



Neutron irradiation effects on carbon based materials at 350°C and 800°C

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Abstract

An experimental study on the neutron irradiation induced thermal properties and dimensional changes of various carbon based materials has been carried using the High Flux Reactor (HFR) of Petten. The investigated materials include: DUNLOP concept 1 and concept 2, SEPCARB N112, SEPCARB N312B, SEPCARB NS11 and RGTi (91) graphite. The two irradiation temperatures are about 350°C and 800°C, and the neutron dose is about 0.3 dpa. This paper presents the experimental results on the neutron induced dimensional, specific heat capacity and thermal conductivity changes. © 2000 Elsevier Science B.V. All rights reserved.

1. Introduction

Carbon fiber composites (CFCs) are considered as an attractive choice for high heat flux components in existing and forthcoming tokamaks, because of their low atomic number leading to favorable plasma compatibility, and their excellent thermal shock resistance. However, carbon materials have some disadvantages such as high tritium uptake during plasma operation, high chemical sputtering, radiation enhanced sublimation (RES) at $T > 1000^\circ\text{C}$ and high rate of reaction with water and oxygen at $T > 1000^\circ\text{C}$. To improve the properties of carbon materials, the tritium inventory should be reduced, chemical erosion and RES have to be suppressed to increase the resistance to water/oxygen at elevated temperatures [1]. This can be done by using silicon doped CFC. Moreover, the neutron irradiation effects on the thermal conductivity of the CFC is a crucial problem, leading to significant decrease especially at low irradiation temperature ($<600^\circ\text{C}$) [2–7].

In the framework of the European fusion R&D programs, an extensive work on neutron effects is being undertaken. This paper describes the results of the in-

vestigation on the thermal conductivity and dimensional stability results of high conductivity CFCs, silicon doped CFC and titanium doped graphite irradiated in the neutron damage range of 0.30–0.35 displacement per atom of graphite (dpa g), and at low (335°C) and medium (775°C) irradiation temperatures.

2. Experimental

2.1. Materials

The investigated materials are four CFCs, one silicon doped CFC and one titanium doped graphite:

Concept 1. 3D CFC constituted of long fibers woven in the plane, with ex-pitch fibers (P120–P130) in x direction and PAN fibers in y direction; then there is a needling giving an orientation of fibers in the perpendicular direction (z direction). The volume fraction of the fibers is 30%. The densification is performed by three cycles of chemical vapor infiltration (CVI) of pyrocarbon.

Concept 2. The same material as Dunlop, concept 1 but with a fiber volume fraction of 38%. The densification is operated by three cycles of CVI of pyrocarbon.

N112. 3D CFC constituted by an ex-PAN NO-VOLTEX preform (fiber volume fraction about 30%)

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which is needed in the perpendicular direction (z). The densification is performed by chemical infiltration of pyrocarbon followed by a heat treatment at high temperature. The last phase of densification is treated by a pitch impregnation at 1000 bars/1000°C followed by a heat treatment at 2200°C.

N312B. 3D CFC constituted by a Novoltex preform, with P25 ex-pitch fibers in x direction and ex-PAN fibers in y direction. Then there is a needling which orientates a percentage of fibers in z direction. The volume fraction of the fibers is 35% (27% in x direction, 4% in y direction and z direction). Then there is a densification by chemical infiltration of pyrocarbon at 1000°C followed by a heat treatment at 2800°C. The last phase of densification is made by chemical infiltration of pyrocarbon at 1000°C followed by a pitch impregnation at 1000 bars/1000°C.

NS11. 3D CFC N11 type, which has undergone a final infiltration of liquid silicon leading partly to the formation of silicon carbide (10 at.% Si). The porosity of this material is very low (1%).

RGTi(91). This material has been provided by the D.V. Efremov Institute Team in St. Petersburg (Russia). This recrystallized graphite is obtained by adding an organic binder and 7 wt% of Ti into the charge which serve as baking activator and carbon graphitization catalyst during high-temperature thermomechanical treatment. The adding of titanium reduces the chemical sputtering of the graphite material [2].

The thermal properties of these materials before irradiation are given in Table 1.

2.2. Irradiation conditions

The irradiation experiments have been performed in the High Flux Reactor (HFR), Petten, The Netherlands. The details of the Paride D302-01 and Paride D302-02 irradiation conditions are given in Table 2. The Paride D302-01 and Paride D302-02 experiments were irradiated during two cycles in core position H2, and during one cycle in core position F8, respectively [8].

