MODIFICATION OF THE FIBRE-MATRIX BONDING IN A BRITTLE CARBON-CARBON COMPOSITE BY CONTROLLED OXIDATION

Catherine Ahearn and Brian Rand
School of Materials, University of Leeds
Leeds, LS2 9JT, UK.

1. Introduction

Oxidation degrades the properties of carbon materials, including carbon-carbon composites[1,2,3,4]. C/C composites are brittle materials whose mechanical properties are controlled by the nature of the fibre-matrix interface. Evans *et al* [5] have classified the behaviour of such materials into three general classes. In type I the interfacial bonding is weak allowing debonding under stress. In type II, the sliding friction is higher resulting in a single matrix crack but with pull-out effects. However, if the interface is strongly bonded catastrophic type III failure occurs. C/C composites show all these types of failure. It is shown that oxidation of C/C composites can take place selectively at the fibre-matrix interface. However, the possibility of using controlled oxidation as a means of modifying the fibre-matrix interfacial characteristics to better utilise fibre properties by promoting multiple matrix cracking appears not to have been examined hitherto. The work here was carried out to test this hypothesis, using a C/C composite which initially showed brittle fracture behaviour characteristic of very strong interfacial bonding.

2. Experimental.

Uni-directional, pitch-based-matrix composites were fabricated from PAN-based Courtaulds fibres impregnated with Ashland Aerocarb 80 pitch from a solution in THF, following the route developed by Appleyard [6]. Oxidation of the carbonised composites was carried out in air at 420 °C and 730 °C to weight losses in the range 0.1 to 3.5% . The transverse structures of the material before and after oxidation were studied using scanning electron microscopy. The mechanical properties of the un-oxidised and oxidised composites were determined using three-point flexural tests [7] on specimens of span-to-depth ratio in excess of 40:1.

3. Results and discussion.

3.1. Microstructural observations.

The samples oxidised at the lower temperature (420 °C) suffered material loss at the fibre-matrix interfaces both at the outer regions of the composites and the areas close to internal pores and cracks. Oxidation at the higher temperature (730 °C) occurred at a more rapid rate. The porosity here appears to be limited to the outer regions of the composite only. The damaged parts form V-shaped areas within which a total separation of fibres and matrix has occurred.

3.2. Fracture behaviour.

The un-oxidised, carbonised material failed in a totally brittle, catastrophic manner exhibiting the class III type behaviour characteristic of these materials after carbonisation. The strength and modulus values of the composite were 216 MPa and 124 GPa respectively. The material failed at a strain of 0.19%. Only 9% of the fibre strength was utilised whereas 84% of the fibre modulus was achieved.

3.2.1. Low temperature oxidation.

The changes in the mechanical properties of the oxidised composites are shown as a function of weight loss in figure 1 while figure 2 shows a schematic illustration of the load extension curves generated. Class III type behaviour continued after oxidation until a weight loss of 0.4% was attained, although increases in both flexural strength and strain-to-failure occurred. The material oxidised to 0.4% weight indicated the development of class II type behaviour. After 1% weight loss the fracture behaviour changed towards class I type. The load-extension curve was linear up to a strain of 0.17% at which point a deviation from linearity occurred. Failure of the fibres occurred at a stress of 337 MPa, an increase in strength of 56% and a utilisation of fibre strength of 16%, almost double that of the un-oxidised material. Failure of the composite was accompanied by a gradual reduction in the load. Class I behaviour continued to dominate although the increase in strength reached a maximum of 85% of the un-oxidised composite at a weight loss of 1.4%. After a weight loss of 2.2% the strength of the material had declined to be virtually equal to that of the un-oxidised composite. However the failure mode remained as a class I . Further oxidative weight losses were detrimental to the ultimate composite strength but served to prevent catastrophic failure as has been observed by other workers [4].

3.2.2. High temperature oxidation.

In the samples heat treated at 730 °C, strength decreased with increasing weight loss. The modulus of the materials also decreased with increasing amounts of oxidation by a maximum of 26% after 3.5% weight loss. Failure strains, however, varied very little.The materials continued to show class III type failure behaviour after oxidation indicating that weakening of the fibre-

matrix interfacial bond was insufficient to allow debonding and a change in the mode of fracture.

3.3. Discussion.

At low temperature it is clear that the reactant gas has been able to react preferentially at the fibre-matrix interfaces to create porosity at these sites so changing the nature of the bond to allow debonding and fibre sliding. Increases in composite strength occur as a result of greater utilisation of fibre strength and the weaker bond prevents brittle failure. It appears that the fibre-matrix debond energy is progressively reduced by oxidation up to a burn off of 0.4% since there is a change in the fracture behaviour from class III to II.

