JOURNAL OF MATERIALS SCIENCE 29 (1994) 419–423

Effect of fibre misalignment on the fracture behaviour of fibre-reinforced composites

Part I. *Experimental*

W. B. HILLIG*
General Electric Company, Corporate Research and Development, Schenectady, NY 12301, USA

Deviations from ideal parallel packing in a unidirectional fibre-reinforced composite affect its resistance to splitting. In order to relate, quantitatively, the failure processes to such misorientation, it is necessary to characterize the departure from ideality and to measure the resistance to failure. Experimental observations are presented relating to: (i) a tendency for the carbon fibres in a tow to group into bundles that deviate somewhat from being parallel with each other, and (ii) the fracture toughness for the splitting of several imperfectly aligned composites. Statistical representations are offered for quantifying or modelling the degree of misalignment.

1. Introduction

Micromechanical models of non-woven fibre composites typically consider the fibres to be parallel and regularly spaced. However, truly parallel alignment is not feasible in real composites made of multifilamental tows and yarns comprised of filaments which are too fine to be handled individually. Within a tow, the fibres tend to form local groups in which the fibres are parallel. Such local groups may be of various sizes and are termed bundles; that is, a tow is comprised of an array of misoriented bundles. This paper examines how various deviations from ideal parallel packing affect the resistance to splitting parallel to the mean fibre orientation.

Wood illustrates the effect of misalignment: American elm is notoriously difficult to split, whereas it is difficult to avoid splitting oak. The constituents and compositions of these woods (the cellulosic reinforcing fibres and the matrix composed of hemicelluloses and lignin) are substantially the same in both species. The twisting and intertwining of the fibres in elm are apparently responsible for its toughness since they preclude the possibility of a cleavage plane parallel to the fibres in the radial direction. In contrast, the fibres in oak are nearly parallel along the axis of the trunk and do not interfere with radial cleavage.

The effect of misalignment on the toughness of epoxy resin–E-glass-fibre composites has been previously noted. The work of fracture for crack initiation in a splitting mode of failure was found not to be affected by fibre misalignment. However, the work of fracture for crack propagation increased with increasing degrees of misalignment.

Relating the failure behaviour, quantitatively, to the misorientation requires a measure of the bundle size and the misalignment of a bundle with respect to the mean direction of the fibres. The term *failure* is defined here as the separation of the original composite body into two bodies. This paper presents experimental observations and measurements relating to: (i) a tendency for fibres to group into bundles that are misaligned relative to each another, but within which the fibres are substantially parallel; (ii) the interaction of a crack with a single misaligned fibre; and (iii) the fracture toughness for the splitting of imperfectly aligned composites. A separate paper will offer a model for predicting the effect of misalignment on composite-failure behaviour.

2. Microstructure of multifibre unidirectional composites

2.1. General

The imperfections in the alignment of individual fibres in a representative, nominally unidirectional, composite can be revealed from examination of polished sections cut at a slight angle to the mean fibre direction. Fig. 1 shows such a section for the case of an epoxy-matrix composite reinforced with parallel tows of continuous carbon fibres. This micrograph reveals a substructure consisting of fibre colonies within which adjacent fibres have nearly the same ellipticity and the same tilt of their major axis. Often, many of the neighboring colonies (bundles) exhibit distinctly different eccentricities and orientations. Furthermore, in some regions the fibres can be seen to be packed in square arrays, and in other regions they are packed in

* Presently at Rensselaer Polytechnic Institute, Department of Materials Engineering, Troy, NY 12180-3590, USA.

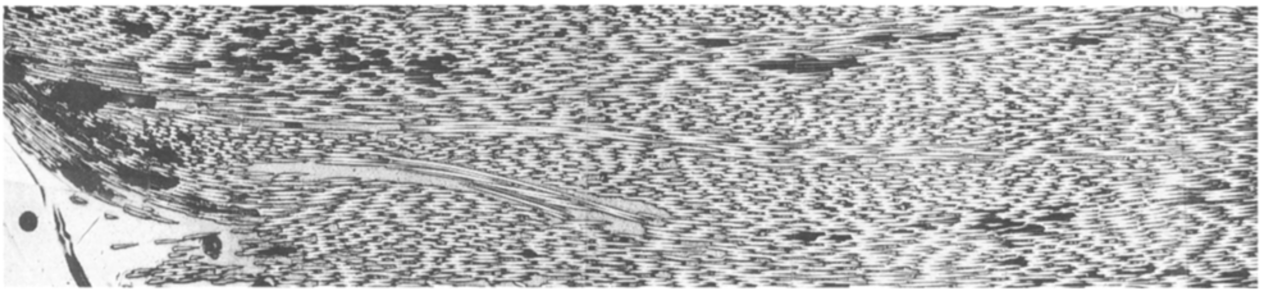

Figure 1 A polished section through an epoxy/graphite-fibre composite cut at 10 °C relative to the nominal unidirectional fibre axis.

hexagonal arrays. The bundles are estimated to contain between about ten and a hundred filaments.