3. Results

3.1. Dimensional changes

Dimensional measurements have been carried out before and after irradiation using disks ($\varnothing = 12$ mm, $h = 5$ mm) and cylinders ($\varnothing = 5$ mm, $h = 15$ mm). Post-irradiation measurements of the 6 materials (concept 1, concept 2, N112, N312B, NS11, RGTi(91)) have been realized in a glove box using micrometry. The grades concept 1, concept 2, N312B and NS11 have been measured in the x , y and z directions, when the grades N112 and RGTi(91) have been measured in the parallel and in the perpendicular directions. Dimensional change ($\delta L/L_0$) results of the different materials are given in Table 3. Dimensional changes of carbon based materials irradiated at 775°C or 335°C in the range 0.31/0.35 dpa g are very low independent of the irradiation temperatures and the measured directions. Neutron irradiation in-

Table 1
Thermal properties of unirradiated materials

Materials		Thermal conductivity (W m ⁻¹ K ⁻¹)		Coefficient of average linear thermal expansion (20–800°C) (10 ⁻⁶ K ⁻¹)	Specific heat capacity (J K ⁻¹ kg ⁻¹)		Density (g/cm ³)
		25°C	800°C		25°C	800°C	
Concept 1	x dir.	431	199	-0.55	692	1791	1.89
	y dir.	102	53	0.74			
	z dir.	76	37	3.51			
Concept 2	x dir.	377	181	-0.38	705	1823	1.88
	y dir.	118	62	0.67			
	z dir.	76	41	2.75			
N112	(//) dir.	248	116	0.62	712	1821	1.99
	(⊥) dir.	213	94	0.55			
N312B	x dir.	303	140	-0.16	703	1793	1.92
	y dir.	117	53	0.97			
	z dir.	122	53	1.57			
NS11	x dir.	222	103	0.99	712	1671	2.13
	y dir.	213	94	1.25			
	z dir.	174	77	2.25			
RGTi (91)	(//) dir.	401	150	1.50	712	1793	2.21
	(⊥) dir.	132	45	8.69			

Table 2
Paride D302 irradiation conditions

Irradiation experiments	Paride D302-01	Paride D302-02
Cumulative time (EFPD)	49.64	23.75
Temperature (°C)	335	775
ϕ ($E > 0.1$ MeV) (10^{21} cm $^{-2}$)	0.301	0.372
Damage (dpa g)	0.31	0.35

duced dimensional changes (Δ) are presented by the following equations:

- for concept 1, concept 2, N312B and NS11,

$$\Delta = \left| \frac{\delta L}{L_0}(x \text{ dir.}) \right| + \left| \frac{\delta L}{L_0}(y \text{ dir.}) \right| + \left| \frac{\delta L}{L_0}(z \text{ dir.}) \right|;$$

- for N112 and RGTi(91),

$$\Delta = 2 \left| \frac{\delta L}{L_0}(\parallel) \right| + \left| \frac{\delta L}{L_0}(\perp) \right|.$$

If the Δ value is close to zero, the material exhibits a very good dimensional stability (Table 4).

At an irradiation temperature of 335°C, NS11 and N112 which were made of an ex-PAN Novoltex preform and which undergone a needling in the perpendicular direction, have the lowest Δ -values, hence the best dimensional stabilities. At an irradiation temperature of 775°C, NS11 and N312B have the lowest Δ -values. At both irradiation temperatures, NS11 shows the best dimensional stability and RGTi(91) exhibits the worse behaviour. Moreover, after irradiation at 775°C, N112 shows a shrinkage in both directions.

The N112 reference material has already been irradiated in the Macif irradiation, carried out in the Osiris reactor, Saclay, France within the temperature range 390–430°C and the neutron damage range 0.41–0.85 dpa g [6]. It has also been irradiated in the Ceram D217-17 irradiation, carried out in the HFR, Petten, The Netherlands within the irradiation temperature range 800–840°C and the neutron damage range 1.05–1.85 dpa g [9]. These irradiation temperatures are very similar to those of the present study. The dimensional changes of N112 as a function of neutron damage are shown in Fig. 1. It is seen, that the shrinkage in parallel and perpendicular directions increases with neutron damage. At a damage of 0.3 dpa g, N112 dimensional changes are almost zero for both irradiation temperatures.

