref. No.)	<u>Density</u> (g/ml)	<u>Change</u> (Per cent)
Unirradiated	WP 441	3.2148	
IW 90	WP 447	3.1320	2.57
IW 95	WP 443 ^a	3.1323 ^a	2.47
IW 52	WP 418	3.148	2.08
IW 53	WP 425	3.1263	2.75
IW 32	WP 442	3.1797	1.09
IW 106	WP 387	3.0995	3.58

^aBlack SiC, density before irradiation 3.2117 g/ml.

DISCUSSION

The dilatation calculated from the lattice constant changes of diamond (Table II) irradiated at Hanford are plotted against exposure in Figure 1. For comparison, there is also plotted the data reported by Primak, Fuchs, and Day¹ for samples exposed in double casings. It is seen that the damage rates per unit exposure are somewhat greater for the more recent samples. The samples were exposed in different locations, but the locations did have a similar geometry. If it is assumed that the dosage per unit exposure is approximately the same for the two sets of samples, the more recent samples show a larger damage rate per neutron, an effect which was attributed¹ to the samples in the single casings having been at a lower temperature than the samples in the double casings.

²C. A. Hutchison and H. L. Johnston, J. Am. Chem. Soc. 62, 3165 (1940)

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FIGURE 1.

IRRADIATION DILATATION
OF DIAMOND.

○ Single Casings ● Double Casings

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Other evidence for the sensitivity of the dilatation of diamond to the temperature of irradiation is obtained by comparing the change in density observed for sample WP 450 (Table IV), irradiated for the most part at temperatures in excess of 200°C, with the changes in lattice constant reported in Table II for samples subjected to a much lower integrated damaging neutron flux at temperatures probably below 100°C. It is seen that the samples irradiated at the lower temperature for a shorter dosage experienced a greater dilatation.

The dilatation of silicon carbide (from the data of Tables III and IV) irradiated in type B facilities at Hanford is plotted against exposure in Figure 2a. The curve is extended using the presumed equivalent cold test hole exposure of the MTR irradiated sample in Figure 2b. It can be seen that the dilatation as a function of exposure is a curve which tends toward saturation. The equivalent cold test hole exposure of IW 106 is somewhat in doubt, but the damage rate has fallen so greatly at this exposure that altering its abscissa could hardly affect the curve. The reciprocal dilatation is plotted against reciprocal exposure in Figure 3. It is seen that the Neubert-Novick law nearly applies to the first stages of the irradiation dilatation. The deviation of the data from the Neubert-Novick law is in the opposite direction from that of diamond. The Neubert-Novick expression is the solution to the differential equation

$$dy/dx = (1 - \phi y)^n$$

for $n = 2$ where y is the fractional property change and x is a unit of dosage. For diamond $n > 2$, about 2.3 for the data given previously,¹ while for the silicon carbide data presented here $n < 1$. The effect in the final stages of irradiation might be termed a delayed saturation in the case of diamond and an accelerated saturation in the case of silicon carbide. This determines the form of the curve obtained by plotting the irradiation dilatation of silicon carbide against the irradiation dilatation of diamond as shown in Figure 4. From the position of the point obtained from the data of IW 90, it is seen that the irradiation dilatation rate of silicon carbide is less sensitive to temperature in this range of temperatures than the irradiation dilatation rate of diamond. The irradiation IW 95 was conducted for an exposure about 10 times that of IW 106 in an equivalent reactor position. The much smaller damage rate is attributed mainly to a higher irradiation temperature than for IW 106 with moderation and scattering by surrounding samples a contributing factor. It is seen that in this irradiation, presumably at a lower temperature than IW 90, the dilatation rate in silicon carbide and diamond appear to have been the same as in the Hanford-irradiated samples

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IRRADIATION DILATATION OF α -SiC.

FIGURE 3.
NEUBERT-NOVICK RELATION
FOR α -SiC.

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FIGURE 4.
COMPARISON OF THE IRRADIATION DILATATION
IN α -SiC WITH DIAMOND.

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I am indebted to Stanley Siegel for determining all the lattice constants reported here. Louis H. Fuchs packaged the samples for irradiation IW 32 and subsequently opened the irradiated slug. I am grateful to A. Whatley (HEW) for irradiating samples WP 346, and WP 443. The EBR irradiation was performed at the suggestion of W. H. Zinn. I am indebted to the members of the staff of the EBR project for having performed the irradiation.

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