A pultruded, unidirectional, epoxy/glass composite, used in the measurements presented later, exhibited a similar bundle substructure. These observations suggest that, in addition to the conventional parameters for mean fibre orientation (the volume fraction and mean fibre diameter), a more complete geometric characterization is needed that includes specification of the size and orientation distributions of fibre bundles. These new parameters can be expected to relate to the splitting toughness, the interlaminar shear strength, and the compressive strength through their effect on buckling.

The term *bundle* is not meant to suggest a spatially persistent entity; it is probable that any given fibre may be associated with one bundle over some length and then it can be correlated with another bundle after some distance. The occurrence of a bundle-like substructure is not unexpected. Efficiency in filling space requires local correlation of fibre orientations. Thus, such a substructure is expected to be a common feature of composites, particularly those made from strands of fibres. Although automatic computerized stereological methods are available for determining the size distribution and volume fraction of individual fibres, to the author's knowledge, such a capability has not yet been developed for quantifying the orientations of the individual fibres or the size and orientation of such spatially correlated regions. Nevertheless, the manual procedure, discussed next, can provide a quantitative statistical description of the degree of alignment.

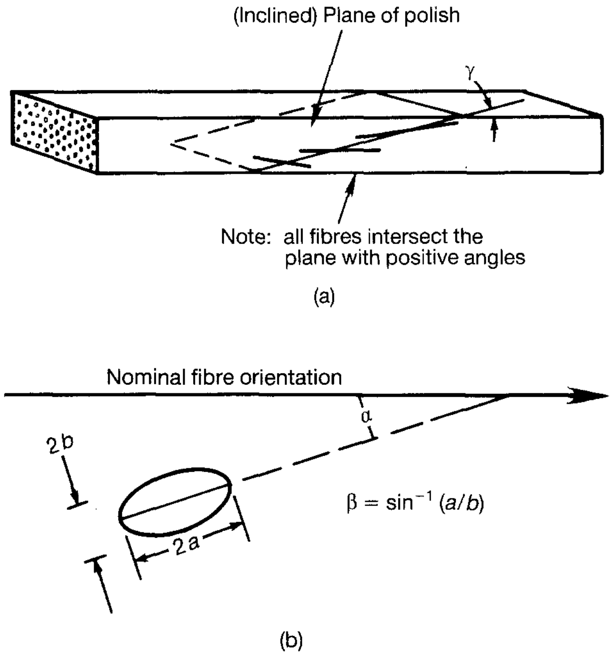

Figure 2 A schematic showing: (a) determination of the orientational parameters, and (b) tilting of the polish plane to eliminate orientational ambiguity.

2.2. Statistical description and methodology

Statistical representations of orientational distributions have been offered [2] by geologists and biometricists. However, these standard representations are not convenient for the present purposes. An approach similar to that given by Hull [3] is adopted instead. Assuming that the fibres have circular cross-sections, ellipses result when the fibres intercept the plane of polish. The eccentricity of the ellipses can be used to determine the angle that the fibres make with the mean fibre direction, as shown in Fig. 2a.

Consider a nominally unidirectional, composite bar specimen made up of fibres with circular cross-sections and aligned so that the fibres are approximately parallel with the long axis of the bar. Let the top surface of the bar be polished to reveal the fibres. If the fibres were perfectly aligned, the exposed fibres would lie in the plane of the polish over the entire length of the bar, and their traces would be parallel to the bar axis. Fibres lying in the plane of polish, but not aligned parallel to the bar axis, would have their traces cocked with respect to the bar axis. However, fibres not lying in the plane of polish would, at some point, intersect that plane at an angle β, so that their polished cross-sections will be ellipses as shown in Fig. 2b. This crossing angle, β, is given by $\sin \beta = b/a$, where b/a is the ratio of the minor to the major axis. The angle that the long axis makes with the bar axis is simply α. Hence, from measurements of α and β, the spatial angle, θ, that the fibre makes relative to the bar axis can be determined.

