JOURNAL OF MATERIALS SCIENCE 6 (1971) 419–426

Some Mechanical Property and Fracture Studies of Carbon Fibre/Nickel Composites

D. M. BRADDICK, P. W. JACKSON*, P. J. WALKER
Rolls-Royce Ltd, Old Hall, Littleover, Derby, UK

A carbon fibre reinforced nickel composite has been fabricated, and some of the mechanical properties investigated. The composite contains some misaligned and broken fibres, and a poor bond exists between the carbon fibre and the nickel. The mechanical properties are, to a very large extent, influenced by these factors. The oxidation resistance was found to be poor, and therefore a serious limitation for high temperature use.

1. Introduction

The advantages to be gained from fibre reinforcement are well documented [1, 2]. However, while the potential of such materials is apparent, the practical difficulties encountered in their development have mainly limited the successful applications to the reinforcement of resin matrices. This is especially true where carbon fibres have been considered as the reinforcing filaments, and the reasons are largely two-fold.
(1) The incompatibility of carbon fibres with many metals at elevated temperatures, either by reaction to form a metal carbide or by recrystallisation [3].
(2) The inherent difficulties when fabricating a composite from a multifilament tow of fibres.
In addition there is also the relatively poor oxidation resistance of carbon at temperatures above 400°C, and the uncertainty of the degree of bonding of the metal to the fibres.

A systematic investigation of some of the mechanical and physical properties of a composite consisting of graphitised fibres in nickel has been performed by R. V. Sara [4] and by D. E. Niesz and C. W. Kistler Jr [5, 6]. In both instances the work was directed towards the development of optimum properties, and included the effects of varying the fabrication parameters. The aim of the present investigation was to study some of the mechanical properties of a graphite fibre reinforced nickel composite, with special emphasis placed on the evaluation of failure mechanisms by both optical and scanning electron microscopy.

2. Composite Fabrication

Throughout this investigation Courtaulds surface treated graphitised fibre was used having UTS = 2 GN/m²,† E = 345 GN/m² and being in the form of a continuous 10^4 filament tow.

A coating technique was developed using an electrodeposition process, which was capable of continuously coating a 10^4 filament tow with up to 50 vol % nickel [7] and which provided each of the fibres within the tow with a uniform covering of nickel (fig. 1).

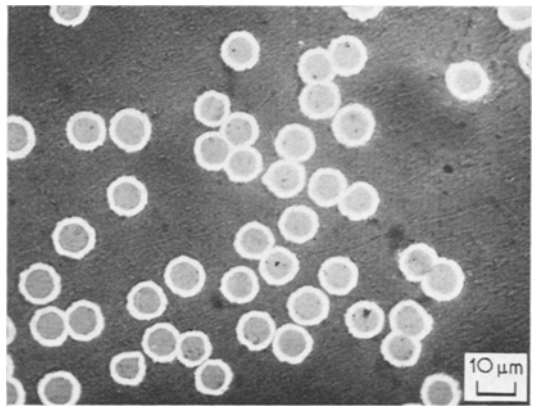


Figure 1 Cross-section of part of a fibre tow coated with nickel.

Both electrodeposition and electroless deposition have been successfully used in the past [4, 5] to deposit nickel onto graphite fibres, although in both cases the size of the tow has been some-

*Now at I.R.D. Co Ltd, Newcastle-on-Tyne.
†1 GN/m² = 145,000 lb/in².

what less, containing approximately 750 filaments.

Consolidation of the coated fibre was accomplished by a hot pressing operation. 50 mm lengths were laid longitudinally in a die and the assembly heated to a temperature of 800°C *in vacuo*. A pressure of 25 MN/m² was applied to the die and maintained for 1½ h, after which it was allowed to cool. By this procedure a flat plate was produced about 1.5 mm thick, which could be either file-cut or machined to the test-piece profile required. A typical cross-section of the composite is shown in fig. 2. The volume

