Neutron Irradiation Damage in Graphite and Its Effects on Properties

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Abstract: Neutron irradiation is known to markedly influence the structure and properties of nuclear graphite. Here we review the mechanism of neutron damage and discuss its effect on the structure and properties of graphite. The results of recent irradiation experiments (HTN series capsules) on H-451 graphite are reviewed and contrasted with existing data. The graphite was irradiated in the High Flux Isotope Reactor at Oak Ridge National Laboratory (ORNL) to high dose at irradiation temperatures of 600 and 900°C. The peak doses attained were 3.8 x 10^{22} n/cm^2 [E>50 keV] and 2.2 x 10^{22} n/cm^2 [E>50 keV] at irradiation temperatures of 600 and 900°C, respectively. At these doses the graphite has passed through volume shrinkage turnaround into swelling and exceeded its original dimensions. Here, data for the dimensional, strength, and modulus changes are reported and discussed in terms of the structural changes that occur in the graphite at high neutron doses.

Keywords: A. Nuclear Graphite; D. Radiation Damage

1. INTRODUCTION

Radiation damage in graphite [1-5] occurs when energetic particles, such as fast neutrons, impinge on the crystal lattice and displace carbon atoms from their equilibrium positions – creating a lattice vacancy and an interstitial carbon atom. The displaced carbon atoms recoil through the lattice and produce other carbon atom displacements in a cascade effect. The cascade carbon atoms tend to be clustered in small groups of 5-10 atoms and it is generally satisfactory to treat the displacements as if they occur randomly. However, not all of the carbon atoms remain displaced. The displaced carbon atoms diffuse between the graphite layer planes in two dimensions and a high proportion of them will recombine with lattice vacancies. Others will coalesce to form linear molecules, which in turn may form the nucleus of a dislocation loop – essentially a new graphite plane. Interstitial clusters, on further irradiation, may be destroyed by impinging neutrons or energetic displaced carbon atoms (irradiation annealing). Adjacent lattice vacancies in the same graphite crystal basal plane are believed to collapse parallel to the basal plane, thereby forming sinks for other vacancies that are increasingly mobile above 600EC, and hence can no longer recombine and annihilate interstitials. This mechanism is illustrated in Fig. 1 below.

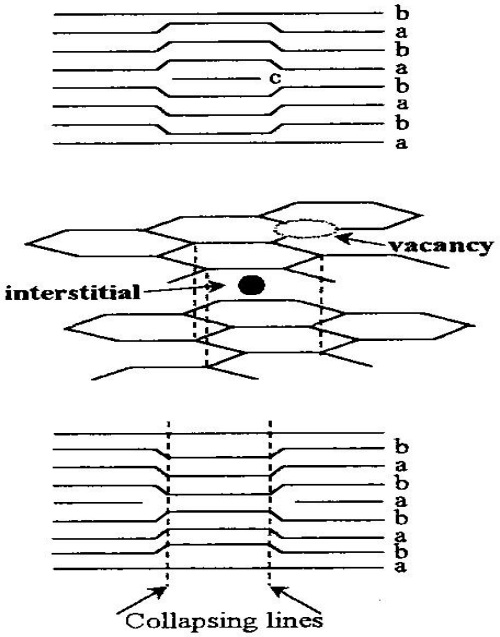

Figure 1. Displacement damage mechanism in graphite

The lattice strain that results from displacement damage causes significant structural and property changes in the graphite.

Graphite is used as the moderator and structural element in high-temperature gas cooled reactors such as the Gas Turbine Modular Helium Reactor (GT-MHR) and the Pebble Bed Modular Reactor (PBMR) [1]. Consequently, the effect of neutron irradiation on the properties of graphite is very important in the design and operation of graphite moderated nuclear reactors.

Here we report the results of neutron irradiation experiments on nuclear graphite grade H-451 to irradiated high dose in the High Flux Isotope Reactor (HFIR) at the Oak Ridge National Laboratory. The observed dimensional and property changes are reported and explained in terms of the mechanism of damage described above.

2. EXPERIMENTAL

2.1 *Material*

An extruded near-isotropic medium-grained nuclear graphite (grade H-451) was used for this work. The graphite was obtained from SGL Carbon from production lots made in the USA during the late 1970s. Table 1 reports typical properties data for H-451 and, for comparison, two other nuclear graphite grades used as moderators in high temperature reactors.