3.2. Specific heat capacity

The specific heat capacities of the different materials have been measured before and after irradiation with a differential scanning calorimeter Setaram DS 111 G. For the samples irradiated at 335°C, the specific heat capacities have been measured from room temperature

to 335°C (Table 5); those irradiated at 775°C have been examined between room temperature and 800°C (Table 6). The specific heat capacity changes at 350°C ($(C_{pi} - C_{p0})/C_{p0}$)_{350°C} of various materials irradiated at 335°C/0.31 dpa g range between –0.9% and 2.3% and the specific heat capacity changes at 800°C ($(C_{pi} - C_{p0})/C_{p0}$)_{800°C} are between –3.4% and 2.6% at 775°C/0.35 dpa g. The uncertainty of the C_p measurements is about 2.5%; this indicates that there is no neutron induced change in the specific heat capacity. Generally, neutron irradiation induces a small increase in specific heat capacity of graphite. Graphite specific heat capacity increases up to 10% after irradiation at 30°C/1 dpa g [10] or after irradiation at 490°C/29 dpa g [11]. Previous studies show that the specific heat capacity at 800°C of N112 is increased by about 12% after irradiation at 1000°C/1.8 dpa g [7]. It is seen that neutron damage of 0.3 dpa g is likely too low to induce any change in the CFC specific heat capacity.

3.3. Thermal diffusivity

Thermal diffusivities before and after irradiation have been measured by the laser flash method using an apparatus located in a glove box [12]. The thermal diffusivity, D , of the sample is obtained with Parker's method. The samples are disks (diameter: 12 mm; thickness: 5 mm). The method consists in illuminating the front face of these disks with a short heat pulse. The thermal diffusivity D is deduced from the heat equation applied to the thermal transient of the rear face, called thermogram. The analysis of the thermograms according to Parker's method allows to solve the heat equation in 1D; and in the total absence of heat exchange between the sample and the furnace atmosphere, the following equation can be used:

$$D = \frac{\gamma L^2}{t_{1/2}},$$

where L is the thickness of the sample, $t_{1/2}$ is the time needed to reach 50% of the maximum temperature increase T_{\max} , and γ is 0.13878. In case that there are heat losses by the front and rear faces of the disk, γ is estimated by a method proceeding from the Cowan's method [13]. In order to avoid annealing effects the

Table 3
Dimensional changes of carbon-based materials after irradiation at 335°C and 775°C

$\delta L/L_0$ in % after irradiation	Concept 1			Concept 2			N112			N312B			NS11			RGTi(91)	
	x dir.	y dir.	z dir.	x dir.	y dir.	z dir.	(//)	(\perp)	(\perp)	x dir.	y dir.	z dir.	x dir.	y dir.	z dir.	(//)	(\perp)
335°C/0.31 dpa g	-0.02	0.01	-0.45	0.10	0.25	0.04	-0.08	0.05	0.05	-0.55	-0.02	-0.04	-0.07	0.06	0.12	0.30	0.18
775°C/0.35 dpa g	0.04	-0.02	0.12	0.02	0.16	0.27	-0.12	-0.04	-0.04	-0.07	-0.02	0.05	-0.07	-0.05	0.00	0.38	0.17

Table 4

Δ -values of carbon-based materials after irradiation at 335°C and 775°C

Materials	Δ -values in %	
	T_{irr} : 335°C 0.31 dpa g	T_{irr} : 775°C 0.35 dpa g
Concept 1	0.48	0.18
Concept 2	0.39	0.45
N112	0.21	0.28
N312B	0.61	0.14
NS11	0.25	0.12
RGTi(91)	0.78	0.93

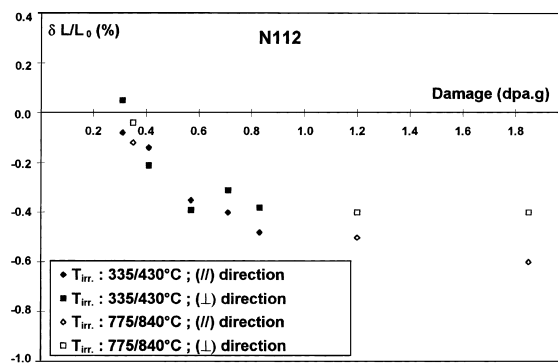


Fig. 1. N112 dimensional changes as a function of the neutron damage.

irradiated samples have been measured up to about their irradiation temperatures (350°C or 800°C).