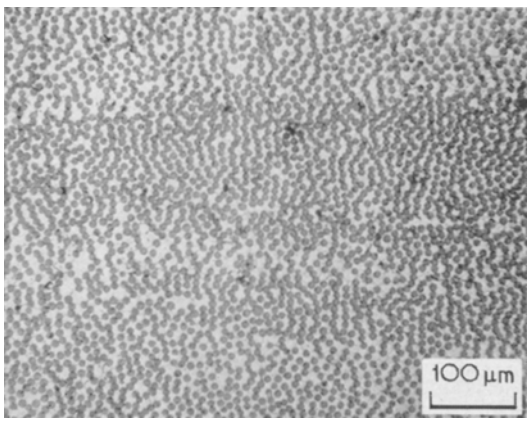


Figure 2 Cross-section of as-pressed composite.

fraction of fibre in the composite was maintained at between 50 and 60% for the majority of this work and was periodically checked by weighing the amount of fibre in a weighed sample after dissolving out the nickel.

The specific gravity of the composite was of the order of 5.3, but full densification was only achieved if the fibre tow had been efficiently coated with nickel, prior to hot pressing.

Heat treatment of the "as-pressed" composites was performed *in vacuo* at a temperature of 900°C for 75 h, and was usually followed by a final pressing operation similar to that described above.

3. Mechanical Testing

3.1. Tensile Strength

"Dog-bone" shaped test-pieces with a reduced cross-section of 4 to 5 mm² were stressed to failure in an Instron Universal testing machine, at a crosshead speed of 0.5 mm/min. Fig. 3 shows a typical example of a tensile fracture,

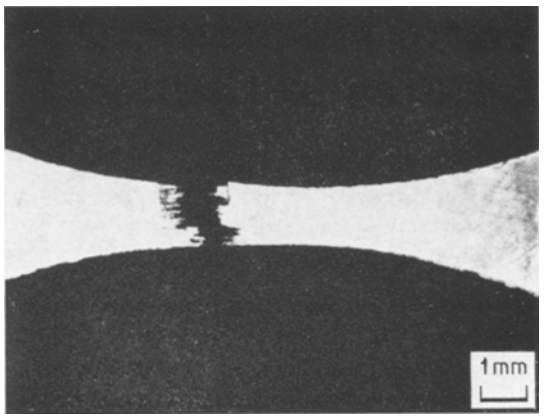
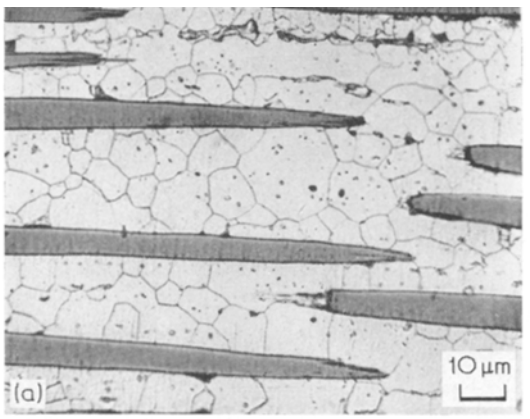


Figure 3 Typical tensile fracture of composite containing 50% fibre by volume.



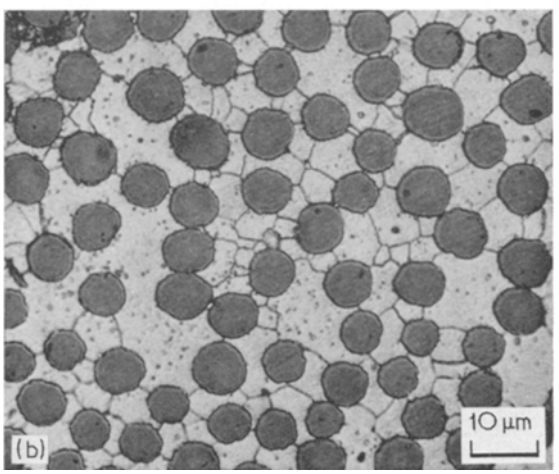


Figure 4 Optical micrographs of as-pressed composite (etched). (a) longitudinal section (b) cross-section. (Recrystallisation of the nickel has assisted in producing a well-bonded matrix with no signs of the original coating boundaries.)

although occasionally failure appeared to initiate in the gripped region. The fracture is essentially of a fibrous nature, and since the matrix is fully bonded (figs. 4a and b) it would appear that "pull out" of the fibres, from the nickel, had occurred. Stereoscan observations confirmed this and also showed that the matrix had suffered severe grain boundary embrittlement (figs. 5a and 5b). In addition, the alignment of the fibres