Table 1. Physical Properties of Several Nuclear Graphite Grade

PROPERTY	**GRAPHITE GRADE**		
	H-451	IM1-24	IG-110
Forming Method	E	M	I
Bulk Density (g/cm^3)	1.75	1.81	1.75
Elastic Modulus (GPa)	*11/9.6	11	10
Tensile Strength, MPa	*15/13	27.5	25
Bend Strength, MPa	*20/24	23	34
Compressive Strength, MPa	*60/60	70	71
Thermal Conductivity (25°C), W/m‧k	*150/135	131	*124/138
CTE, 10^{-6}/EC	*3.5/4.5	4.3	*4/3.6

E-extruded, M-molded, I-isostatic pressing; *∥/⊥ to forming axis

2.2 *Irradiation Conditions*

Small ring samples of H-451 graphite (≈ 12-mm outside diameter, 3-mm inside diameter, and 6-mm length) were irradiated in capsules in the target region of the HFIR. The peak neutron dose and design temperature is reported in Table 2 for each irradiation experiment. The accumulated neutron dose varies along the length of the target capsule and is symmetrical about the capsule midpoint, where the peak flux occurs. The flux ratio increases from 0.38 at the top and bottom of the capsule to 1 at the capsule midpoint.

Table 2. Capsule irradiation design temperature and peak neutron dose.

HFIR Target Capsule	**Design Irradiation Temperature, °C**	**Peak Neutron Dose (n/m^2) [E>50 keV]**
HTFC-1	600	0.24 x10^{26}
HTFC-2	600	0.73 x10^{26}
HTF-3	600	1.35x10^{26}
HTN-1	900	1.0x10^{26}
HTN-2	600	3.8 x10^{26}
HTN-3	900	2.2x10^{26}
HTK-7	600	3.8x10^{26}

The sample temperature in the HTN series of irradiation capsules was achieved by (i) nuclear heating of the graphite specimen and graphite center spine; (ii) selection of the capsule filler gas (Ne in the 600°C capsules, and Ar in the 900°C capsules); and (iii) sizing the annular gas gap between the graphite specimen and the capsule.

2.3 *Property Determinations*

Graphite specimen dimensions were measured using a bench top precision micrometer before and after irradiation. The elastic modulus of the samples was determined ultrasonically by measuring the velocity of sound through the graphite. The elastic modulus E, is given by

$$E=\rho v^2[(1+\mu)(1-2\mu)/(1-\mu)] \text{ Pa} \quad (1)$$

where ρ is the specimen bulk density (kg/m^3), v is the longitudinal velocity of sound (m/s), and μ is Poisson's ratio given by:

$$\mu=\{1-[2(v_s/v_l)^2]\}/\{2-[2(v_s/v_l)^2]\} \quad (2)$$

where v_s and v_l are the shear and longitudinal velocities (m/s) respectively.

Strength was determined by testing the ring specimens in compression. The "brittle ring strength" was calculated from:

$$\sigma = [6K_tP(D_2+D_1)]/[\pi h(D_2-D_1)^2] \quad (3)$$

where K_t is the stress intensity factor, equal to 1.25 (m^{-1})

for the geometry used here, P is the load at failure (N), D_2 is the outside diameter (m), D_1 is the inside diameter (m) and h is the ring thickness (m).

3. RESULTS AND DISCUSSIONS

3.1 *Dimensional Changes*

The irradiation-induced volume changes in H-451 graphite are reported in Fig. 2 below.

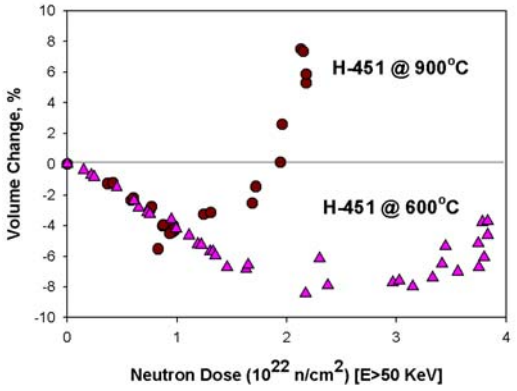

Figure 2. Irradiation-induced volume changes for H-451 graphite at two irradiation temperatures

The graphite initially shrinks but with increasing dose the rate of shrinkage falls to zero and the graphite begins to swell, eventually reaching and exceeding the sample's original volume and expanding into net growth. At an irradiation temperature of 900°C the dose at which the graphite returns to its original volume is approximately $2x10^{26}$ n/m^2 [E>50 keV]. At an irradiation temperature of 600°C the point at which H-451 returns to its original volume was at a dose > $4x10^{26}$ n/m^2 [E>50keV].