3.4. Thermal conductivity

The evaluation of the thermal conductivity of irradiated CFCs is performed using the following equation :

$$K_i = D_i \rho_i c_{pi},$$

where D_i is the thermal diffusivity after irradiation, c_{pi} is the specific heat capacity after irradiation and ρ_i is the measured density after irradiation according to the damage level.

For the different materials, the evaluation of ρ_i versus temperature is performed using the average linear thermal expansion coefficient (20–800°C) of the material in the different directions [6,7].

The ratios of the thermal conductivities in x or ($//$) direction, after irradiation (K_i) to that of unirradiated values (K_0) are given in Table 7 for the materials irradiated at 335°C/0.31 dpa g and in Table 8 for those irradiated at 775°C/0.35 dpa g.

Table 5
Materials specific heat capacities before (C_{p0}) and after irradiation at 335°C/0.31 dpa g (C_{pi})

Temperature (°C)	Specific heat capacity (J K ⁻¹ kg ⁻¹)											
	Concept 1		Concept 2		N112		N312B		NS11		RGTi(91)	
	C_{p0}	C_{pi}	C_{p0}	C_{pi}	C_{p0}	C_{pi}	C_{p0}	C_{pi}	C_{p0}	C_{pi}	C_{p0}	C_{pi}
25	692	715	705	718	712	724	703	724	712	717	712	710
100	935	925	931	934	946	937	937	938	906	917	911	903
200	1177	1160	1172	1171	1187	1174	1175	1179	1112	1132	1139	1128
300	1352	1341	1354	1352	1366	1356	1350	1367	1268	1295	1318	1305
350	1423	1416	1428	1427	1438	1431	1421	1445	1331	1362	1391	1378

Table 6
Materials specific heat capacities before irradiation (C_{p0}) and after irradiation at 775°C/0.35 dpa g (C_{pi})

Temperature (°C)	Specific heat capacity (J K ⁻¹ kg ⁻¹)											
	Concept 1		Concept 2		N112		N312B		NS11		RGTi(91)	
	C_{p0}	C_{pi}	C_{p0}	C_{pi}	C_{p0}	C_{pi}	C_{p0}	C_{pi}	C_{p0}	C_{pi}	C_{p0}	C_{pi}
25	692	717	705	713	712	715	703	711	712	701	712	702
100	935	942	931	943	946	937	937	940	906	897	911	912
200	1177	1182	1172	1185	1187	1176	1175	1181	1112	1096	1139	1136
300	1352	1364	1354	1366	1366	1357	1350	1362	1268	1243	1318	1304
400	1484	1505	1493	1504	1502	1497	1483	1501	1387	1354	1457	1434
500	1587	1615	1603	1613	1608	1607	1586	1610	1481	1441	1568	1536
600	1669	1704	1691	1700	1693	1696	1669	1698	1557	1510	1657	1618
700	1736	1776	1763	1772	1763	1769	1737	1770	1619	1567	1731	1686
800	1791	1837	1823	1832	1821	1829	1793	1829	1671	1614	1793	1742

3.4.1. Comparison of the different materials

The thermal conductivity of the unirradiated materials in x or (l) direction, are ranked as follows:

$$\text{concept 1} > \text{concept 2} > \text{RGTi (91)} \approx \text{N312B} > \text{N112} > \text{NS11}.$$

After irradiation at 335°C, the sequences are (Fig. 2):

$$\text{concept 1} \approx \text{RGTi (91)} > \text{N312B} > \text{concept 2} > \text{N112} > \text{NS11}.$$

Concept 1 shows the best thermal conductivity before and after irradiation. Nevertheless concept 2 which has a higher thermal conductivity than N312B or RGTi(91) before irradiation, shows a lower one after irradiation at 335°C/0.31 dpa g. Before and after irradiation, NS11 appears to be the material with the lowest thermal conductivity. After irradiation at 775°C, the thermal conductivities of the materials in x or (l) direction are ranked as follows (Fig. 3):

$$\text{concept 1} \approx \text{concept 2} > \text{RGTi (91)} > \text{N312B} > \text{N112} > \text{NS11}.$$

It is interesting to note that it is almost the same ranking for the unirradiated materials. This means that the materials with the higher initial thermal conductivities keep their advantages after irradiation.