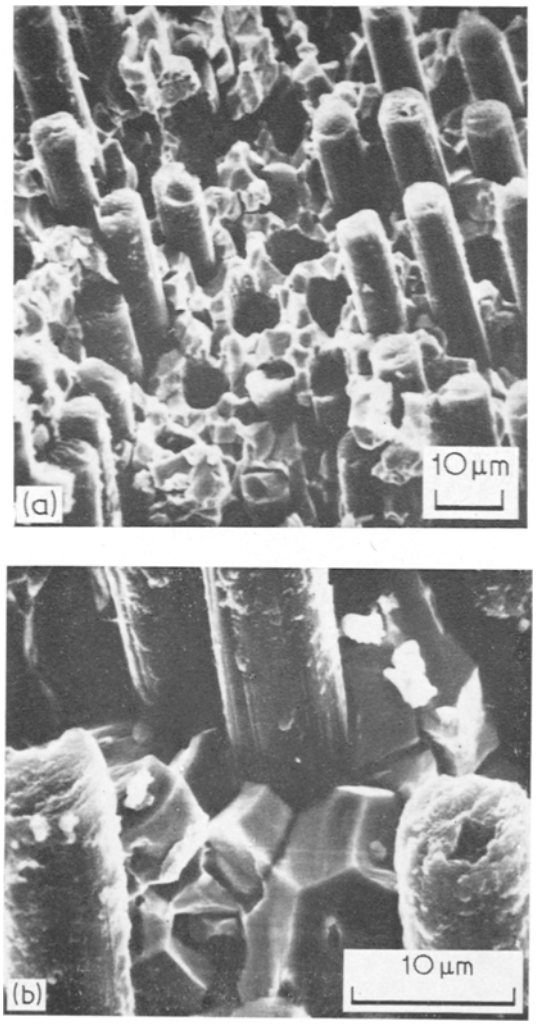


Figure 5 Scanning electron micrographs of a tensile fracture face. (a) fibre pull out from the brittle matrix (b) grain boundary embrittlement of the nickel.

was less than perfect, and the composite contained broken fibres (fig. 6).

Tensile strength values were generally in the

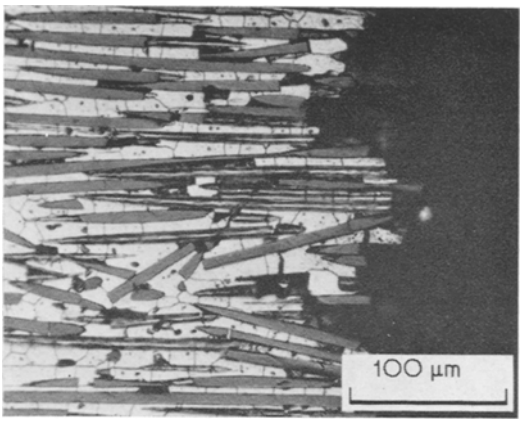


Figure 6 Optical micrograph of a composite tensile fracture face. Broken and misaligned fibres are present both at and away from the fracture.

range 450 to 590 MN/m^2 with the highest recorded value of 770 MN/m^2. It was evident that there were three reasons for both the low strengths and the large degree of scatter in the results:

(1) The bond between the fibre and nickel matrix was poor.

(2) Brittle nickel was preventing efficient load transfer to the fibres.

(3) The composite contained broken fibres.

The problem of the grain boundary embrittlement of electrodeposited nickel has also been encountered by Sutton *et al* [8], who were unable to identify the cause, but eliminated it by heat treating *in vacuo* for 24 h at 1275°C.

Degradation of the graphite/nickel composite would, however, result from such a treatment [3], but our own investigation revealed that longer times at lower temperatures could be used. It was found that ductile nickel could be produced (fig. 7) by heat treating the composite *in vacuo* for 75 h at 900°C. On tensile testing, however, the fracture was very fibrous, and the strength low, indicating that the fibre/matrix bond strength had been reduced still further. This was partially restored by a further hot pressing treatment similar to that used originally, but the tensile strength was not significantly improved.