In the samples oxidised at the higher temperature, the localisation of the porosity at the outer surfaces indicates that the oxidising gas was rapidly consumed as the reaction advanced into the specimen, giving rise to the V-shaped features. This diffusion controlled reaction tends to restrict the damage to regions close to the surface of the composite. Extensive weakening of the interfaces throughout the body of the composite does not take place allowing easy crack propagation across the strongly bonded interface and the retention of brittle, catastrophic failure. The localised areas of damage at the outer regions of the test piece probably contributed significantly to the reduction in strength. The decrease in modulus is attributed to the greater porosity at the exterior of the oxidised samples.
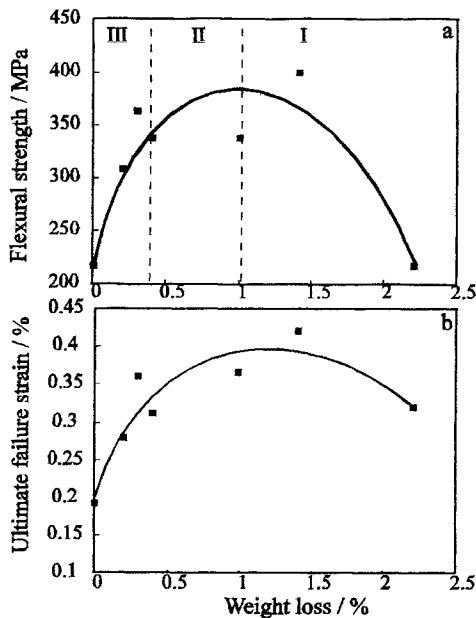
4. Conclusions.

The beneficial effects of low temperature oxidation described here have not been reported by any other workers. The results underline the importance of the fibre-matrix interface in controlling composite fracture behaviour and show that even small weight losses can have a dramatic effect. Of interest is whether the observed increases in strength are limited to this particular combination of fibres and matrix or whether other strongly bonded interfaces show similar results. It may be possible to modify the fibre-matrix properties by immersing the components in oxidising acids.

REFERENCES.
1. J.X. Zhao, R.C. Bradt and P.L. Walker, Carbon, 23, 9, (1985).
2. J.L. Wood, R.C. Bradt and P.L. Walker, Carbon, 18, 179, (1980).
3. I.M. Pickup, B. McEnaney and R.G. Cook, Carbon, 24, 535, (1986).
4. P. Crocker and B. McEnaney, Carbon, 29, 881, (1991).
5. A.G. Evans, F.W. Zok and J. Davis, Composites Science and Technology, 42, 3, (1991).
6. S.P. Appleyard, Ph.D. Thesis, University of Leeds, (1993).
7. ASTM D790M-84 in ASTM Standards and Literature References for Composite Materials, Ed. Committee D-30, 1st Edition, ASTM, Philadelphia, 360, (1987).

Figure 1. Change in mechanical properties of oxidised composites as a function of weight loss.
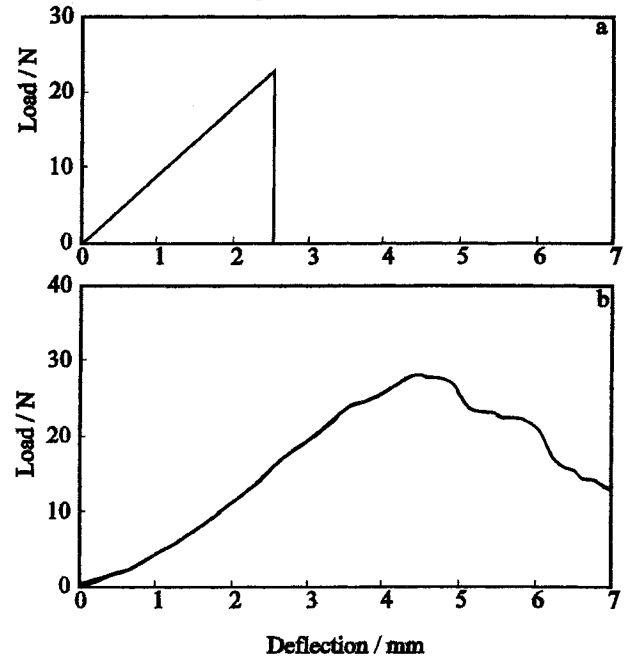
Figure 2. Schematic illustration of the load-extension curves of (a) un-oxidised and (b) oxidised to 1.4% weight loss.