3.4.2. Effect of the irradiation temperature

The different carbon based materials have been irradiated at the same neutron damage (≈ 0.3 dpa g) and at two different irradiation temperatures (335°C and 775°C). For the different materials taken in x or (l) direction, it is interesting to compare their ratios $(K_i/K_0)_{T_{irr}}$ at these two irradiation temperatures (Table 9).

It is obvious that the thermal conductivity loss after irradiation at 335°C is significant. The normalized thermal conductivities at 335°C range between 0.26 (concept 1) and 0.35 (RGTi(91)). After irradiation at 775°C, the thermal conductivity loss is much lower. The normalized thermal conductivities at 775°C range between 0.62 (N112) and 0.70 (concept 1) except for NS11 which has a particularly lower value (0.51).

The NS11 material does not behave as we have expected. In previous work [7] we have shown that the ratio $(K_i/K_0)_{T_{irr}}$ is higher for CFCs with low initial thermal conductivity than for CFCs with higher initial

Table 7
Thermal conductivities in x or (//) direction before (K_0) and after irradiation (K_i) at 335°C/0.31 dpa g

Temperature (°C)	Concept 1		Concept 2		N112		N312B		NS11		RGT1(91)					
	K_0 (W m ⁻¹ K ⁻¹)	K_i (W m ⁻¹ K ⁻¹)	K_0 (W m ⁻¹ K ⁻¹)	K_i (W m ⁻¹ K ⁻¹)	K_0 (W m ⁻¹ K ⁻¹)	K_i (W m ⁻¹ K ⁻¹)	K_0 (W m ⁻¹ K ⁻¹)	K_i (W m ⁻¹ K ⁻¹)	K_0 (W m ⁻¹ K ⁻¹)	K_i (W m ⁻¹ K ⁻¹)	K_0 (W m ⁻¹ K ⁻¹)	K_i (W m ⁻¹ K ⁻¹)				
25	447	65	365	53	247	47	0.19	320	63	0.20	231	40	0.17	386	69	0.18
200	363	77	294	65	213	55	0.26	305	69	0.23	209	45	0.22	328	72	0.22
100	416	72	336	60	236	52	0.22	273	73	0.27	184	47	0.26	273	75	0.27
300	318	79	260	67	189	55	0.29	246	73	0.30	162	46	0.28	233	77	0.33
350	300	79	245	67	179	55	0.31	229	72	0.31	153	45	0.29	217	77	0.35

Table 8
Thermal conductivities in x or (//) direction before (K_0) and after irradiation (K_i) at 775°C/0.35 dpa g

Temperature (°C)	Concept 1		Concept 2		N112		N312BR		NS11		GT1(91)					
	K_0 (W m ⁻¹ K ⁻¹)	K_i (W m ⁻¹ K ⁻¹)	K_0 (W m ⁻¹ K ⁻¹)	K_i (W m ⁻¹ K ⁻¹)	K_0 (W m ⁻¹ K ⁻¹)	K_i (W m ⁻¹ K ⁻¹)	K_0 (W m ⁻¹ K ⁻¹)	K_i (W m ⁻¹ K ⁻¹)	K_0 (W m ⁻¹ K ⁻¹)	K_i (W m ⁻¹ K ⁻¹)	K_0 (W m ⁻¹ K ⁻¹)	K_i (W m ⁻¹ K ⁻¹)				
25	397	219	389	163	250	94	0.38	286	113	0.40	213	66	0.31	431	215	0.50
200	349	209	342	185	206	100	0.49	246	122	0.50	170	68	0.40	275	194	0.71
400	271	176	277	166	163	91	0.60	193	109	0.56	136	62	0.46	197	155	0.79
600	215	146	228	145	135	81	0.64	154	93	0.60	114	56	0.49	166	123	0.74
800	174	123	192	129	118	73	0.62	127	80	0.63	100	51	0.51	154	98	0.64

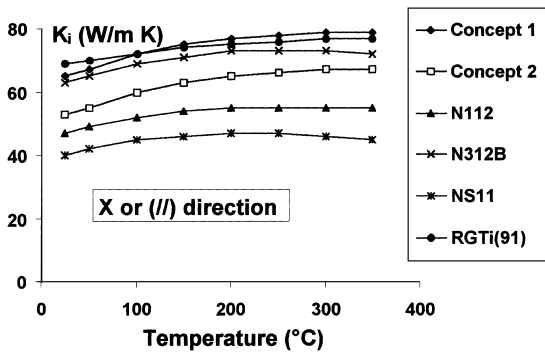


Fig. 2. Thermal conductivity of the materials as a function of temperature after irradiation at 335°C/0.31 dpa g.