The mechanism of failure appears to be largely one of "pull out" of the fibres, which is facilitated by the presence of broken or damaged fibres. In the "as-pressed" composites, cracks propagate along the nickel grain boundaries and also along the fibre/matrix interface. However, the improved ductility of the nickel, resulting

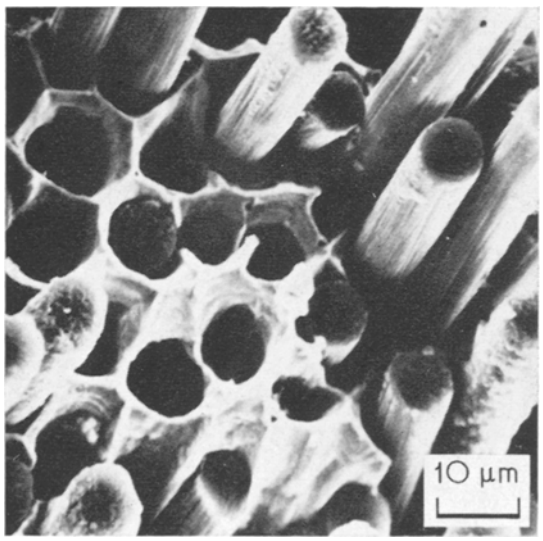


Figure 7 Scanning electron micrograph of a tensile fracture face. Heat-treatment has restored the ductility allowing localised necking of the nickel between the fibres.

from heat treatment, fails to increase the tensile strength of the composite. Thus the overriding weakening mechanism is the poor adhesion of carbon fibres to nickel coupled with a significant proportion of broken fibres. This was also apparent when a few tensile tests were performed at elevated temperatures, the bond strength being further weakened and the fracture so fibrous that the majority of test-pieces failed in the grips at low strength values.

Increased fibre "pull out" on tensile testing at elevated temperatures has also been attributed to a poor bond strength [4] and to filament breakage coupled with a reduction in the matrix shear strength [6].

3.2. Stress-Strain Behaviour

The essential features of this test were similar to those described by Baker and Cratchley [9]. A parallel sided, 12.5 mm wide, "as-pressed" composite test-piece was cycled a number of times at various load values, and the resultant stress-strain curve measured using a strain gauge extensometer and the Instron X-Y recorder. From the curves obtained the moduli could be measured and the response to load cycling of the material assessed. In addition, dynamic modulus measurements were made, calculated from the resonant frequency of the composite when subjected to sonic vibration, and hence at very low values of strain.

Load cycling to a given stress level resulted in the first cycle being an open loop followed by loop closure on subsequent cycling. Also in the initial cycle, yielding of the nickel brought about a change in slope and hence a second value of modulus. On raising the stress level, the composite followed the same pattern of hysteresis followed by loop stabilisation, as previously encountered with silica fibre reinforced aluminium [9]. The modulus values obtained from both the static and dynamic tests showed a large degree of scatter, with the majority of results falling in the range 207 to 276 GN/m^2 compared with the maximum theoretical primary modulus of 276 GN/m^2 calculated from the rule of mixtures. Secondary modulus values, obtained from the slope of the curve after the nickel had yielded were also low, being in the range 103 to 138 GN/m^2.

The occurrence of some low values of elastic modulus appeared to coincide with composite samples which had not been fully consolidated. This was probably due partly to poorly aligned fibre in the original tow (which in itself would also be expected to contribute slightly to a modulus reduction) and also to non-uniform fibre coating.

3.3. Three-Point Bending

The flexural strength of the composite was measured in three-point bending on test-pieces which had an aspect ratio (length/thickness) of $\sim$ 25:1 consisting of a 38 mm span and a thickness of 1.5 mm. Rollers were used to apply the bending moment in order to prevent local damage to the composite as might occur with knife edges. There was a large scatter in the results, the majority being in the range 0.83 to 1.1 GN/m^2. The onset of failure was usually rapid, beginning with a crack propagating from the side of the test-piece in tension, towards the neutral axis (fig. 8a), followed by interlaminar shear along the longitudinal axis (fig. 8b).

3.4. Impact Resistance

The type of test used was designed to break a notched test-piece in a single blow in four-point bending and to compare the energy absorbed with that of other materials.

