PROGRESS IN THE IRRADIATION OF CARBON-CARBON COMPOSITES

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Introduction

We have previously described investigations of carbon-carbon composites exposed to fast neutrons in the HERALD 5MW, light water moderated reactor (1,2). The property improvements obtained, the structural modifications and the potential use of these materials in nuclear energy generation and aerospace warranted further work.

Experimental

Carbon-carbon composites of thickness varying from 1 mm to 10 mm were prepared using PAN based HT and HM fibres and rayon based cloth. The composite structures were unidirectionally reinforced (UNI), 0°-90° cross-plied and all cloth. Densification was by chemical vapour deposition (CVD) or resin re-impregnation (RIP). Using two different fibres, two structures and two densification routes yielded eight different composites under evaluation plus one woven cloth composite. The properties measured are dimensional stability, longitudinal and transverse flexural strength, modulus, shear strength and impact strength after exposure to fast neutron doses varying from 10^{17} to 10^{20} n cm^{-2}.

Experiments have been done to study the effects of exposure in air and in inert atmosphere. The effect of temperature is also being studied. So far only moderate temperatures have been used eg 50°C and 250°C but these cover a range where interesting effects are observed using other graphites.

One of the problems in this work is the activation of impurities by thermal and epi-thermal neutrons causing handling difficulties during post exposure evaluation. A rig is under design which will incorporate boron carbide and cadmium shielding to reduce the residual activity. The use of boron carbide to provide γ-heating and helium flow for cooling also assists in achieving different exposure temperatures. Observations using HT fibre exposed to 10^{19} n cm^{-2} with and without shielding, showed that for boron carbide the activity after 100 days removal from the reactor was reduced from 3.8×10^{-6} to 4.5×10^{-7} curies/g/10^{19} n cm^{-2} and for cadmium was reduced from 7.7×10^{-7} to 1.6×10^{-7} curies/g/10^{19} n cm^{-2}, reductions of 8.4 : 1 and 4.8 : 1 respectively.

Results

Figure 1 shows the effect of exposure in air at 150°-250°C to fast neutron doses up to 1.5×10^{20} n cm^{-2} on the flexural strength and modulus of UNI-HT-CVD composites. Both properties increased steadily up to approximately 5×10^{19} n cm^{-2} then both fell abruptly such that at 1.5×10^{20} n cm^{-2} their values were less than the unirradiated values. Figure 2 shows some structural data obtained by neutron diffraction on the same material after similar exposures. The apparent d_{002} increased steadily and the apparent L_c decreased up to 5×10^{19} n cm^{-2}, then the rate of change increased sharply. Under the conditions of this experiment an exposure of approximately 10^{20} n cm^{-2} seems to be a threshold for the UNI-HT-CVD material, below which, property improvements occur and above which the high concentration of interstitial clusters disrupt the structure bringing severe reduction in mechanical properties.

Table 1 gives the dimensional changes with exposure for the nine composites under evaluation. In almost every case where change occurred it was a shrinkage. The one small length increase is thought to be due to measurement error. Measurements were made to an accuracy of 0.0005 in. but changes of 0.001 in. or less have been recorded as zero. The shrinkage in length of UNI composites, in length and breadth of cross-plied composites and no change in depth suggest that the fibres are changed in length but not in diameter. The structural evidence in Figure 2 indicated that at exposures around 10^{20} n cm^{-2}, there was large expansion in the c-axis direction which would be expected to manifest itself as an increase in diameter. The absence of dimensional change perpendicular to the fibres suggests that the c-axis expansion may be accommodated in cylindrical pores or Mrozowski cracks in the fibres themselves or within the cylindrical matrix sheaths surrounding the fibres. The former effect would lead to changes in fibre properties, the latter to changes in composite properties and may account for the increased shear strength and reduced impact strength previously reported.

The HT fibre length appears to shrink more that the HM fibre length; at an exposure of 10^{20} n cm^{-2} almost twice as much. Whereas the UNI-HM and 0°-90°-HT composites appear not to shrink at 10^{18} n cm^{-2}, the 0°-90°-HM and low modulus cloth composites shrink slightly. This may be due to the lower shear strength of the cross-plied HM and woven cloth composites or increased crystallinity.