However, identical ellipses result for positive or negative values of β. This ambiguity can be resolved by inclining the plane of polish, relative to the bar axis, by a known angle, γ, as indicated in Fig. 2a. This angle γ is chosen to exceed any of the fibre misalignments. In this way, all fibres pass through the plane of polish with gradients of the same sign. The angles α and β in the inclined plane of polish are determined as described above. The spatial angle θ by which a fibre deviates from the bar axis is given by

$$\cos\theta = \cos\alpha\cos\beta\cos\gamma + \sin\beta\sin\gamma. \quad (1)$$

Note that the longitudinal axis of the specimen need not coincide with the mean local fibre direction. By replacing the arbitrarily chosen angle γ by β_{av}, the computed values for θ represent the deviations from the mean direction. The difference between β_{av} and γ is a measure of the difference between the nominal and the local mean fibre directions.

The distribution of the crossing angle, β, is given in Fig. 3a as derived from measurements on 128 fibres which intersect random lines placed across the micrograph. The derived distribution for θ, based on using β_{av}, is shown in Fig. 3b to be strongly peaked around $\theta = 0$, and practically to vanish for $\theta > 6°$.

Orientational correlations between neighboring fibres are more difficult to specify. However, indications of such correlations can be seen by plotting the

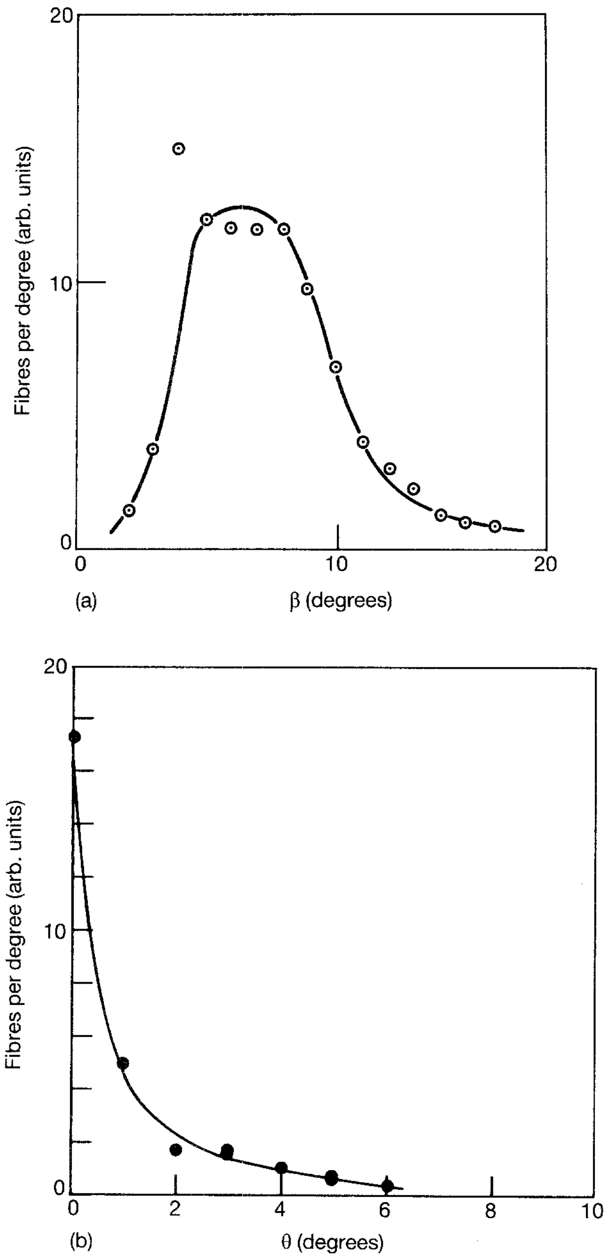

Figure 3 The orientational distribution of the fibres shown in Fig. 1: (a) on the plane of polish, and (b) around the axis of unidirectional symmetry.