The test-piece is illustrated in fig. 9. A small Hounsfield machine was used, with a 57 g tup, an anvil separation of 38 mm and a 6.3 mm wide centre hammer.

The effect of varying the fabrication conditions

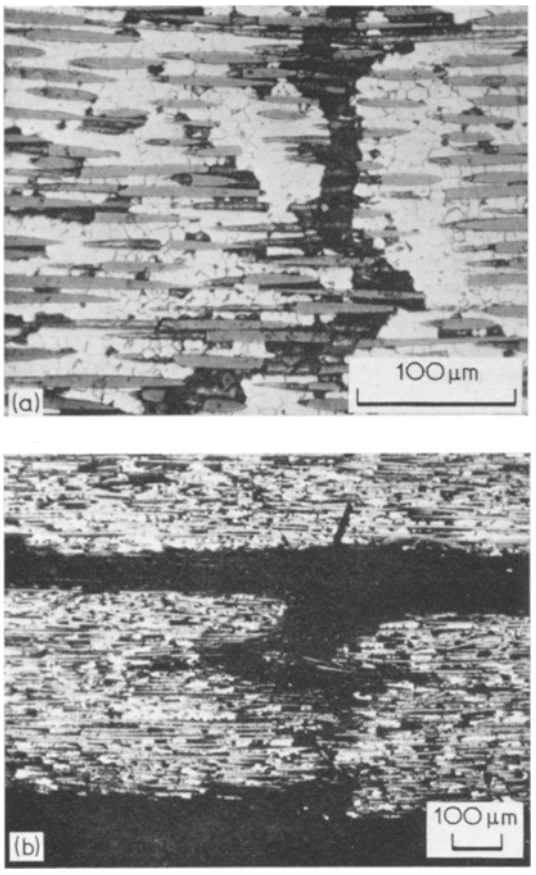


Figure 8 Optical micrographs of longitudinal sections of composite tested in three-point bending. (a) Propagation of tensile crack via grain boundaries assisted by fibre pull-out (b) Interlaminar shear cracking following tensile fracture.

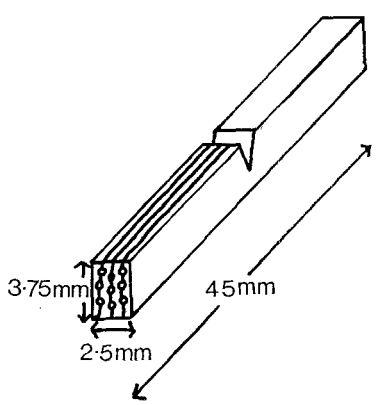


Figure 9 Notched impact test-piece configuration.

of the composite and of heat treatment were investigated, but in all cases no significant improvement could be made over the normal as-pressed condition. Table I compares results for the graphite/nickel system with those obtained from other materials tested under similar conditions.

TABLE I Longitudinal impact data

Material	Energy absorbed (Joules)*
C/Ni (as-pressed)	0.08-0.12
C/Ni (heat-treated)	0.08-0.11
RR58	0.19
B/Al	0.095
0.002 in. SiO_2/Al	0.43
0.005 in. SiO_2/Al	0.68

*1 Joule = 0.738 ft. lbs.

A weak interface, as is present in the silica fibre reinforced aluminium system, would be expected to hinder crack propagation through the material [9], and thereby improve the impact resistance, but in the case of graphite/nickel, with an apparently weak fibre/matrix interface, the fracture was generally of a more brittle nature (fig. 10b). The failure mode did however appear to be typical of a notch-insensitive material, with initial crack propagation often perpendicular to the direction of the applied stress (fig. 10a). Because the test is essentially in four-point bend, the remainder of the fracture mechanism is dependent on the point of impact of the two edges on the tup, since it is from these areas that the final fracture often occurred.