The data reported in Fig. 2 for irradiation-induced volume changes at 600°C derive from several separate irradiation capsules, namely HTFC-1, HTFC-2, HTF-3, HTN-2, and HTK-7 (Table 1), irradiated to differing peak doses over a time period spanning >15 years. Yet, the volume change data falls on a single relatively smooth curve, giving a high measure of confidence in the data. Similarly, the parallel and perpendicular dimensional change data sets at 600°C (Fig. 3) fall on single curves.

The dimensional change behavior of H-451 graphite at irradiation temperatures of 600 and 900°C are reported in Figs. 3 and 4, respectively. Significant anisotropy was observed in the dimensional change behavior in the parallel and perpendicular to the forming direction at both irradiation temperatures.

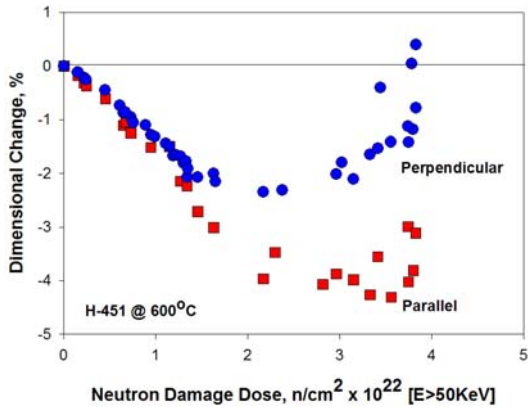

Figure 3. Dimensional change behavior of H-451 graphite at an irradiation temperature of 600°C

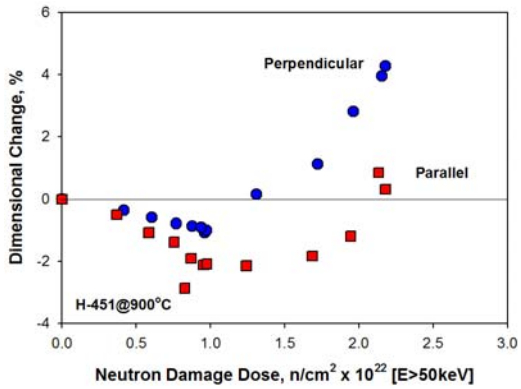

Figure 4. Dimensional change behavior of H-451 graphite at an irradiation temperature of 900°C

A principal result of the carbon atom displacements discussed in the introduction is crystalline dimensional change. Interstitial defects will cause crystallite growth perpendicular to the layer planes (c-axis direction), whereas coalescence of vacancies will cause a shrinkage parallel to the layer planes (a-axis direction). The damage mechanism and associated dimensional changes are illustrated in Fig. 5.

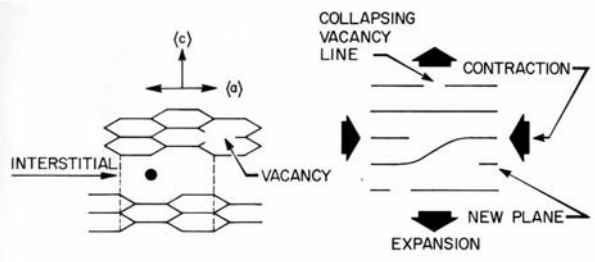

Figure 5. Neutron irradiation in graphite showing the induced crystal dimensional changes

Polygranular graphite exhibits a polycrystalline structure, usually with significant texture resulting from the method of forming during manufacture. Consequently, structural and dimensional changes in polygranular graphite are a function of the crystallites dimensional change and the graphite's texture. In polygranular graphite, thermal shrinkage cracks (formed during manufacture) that are preferentially aligned in the crystallographic a-direction initially accommodate the c-direction expansion, so mainly a-direction contraction is observed. Hence, the graphite undergoes net volume shrinkage. This behavior is seen for H-451 graphite (Figs. 2-4). With increasing neutron dose (displacements), the incompatibility of crystallite dimensional changes leads to the generation of new porosity oriented parallel to the basal planes, and the volume shrinkage rate falls, eventually reaching zero. The graphite then begins to swell at an increasing rate with increasing neutron dose because of the combined effect of c-axis growth and new pore generation. The graphite thus undergoes a volume change "turnaround" into net growth which continues until the generation of cracks and pores in the graphite, due to differential crystal strain, eventually causes total disintegration of the graphite.