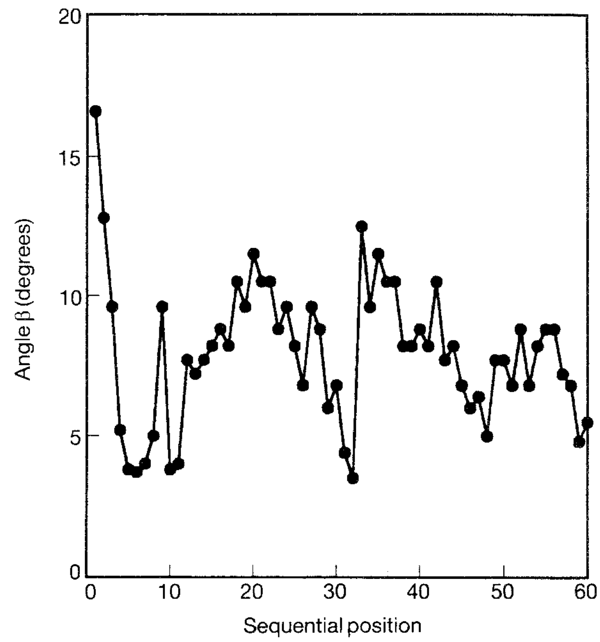

Figure 4 Correlations in fibre orientation shown by plotting β-values for sequential positions across the width of the sample.

α and β-values for the individual fibres against their sequential positions in traversing the polished section as shown in Fig. 4. Points corresponding to fibres that are judged to be in the same bundle are connected by lines. However, visual inspection of Fig. 1 perhaps allows delineation of the bundles even more distinctly. Over a section comprised of 60 adjacent fibres, about eight bundles could be identified, plus possibly two more bundles. In spite of such imprecision, these values indicate that mean bundle diameters span roughly seven fibres in this case. Thus, a typical bundle is estimated to contain about 50 ($\approx 7^2$) fibres.

3. Mechanical Tests of composite splitting

Epoxy-matrix composites in the form of sheets reinforced with either pultruded E-glass fibres (A) or with randomly oriented chopped E-glass fibres (B) were selected for testing. These two composites represent extremes in the degree of misorientation practically achievable using multifilament tows as the starting material. In addition, a completely different composite system (C), consisting of a glass matrix reinforced with nominally unidirectional graphite fibre, was also investigated. The characteristics of the three composites are summarized in Table I.

Composite C, in the form of a slab made by hot-pressing unidirectionally aligned C fibres in a code-7740-borosilicate-glass matrix, was supplied by the kind cooperation of Dr K. Chyung of the Corning Glass Works. The properties of the Hercules HMS graphite fibre are stated by the manufacturer to be: diameter 8 μm; Young's modulus 320 GPa; and failure strain, 0.58%. The properties of the matrix glass reported in Corning property data sheets are: Young's modulus, 91 GPa; failure strain, 0.08%; and Poisson's ratio, 0.20. The composite contained 48 v/o of fibre reinforcement. The flexural strength of the composite

TABLE I Characteristics of the composite systems tested

	System		
	A	B	C
Matrix	Epoxy	Epoxy	Borosilicate
Fibre	E-glass	E-glass	Graphite
Fibre diameter (μm)	10	20	8
Fibre v/o	60	25	45
Fibre length	Continuous	25 mm	Continuous
Bundle diameter[a](μm)	100	200	1100
Misorientation[a](degrees)	5–10	90	5–10
Fibre modulus (GPa)	69	69	345
Fibre fail strain[a]	0.02	0.02	0.007

[a] Estimated value.

is 585 ± 35 MPa and its Young's modulus is ~ 170 GPa.

The ultimate strength and the work of fracture were determined for these three composite materials using a three-point-bend test procedure. The test coupon size for the A and B composites was $75 \times 19 \times 2.8\ mm^3$ with a 1.9 mm deep, center notch; and the test coupon size for the C composite was $50 \times 23 \times 5\ mm^3$ without a notch. The fibres in composites A and C were oriented perpendicular to the axis of the specimen.

The two epoxy-matrix composites behaved quite differently when stressed. The A material, upon loading, first developed a narrow, straight, opaque band extending from the base of the notch to the bottom edge of the specimen. This band consisted of a multitude of parallel microcracks that developed before any gross crack opening was evident. In constrast, the B material showed extensive irregular cracking during early stages of loading, and it displayed only a short opaque zone, limited to the crack-tip region. The specimens are shown in Fig. 5a and 5b during early stages of loading. The opaqueness of the graphite-fibre composite C prevented any possible observation of such a band as developed with composite A. However, the appearance of bundle bridging in composite C as the crack opened was similar to that seen in composite A. The force–displacement curves for composites A, B and C are given in Fig. 6a–c, and the results of the tests are summarized in Table II. These curves are similar in shape to those reported for splitting-mode failure of composites [4], but for which no misorientation information was reported.