3.5. Fatigue Properties

Testing was performed in reversed bending at constant stress using the same test-piece configuration and fatigue testing assembly as previously employed for aluminium matrix composite studies [10]. The results are plotted in the form of an S-N curve (fig. 11) and are compared with Nimonic 80A and RR58 (an aluminium based alloy). Although the results show a large scatter, the curve is a relatively shallow one, as has been found with other composite systems [11, 12]. The endurance limit at 10^7 cycles is $\sim$ 400 MN/m^2 which compares very well with Nimonic 80A, but one serious disadvantage is the reduction in modulus which occurs during the fatigue process as a result of damage to the matrix. The fracture characteristics do not appear very different from those found on tensile test-pieces, with little or no

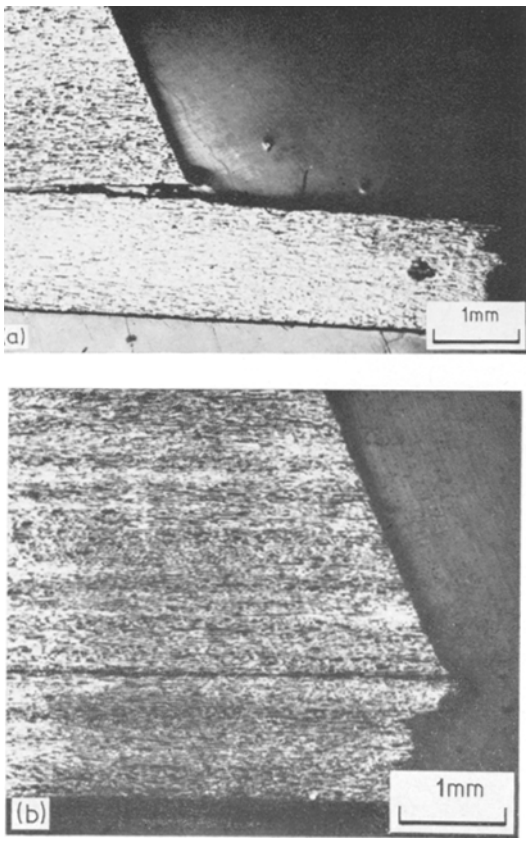


Figure 10 Optical micrographs of longitudinal sections through impacted test pieces. (a) Typically notch insensitive fracture (b) More brittle type of fracture but having associated interlaminar shear crack.

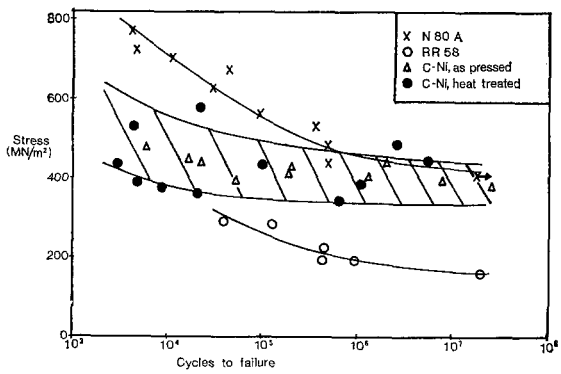


Figure 11 S-N curves obtained from reversed bending fatigue tests.

signs of actual matrix fatigue even after 10^7 cycles. Failure appeared to have occurred by the breakdown of both the nickel grain boundaries and the fibre/matrix interfaces (fig. 12a).

Although heat treatment resulted in the nickel exhibiting ductility when the composite was pulled in tension, it did not appear to significantly affect the mode of fatigue fracture. Fig. 12b shows a fatigue fracture surface and although there is some evidence of fatigue striations in the matrix, the majority of it appears to have failed in an intergranular manner.

3.6. Oxidation Resistance

As-pressed composites were heat treated at 600°C for various times in an open ended tube furnace, in order to determine what degree of protection of the fibres was afforded by the