H-451 graphite is an extruded material and thus the filler coke particles are preferentially aligned in the extrusion axis (parallel direction). Consequently, the crystallographic a-direction is preferentially aligned in the parallel direction and the a-direction shrinkage is more apparent in the parallel (to extrusion) direction, as indicated by the parallel direction dimensional change data in Figs. 3 and 4. The dimensional and volume changes are greater at an irradiation temperature of 600°C that at 900°C, i.e., both the maximum shrinkage and the turnaround dose is greater at an irradiation temperature of 600°C. This temperature effect can be attributed to the thermal closure of internal porosity aligned parallel to the a-direction that accommodates the c-direction swelling. At higher irradiation temperatures a greater fraction of this accommodating porosity is closed and thus the shrinkage is less at the point of turnaround.

The crystallographic and structural changes caused by neutron irradiation of graphite have significant effects on the material's mechanical properties. In the unirradiated condition, polygranular graphite behaves in a brittle fashion and fails at relatively small strains. The stress-strain curve is non-linear and the fracture process occurs by the formation of subcritical cracks, which coalesce to produce a critical flaw [6,7]. When graphite is irradiated the stress-strain curve becomes more linear, the strain to failure reduced, and the strength and elastic modulus increased. As shown in Fig. 6 for H-451, there is a rapid increase in strength attributed to dislocation pinning at irradiation-induced lattice defect sites. This effect is largely saturated at doses > 0.1×10^{26} n/m^2 [E>50 keV]. Above about 1×10^{26} n/m^2 [E>50 keV] a more gradual increase in strength occurs due to structural changes within the graphite. The high-dose strength behavior of graphite is controlled by pore generation occurring after volume "turnaround" when the graphite approaches and exceeds its original volume, i.e., net volume swelling. The data reported here shows that H-451 retains its strength at doses up to 4×10^{22} n/cm^2 [E>50 keV] at an irradiation temperature of 600°C.

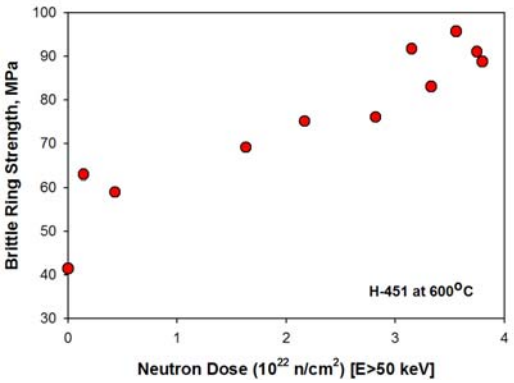

Figure 6. The variation of strength with neutron dose for H-451 graphite irradiated at 600°C

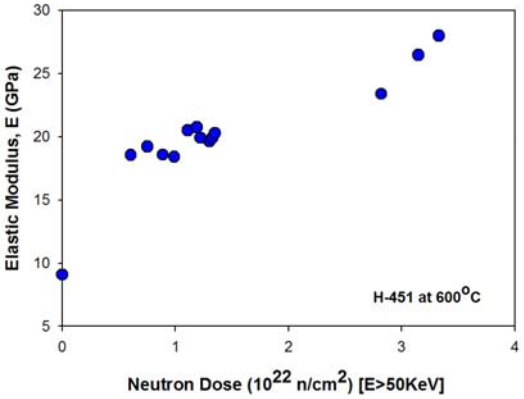

Figure 7. The variation of elastic modulus with neutron dose for H-451 graphite irradiated at 600°C

The strain behavior of polygranular graphite subjected to an externally applied load is largely controlled

by shear of the component crystallites. As with strength, irradiation-induced changes in elastic modulus are the combined results of in-crystallite effects, due to low dose dislocation pinning, and superimposed structural changes external to the crystallite. The modulus data in Fig. 6 reflects these effects – a rapid rise in modulus at low dose followed by a more gradual increase due to structural changes. Both the strength and modulus changes for H-451 graphite follow the expected trend with neutron dose [1]. It has been proposed that changes in the "structure factor" that relates crystallite "microscopic" properties to bulk or "macroscopic" changes are determined by a single parameter, X_T [8,9]. It is planned to extend the work on X_T using these new data reported here.

4. CONCLUSIONS

Grade H-451 graphite has been irradiated at temperatures of 600 and 900°C and data for the volume, dimensional, strength, and modulus changes reported. The new data at 600°C compares favorably with existing data. The data show that H-451 graphite could be utilized in a nuclear reactor application to doses as great as $4x10^{26}$ n/cm^2 [E>50 keV] at an irradiation temperature of 600°C, but to lesser doses at an irradiation temperature of 900°C. The irradiation behavior of H-451 graphite was as expected and was explained with accepted irradiation behavior models.

ACKNOWLEDGEMENTS

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