The work of separation was determined from the area under the force–displacement curves. In these experiments the splitting resulted in a fibrous rent rather than in clean cleavage because many bundles were inclined to the mean plane of the fracture and they continued to bridge the crack. The ultimate-strength and work-of-separation values were greatest in the case of composite B. Because the fibre bundles in composite B were randomly oriented, many lay approximately perpendicularly to the crack faces. The results show that near-perpendicular bundles are more effective in transferring the load across the crack opening than are the more inclined fibre bundles. However, once these normal load-carrying members fail, the load-carrying capacity of the structure drops precipitously, by 79% in this example. Nevertheless, the high values for the work of separation for composites A and C, relative to the work of fracture for the matrix phase alone (typically less than 100 J m^{-2} for epoxy [5] and less than 1 J m^{-2} for inorganic glass [6]), show that substantial toughening can result from the more-inclined fibres. The lower value for the work of separation in composite C, relative to composite A, is in part a reflection of the intrinsically lower energy storable in a graphite fibre relative to a glass fibre prior to fibre failure.

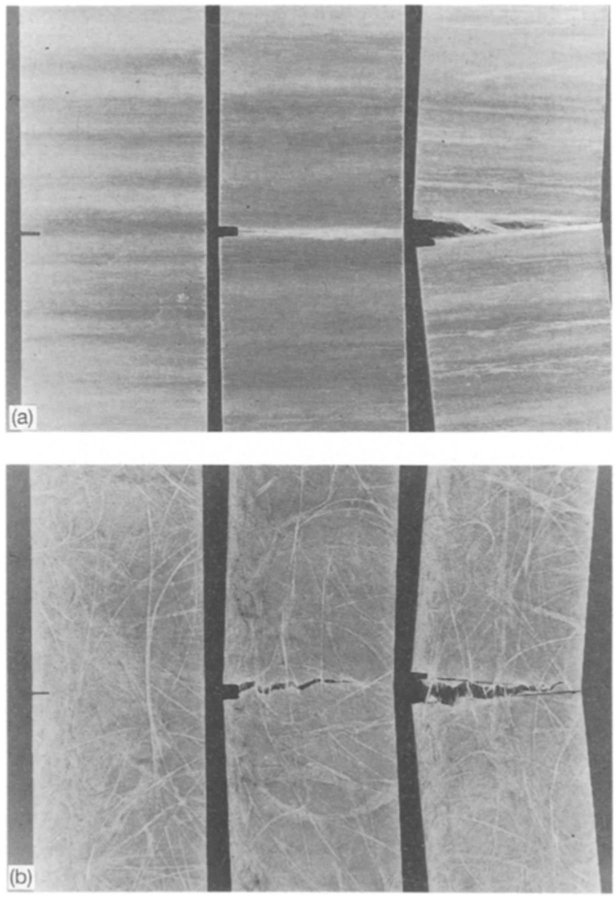

Figure 5 Appearance of cleavage cracks in epoxy–E-glass-fibre composites during the early stages of loading when: (a) fibre tows are pultruded, and (b) fibre tows are randomly oriented.

TABLE II Mechanical-test results

	System		
	A	B	C
Ultimate strength (MPa)	16.6	44.1	5.3
Reduction in strength (%)	27	79	55
Work of separation (J)	2300	5550	225

4. Conclusion

Misalignment of fibres in unidirectional composites is common in composites made from multifilament strands. Locally, the fibres are organized into bundles within which the fibres are essentially parallel. Such a substructure interferes with the clean cleavage of a