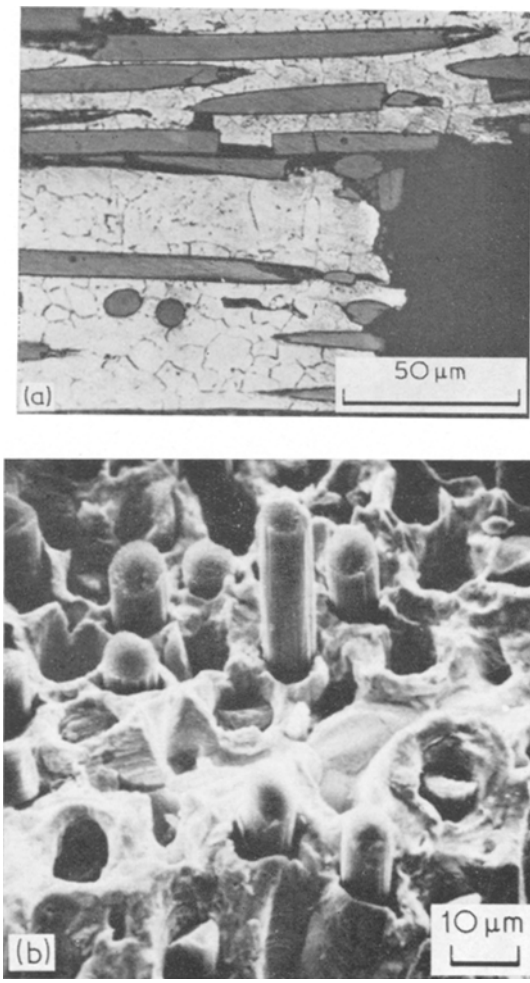


Figure 12 Fatigue fracture surfaces. (a) Optical micrograph of longitudinal section (b) Scanning electron micrograph. showing signs of fatigue striations.

presence of nickel. In a matter of 1 h oxidation of the fibres was apparent throughout the whole structure, and within 5 h the majority of the fibres had disappeared (fig. 13a). Fig. 13b shows

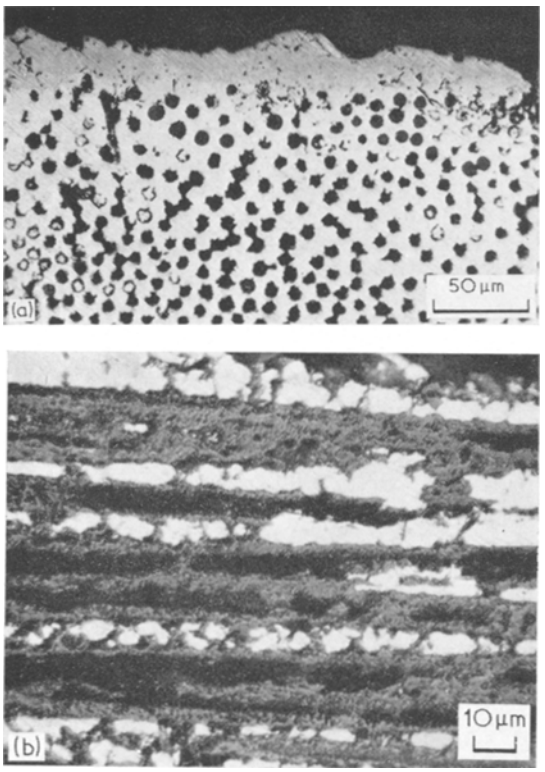


Figure 13 Optical micrographs of oxidised composite. (a) Cross-section after 5 h at 600°3C (b) Longitudinal section after 200 h at 600°C, showing growth of nickel oxide, replacing carbon fibres.

a longitudinal section of an oxidised sample, from which it appears that oxidation begins at the fibre/matrix interface with consequent disintegration of the fibre on one side and formation of nickel oxide on the other. In addition, there is also evidence of oxygen penetration through the nickel itself, probably via grain boundary diffusion.

4. Discussion

The tensile fracture surface of the composites showed fibre "pull out", but in no instance could any nickel be seen adhering to the fibre surfaces. In addition, the few transverse tests which were performed indicated that the fibre/matrix bond strength was very low, although this form of test is susceptible to fracture occurring from localised areas of weakness, e.g. fibre/fibre contact. Niesz *et al* [5] also found the transverse strength of a carbon fibre/nickel composite to be low, but nevertheless showed evidence of nickel adhering to the exposed fibres on a longitudinal tensile fracture face. Since they obtained values of up to 80% of the rule of mixtures tensile strength, it is thought that our own composites suffered from poor fibre alignment and hence broken fibres, and also a less-even fibre distribution than the exceptionally carefully prepared Battelle composites.

The presence of a weak interface did not, however, result in a good impact resistance, as was found for instance with the Al/Al interface in silica fibre reinforced aluminium [9]. The presence of broken fibres produced both in fabrication and at the point of impact of the tup would presumably assist both fibre "pull out" and translaminar crack propagation. Although improvements in fracture toughness might be expected from a composite containing less fibre damage, recent work has confirmed that decreasing the fibre diameter reduces the work to fracture [13] unless there is significant energy absorption by an alternative mechanism such as pull-out or delamination.