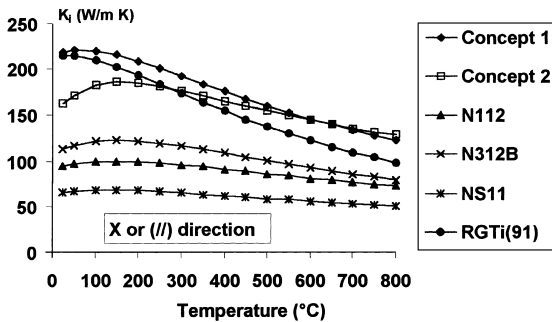


Fig. 3. Thermal conductivity of the materials as a function of the temperature after irradiation at 775°C/0.35 dpa g.

thermal conductivity after irradiation in the temperature range 600–1000°C.

Neutron irradiation induces a displacement of carbon atoms from their initial positions in the lattice to interstitial positions between two graphite planes. Thus, large dislocation loops or defect clusters are created especially at low irradiation temperatures (<450°C). The low irradiation temperature (335°C) avoids the annealing of these defects and slows down the propagation of the phonons along the graphite planes. Therefore, there is a beneficial effect of high irradiation temperature, allowing an annealing effect. The carbon atoms displaced from their lattice sites can return to their initial positions.

These results confirm that the normalized thermal conductivity temperature increases with irradiation temperature between 300°C and 1000°C [6].

3.4.3. Comparison of this study with previous work

N112 CFC has been used as a reference material because it has been irradiated in several irradiation experiments, particularly in the Macif irradiation at 390°C/0.52 dpa g and at 430°C/0.85 dpa g [14]. So it is interesting to compare the normalized thermal conductivity results of N112 irradiated in Paride D302 and in Macif, at irradiation temperatures ranging between 335°C and 430°C and neutron damages ranging from 0.31 to 0.85 dpa g.

We observe that the ratios (K_i/K_0) in both directions are almost the same after irradiation at 430°C/0.85 dpa g, 390°C/0.52 dpa g and 335°C/0.31 dpa g (Table 10). This means that the decrease in the irradiation temperature from 430°C to 335°C leads to the same effect on the thermal conductivity that the increase in neutron damage from 0.31 to 0.85 dpa g. The thermal conductivity loss under neutron irradiation depends strongly on the irradiation temperature and weakly on the neutron damage. The results have shown that the thermal conductivity of A05 CFC at 600°C is a logarithmic function of neutron damage, in a wide range of damage (from 9×10^{-4} to 1.8 dpa g) [7].

Birch and Brocklehurst [15] have shown that the fast neutron effects are allowed for the equivalent temperature concept which may be expressed as follows: if the two irradiations are performed at different levels of fast neutron fluxes ϕ_i and ϕ_j , to produce identical damage, the absolute irradiation temperatures T_i , T_j must be related by the expression:

$$\frac{1}{T_i} - \frac{1}{T_j} = \frac{k}{E} \ln \frac{\phi_j}{\phi_i},$$

where k is Boltzmann’s constant and E is the activation energy of the damage species assumed in practice to be represented by a unique value. If we apply this relation in our case, using N112 data (Table 11), of which the normalized thermal conductivity is constant, we can draw a curve,

$$\frac{1}{T_i} - \frac{1}{T_j} = f \left(\ln \frac{\phi_j}{\phi_i} \right).$$

Table 9

Normalized thermal conductivities in x or (//) direction at the irradiation temperature ($(K_i/K_0)_{T_{irr}}$) as a function of the irradiation temperature (T_{irr})

Irradiation temperature (°C)	$((K_i/K_0)_{T_{irr}})$					
	Concept 1	Concept 2	N112	N312B	NS11	RGTi(91)
335	0.26	0.27	0.31	0.31	0.29	0.35
775	0.70	0.67	0.62	0.63	0.51	0.65

Table 10

N112 normalized thermal conductivities (K_i/K_0) in ($//$) and (\perp) directions after irradiation at different conditions