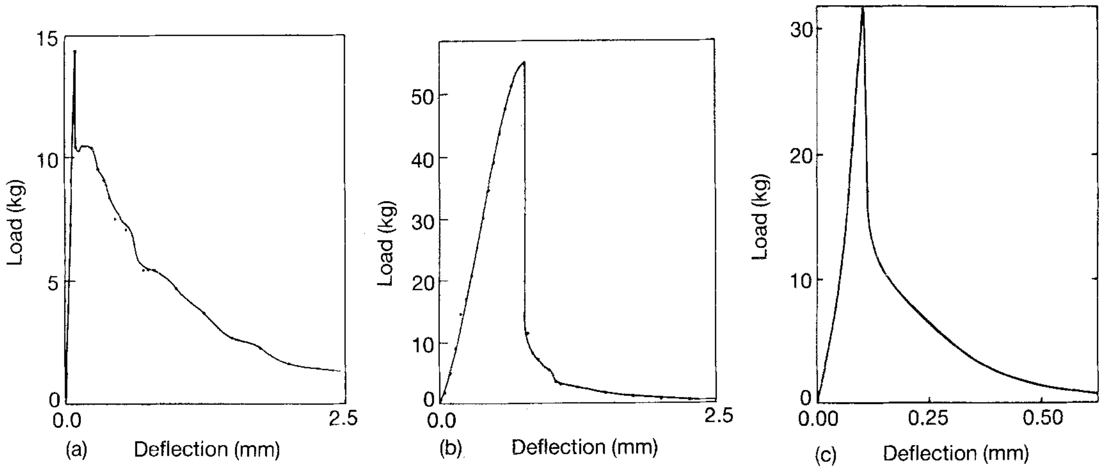

Figure 6 Load versus deflection curves for splitting-type failure in three-point-bend tests for: (a) an epoxy–pultruded-E-glass composite (60 v/o fibre), (b) an epoxy–randomly-oriented-E-glass composite (25 v/o fibre), and (c) a borosilicate-glass–unidirectional-graphite-fibre composite (45 v/o fibre).

composite parallel to the nominal mean fibre direction. In principle, the substructure can be specified in terms of a distribution of bundle sizes, and a distribution of angular deviations of the bundles from the mean fibre direction. A procedure for experimentally determining the misorientation of individual fibres has been given. Visual examination of suitable polished sections reveal the grouping of fibres into bundles, but this approach is somewhat subjective. Quantitative determination of the scale of the substructure and the degree of misalignment would provide valuable additional information to the data now commonly used to specify fibre composites. However, the effort required to obtain such information manually precludes such determinations on a routine basis until such a time as appropriate instrumentation and information processing become available.

Composite C showed crack-bridging fibre toughening even though the matrix was a brittle glass that was comparable in stiffness to the non-ductile reinforcing graphite fibre. These conditions approximate those in a model study [7] of the interaction of a crack with a single fibre. That model study indicated that the required matrix *compliance* for crack-face separation can be achieved by failure of the matrix resisting flexing of the fibre. Thus, secondary matrix cracking may be the process which accommodated crack bridging in composite C.

The experiments on the splitting failure in composites A and B show that the toughness for a highly aligned composite, as measured by the work of separation, can be comparable to that when the fibres criss-cross the crack opening at all possible angles. The criss-crossing is noteworthy, since in composite A the crack traverses the weakest direction, whereas in composite B there is no strongest direction. Hence, it may be advantageous to toughen a weaker direction in a composite by purposely controlling the degree of misalignment.

The purpose of this paper was to document evidence that fibre misalignment and the co-operative local association of such fibres into *bundles* occur in real composites, particularly when they are constituted from multifilament tows, yarns or strands. Such a geometric description lends itself to a theoretical modelling of the strength and toughness of unidirectional and multidirectional composites. Further experimental work is needed to determine the generality of such a geometric description for other composite systems and the effect of such a geometry on mechanical and physical properties. Finally, although this paper has emphasized the effect on crack growth and final composite separation, regions of high misalignment can be expected to serve as nucleation sites for instability-failure processes such as buckling and shear-induced delamination.

Acknowledgement
Discussions with Dr William T. Tucker on statistical procedures for quantifying the microstructural geometry have been helpful and are appreciated.

References
1. I. ENGELBERG and G. MAROM, *Fibre Sci. Technol.* **9** (1976) 63–67.
2. G. S. WATSON, *J. Geology* **74** (1966) 786–797.
3. D. HULL, "*An introduction to composite materials*", (Cambridge University Press, Cambridge 1981) pp. 71 ff.
4. G. MAROM and E. F. T. WHITE, *J. Mater. Sci.* **7** (1972) 1299–1307.
5. D. L. HUNSTON, *Compos. Technol. Rev.* **6** (1984) 176.
6. S. W. FREIMAN, T. L. BAKER and J. B. WACHTMAN jr., NBS Technical Note 1212, (National Bureau of Standards, Gaithersburg MD, 1985).
7. W. B. HILLIG, *J. Am. Ceram. Soc.* **75** (1992) 3172.

Received 30 July
and accepted 12 August 1993