The reversed bending fatigue properties appeared comparable with Nimonic 80A, although the fracture, which showed no evidence of actual matrix fatigue, suggested that a fully bonded composite would perform even better. The 10^7 cycles endurance limit of 400 MN/m^2 was surprisingly high when compared with the average tensile strength of about 520 MN/m^2, although as already pointed out this is only 50% of the rule of mixtures tensile strength. The fatigue process, as has been found with other composite materials [14, 15], appears to seek out weak interfaces so that even the apparently ductile nickel, obtained by heat treatment, showed some signs of intergranular failure. The S-N curve was a shallow one, which appears to be typical of a fibre reinforced metal [11, 12, 16] although in this case the features on the fracture surfaces appeared to be similar on both high and low endurance test-pieces.

Although graphite fibres are not inherently oxidation resistant, it was originally assumed that the presence of nickel would, at very least, inhibit the access of oxygen to them. However, the weak interfaces present in the composite,

including grain boundaries, allowed rapid diffusion of oxygen to the fibres, resulting in internal oxidation of the nickel, and complete degradation of the fibres. In addition to this, the oxidation behaviour of nickel coated graphite fibres has been compared with that of uncoated graphite fibres using a thermobalance [17]. From the few tests performed it was apparent that the presence of nickel served to accelerate the rate of oxidation of the fibre. One possible interpretation of this is that oxygen diffuses through nickel in a highly active monatomic form and would therefore react more readily with the carbon.

5. Conclusions

(*a*) The mechanical properties of a graphite fibre reinforced nickel composite have been investigated, and it is concluded that longitudinal properties could be improved by more careful fibre alignment and possibly better fibre distribution within the matrix.

(*b*) Near theoretical tensile strengths will only be obtainable by a substantial improvement in the fibre/matrix bond strength, particularly at elevated temperatures.

(*c*) The poor oxidation resistance, even at 600° C, is a serious limitation for high temperature use.

Acknowledgements

The authors wish to thank Mr R. Walker for his assistance with the majority of the work in this report, Mr J. Spencer for his help with designing and building the electrodeposition assembly, Miss H. Baker for her work on the stress-strain behaviour, Dr A. A. Baker for his useful comments and suggestions, and Mr B. A. Proctor for his general support and guidance during this programme of work.

The permission of Rolls-Royce Ltd to publish the paper is gratefully acknowledged.

References

1. A. KELLY and G. J. DAVIES, *Met. Reviews* **10** (1965) No. 37.
2. D. CRATCHLEY, *ibid* **10** (1965) No. 37.
3. P. W. JACKSON, *Metals Engng Qtly* **9** (1969) 22.
4. R. V. SARA, 14th Nat. *SAMPE Symposium* Nov. 1968.
5. D. E. NIESZ and C. W. KISTLER JR, *ASM WESTEC Conf.* Los Angeles. March 1969.
6. C. W. KISTLER JR and D. E. NIESZ, AD-854149 May 1969.
7. D. M. BRADDICK, Brit. Pat. No. 1208959, 1970.
8. J. CHORNE, G. A. BRUCH, and W. H. SUTTON, AD-646919 1966.
9. A. A. BAKER and D. CRATCHLEY, *Appl. Mat. Res.* **5** (1966) 92.
10. *Idem, ibid* **3** (1964) 212.
11. H. SHIMIZU and J. F. DOLOWY J., *Composite Mat. Testing and Design ASTM-STP*-460 (1969) 192.
12. A. A. BAKER, *J. Mater. Sci.* **3** (1968) 412.
13. R. E. COOPER, *J. Mech. Phys. Solids* **18** (1970) 179.
14. P. W. JACKSON, A. A. BAKER, and D. M. BRADDICK, submitted for publication.
15. A. A. BAKER, *J. Mater. Sci.* **5** (1966) 143.
16. K. G. KREIDER and G. R. LEVERANT, 10th Nat. *SAMPE Symposium* Nov. 1966.
17. A. A. BAKER, private communication.

Received 10 March and accepted 20 March 1971.