Table 1 also shows that the RIP composites shrink slightly less than the CVD composites. There are two possible reasons for this; the first is that the pyrocarbon deposited by the CVD process has been shown to be oriented preferentially with its a-axis parallel to the fibres and may be shrinking in a similar manner. The glassy carbon from the resin is not well oriented. The second reason is that the RIP composites are generally of slightly higher density (1.6 cf 1.5 g cm^{-3}) than the CVD composites and also have higher shear strengths (30 cf 20 MN m^{-2}). Thus they resist the tendency for the fibres to shrink.

Further details of the mechanical properties of the composites after exposure will be given at the Conference.

References

(1) E J Walker, L F Pain and P B Roscoe, 12th Bienn Conf on Carbon, Pittsburgh, 1975- Paper CE-1.

(2) E J Walker, L F Pain and P B Roscoe, 2nd Int Carbon Conf, Baden-Baden, 1976 - Paper P5.2

TABLE 1

Dimensional Changes

Material	Exposure ($n\ cm^{-2}$)	% Change in: Length	Breadth	Depth
UNI-HT-RIP	10^{18}	0	0	0
	10^{19}	-0.11	0	0
	10^{20}	-0.68	0	0
UNI-HT-CVD	10^{18}	-0.10	0	0
	10^{19}	-0.28	0	0
	10^{20}	-0.55	0	0
UNI-HM-RIP	10^{18}	0	0	0
	10^{19}	+0.11	0	0
	10^{20}	-0.27	0	0
UNI-HM-CVD	10^{18}	0	0	0
	10^{19}	-0.12	0	0
	10^{20}	-0.40	0	0
0/90-HT-RIP	10^{18}	0	0	0
	10^{19}	-0.26	0	0
	10^{20}	-0.68	-1.01	0
0/90-HT-CVD	10^{18}	0	0	0
	10^{19}	-0.20	0	0
	10^{20}	-0.79	-0.81	0
0/90-HM-RIP	10^{18}	-0.13	-0.18	0
	10^{19}	-0.14	-0.27	0
	10^{20}	-0.40	-0.54	0
0/90-HM-CVD	10^{18}	-0.17	-0.18	0
	10^{19}	-0.15	-0.18	0
	10^{20}	-0.49	-0.68	0
Low modulus graphite cloth - CVD	10^{18}	-0.14	-	0
	10^{19}	-0.19	-	0
	10^{20}	-0.53	-	0

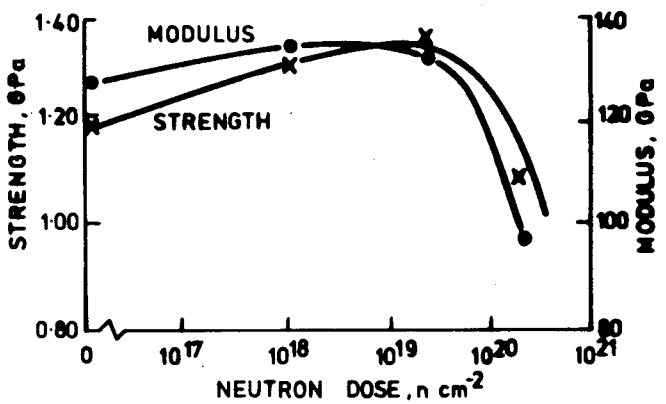

Figure 1 Flexural Strength and Modulus vs. Exposure. UNI-HT-CVD

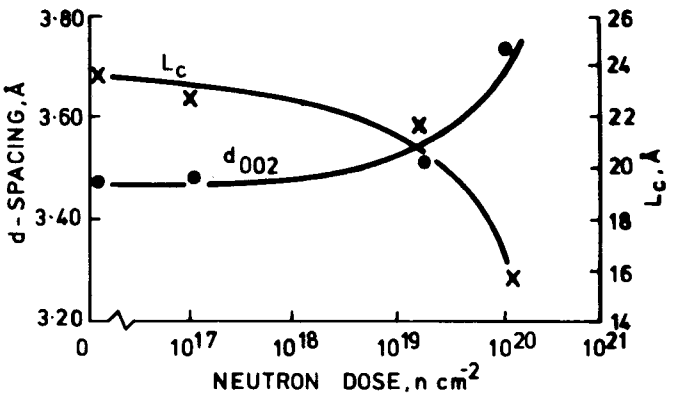

Figure 2 Apparent d-spacing and crystallite size vs. Exposure. UNI-HT-CVD