$T_{\text{measur.}}$ (°C)	Normalized thermal conductivity(K_i/K_0)					
	$//$ direction			\perp direction		
	$T_{\text{irr.}}$: 335°C 0.31 dpa g	$T_{\text{irr.}}$: 390°C 0.52 dpa g	$T_{\text{irr.}}$: 430°C 0.85 dpa g	$T_{\text{irr.}}$: 335°C 0.31 dpa g	$T_{\text{irr.}}$: 390°C 0.52 dpa g	$T_{\text{irr.}}$: 430°C 0.85 dpa g
100	0.22	0.22	0.18	0.23	0.24	0.22
350	0.31	0.34	0.31	0.32	0.36	0.35

Table 11

N112 irradiation conditions

Irradiation experiments	Paride D302	Macif	Macif
Reactor	HFR	Osiris	Osiris
Irradiation temperature (°C)	335	390	430
Damage (dpa g)	0.31	0.52	0.85
Cumulative time (EFPD)	49.6	63.3	63.3
Fluence ($E > 0.1$ MeV) (10^{21} cm $^{-2}$)	0.30	0.69	1.12
Flux ($E > 0.1$ MeV) (10^{14} cm $^{-2}$ s $^{-1}$)	0.62	1.26	2.05

(Fig. 4). So, using a linear fit, it is possible to determine the ratio k/E , therefore the activation energy of the damage species is 0.45 eV.

Snead and Burchell [16] have shown that the thermal conductivity of graphite-based materials can be written as a summation of the thermal resistance due to the scattering mechanisms:

$$K(x, T) = \beta(x) \left(\frac{1}{K_u} + \frac{1}{K_{GB}} + \frac{1}{K_i} \right)^{-1},$$

where $\beta(x)$ is a coefficient depending on the orientation with respect to the basal plane. $\beta(x)$ is constant with temperature. $1/K_u$ defines the phonon–phonon scatter-

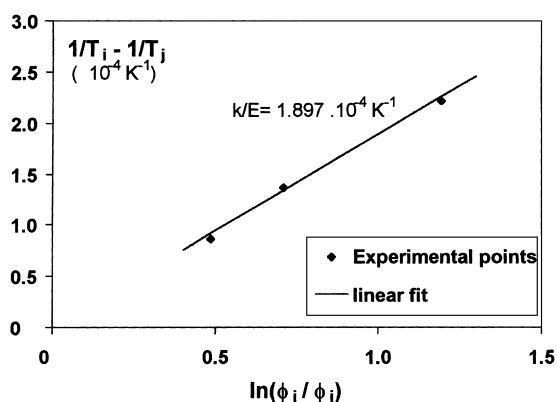


Fig. 4. N112 irradiation temperature and neutron flux equivalence for the irradiation conditions listed in Table 11.

ing effect and dominates at higher temperatures. $1/K_{GB}$ defines the grain boundary phonon scattering which dominates at low temperatures and becomes insignificant above a few hundred degrees Celsius. $1/K_i$ is the thermal resistance due to phonon scattering by defects.

Neutron irradiation induces various types of stable defects depending on the irradiation temperature. Using many references on the thermal conductivity taken at the irradiation temperature of graphites and CFCs, Snead and Burchell have evolved an algorithm which gives the normalized thermal conductivity at the irradiation temperature as a function of the irradiation temperature and of the neutron damage in dpa, T in °C:

$$\left(\frac{K_i}{K_0} \right)_{T_{\text{irr}}} = -M(T_{\text{irr}}) \log(\text{dpa}) + S(T_{\text{irr}}),$$

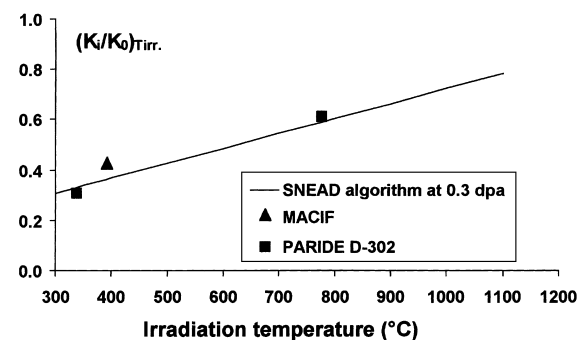


Fig. 5. N112 ($//$) normalized thermal conductivity at 0.3 dpa g as a function of the irradiation temperature.

where

$$M(T_{\text{irr}}) = 0.25 - 0.00017 \cdot T_{\text{irr}}, \quad S(T_{\text{irr}}) = 0.000683 \cdot T_{\text{irr}}.$$

This algorithm is accurate within the temperature range 300–1200°C. The comparison of the Snead algorithm with the Paride D302 and Macif [14] results for N112 (//) in the damage range of about 0.3 dpa g is given in Fig. 5. The normalized thermal conductivities of N112 (//) irradiated at 335°C, 775°C (Paride D302 results) and 385°C (Macif results) show a good agreement between experimental and calculated values.

4. Conclusion

In the present study, an experimental investigation of neutron irradiation effects on the dimensional, heat capacity and thermal conductivity changes of carbon based materials has been performed. The following conclusions can be drawn:

1. In the damage range of 0.3 dpa g and at 335°C or 775°C, the dimensional changes of the studied CFCs, silicon doped CFC and RGTi(91) graphite are very low. Moreover, it appears that NS11 silicon doped CFC presents the best dimensional stability at both irradiation temperatures.
2. A neutron damage of 0.3 dpa g is likely too weak, to induce any change in the CFC specific heat capacity.
3. If the studied materials are ranked with their thermal conductivities, there is almost the same ranking before and after irradiation at 775°C. However, concept 2 which has a higher K_0 than N312B or RGTi(91) before irradiation shows a lower thermal conductivity after irradiation at 335°C/0.31 dpa g.
4. After irradiation at low temperature ($\leq 335^\circ\text{C}$), the thermal conductivity loss is significant for all the studied materials ($0.26 < (K_i/K_0)_{350^\circ\text{C}} < 0.35$). The normalized thermal conductivities at 775°C range between 0.62 (N112) and 0.70 (concept 1) except for NS11 which shows the lowest value ($(K_i/K_0)_{775^\circ\text{C}} = 0.51$). As it was expected these results

confirm that the ratio $(K_i/K_0)_{T_{\text{irr}}}$ increases with irradiation temperature between 350°C and 1000°C.

5. The thermal conductivity loss depends strongly on the irradiation temperature and weakly on the neutron damage. It has been established that irradiation experiments at 335°C/0.31 dpa g, 390°C/0.52 dpa g. and 430°C/0.85 dpa g lead to the same degradation in the thermal conductivity. The calculated activation energy of the damage species is about 0.45 eV.
6. The results of this study are in good agreement with the proposed algorithm from Snead et al. [16].

References

- [1] C.H. Wu, C. Alessandrini, R. Moormann, M. Rubel, B.M.U. Scherzer, J. Nucl. Mater. 220–222 (1995) 860.
- [2] T.A. Burtseva, O.K. Chugunov, E.F. Dovguchits, V.L. Komarov, I.V. Mazul, A.A. Mitrofansky, M.I. Persin, Y.G. Prokofiev, V.A. Sokolov, E.I. Trofimchuk, L.P. Zav'jal'sky, J. Nucl. Mater. 191–194 (1992) 309.
- [3] T. Maruyama, M. Harayama, J. Nucl. Mater. 195 (1992) 44.
- [4] L.L. Snead, T.D. Burchell, J. Nucl. Mater. 224 (1995) 222.
- [5] T.D. Burchell, Phys. Scr. T 64 (1996) 17.
- [6] J.P. Bonal, C.H. Wu, J. Nucl. Mater. 228 (1996) 155.
- [7] J.P. Bonal, C.H. Wu, J. Nucl. Mater. 230 (1996) 271.
- [8] R. Conrad, R. May, Technical Memorandum HFR 95/4242 (1995).
- [9] J.P. Bonal, D. Moulinier, Report DMT/94-262, SETIC/LEMA/94_054, 1994.
- [10] R.E. Nightingale, Nuclear Graphite, Academic Press, New York, 1962, pp. 309–310.
- [11] C.H. Wu, J.P. Bonal, B. Kryger, J. Nucl. Mater. 208 (1994) 1.
- [12] J.P. Bonal, C.H. Wu, Phys. Scr. T 64 (1996) 26.
- [13] R.D. Cowan, J. Appl. Phys. 34–4 (1965) 926.
- [14] J.P. Bonal, D. Moulinier, Report DMT/95-146, SEMI/LEMA/95-067, 1995.
- [15] M. Birch, J.E. Brocklehurst, ND-R-1434 (S), UKAEA, Springfields Nuclear Power Development Laboratories, 1986.
- [16] L.L. Snead, T.D. Burchell, in: 22nd Biennial Conference on Carbon, San Diego, 16–21 July 1995, extended abstracts, 1995, pp. 774–775.