JOURNAL OF MATERIALS SCIENCE 30 (1995) 3747–3751

Solvent and interstitial fibres in continuous fibre composites

P. G. PARTRIDGE
Interface Analysis Centre, University of Bristol, Bristol, UK

The possibility of introducing small-diameter fibres (interstitial fibres) into the interstices between larger diameter fibres (solvent fibres) to produce continuous interstitial fibre composites (IFCs) has been considered. The interstitial void size and shape is dependent on the type of stacking of the cylindrical fibres. It is shown that the maximum total fibre volume fraction can be substantially increased by the introduction of one or more interstitial fibres. The distribution of the fibre clusters creates a new type of composite microstructure. Possible interstitial fibres and the effect of fibre clusters on composite properties are discussed.

1. Introduction

Continuous fibre-reinforced polymeric materials offer a substantial increase in strength and stiffness compared with the unreinforced materials and are now in widespread use. With metal matrix composites (MMCs) it has been more difficult to compete with unreinforced metallic materials on the basis of their combination of properties and cost [1]. Because properties depend on fibre spacing, and in particular on the amount of fibre/fibre contact, controlling and inspecting fibre spacing is a major cost factor in MMC manufacture [1]. A recently reported MMC manufacturing process uses matrix-coated fibres (MCF) [2]. During consolidation the coating flows plastically and fills the voids between the fibres. This prevents fibres touching and allows close control over the fibre spacing, which is dictated by coating thickness.

In parallel with the progress in MMC processing, there has been an increase in the variety of high-strength continuous fibres available [3]. They have diameters from about 10–140 μm and different physical and mechanical properties, such as density, modulus, strain to fracture, thermal expansion and electrical and thermal conductivity. Although mixed fibres have been used in polymer matrices (hybrid composites), they have been less attractive for MMCs, because of the difficulty in consolidation. Using the above matrix coating process, this problem may be overcome, allowing a mixture of fibre sizes and types to be combined and increasing the potential for new continuous fibre composites.

In advanced continuous fibre MMCs, e.g. high-strength composites or composites for use at high temperature, the matrix plays many roles. It holds the fibres together and acts as a load-transfer medium, and provides some toughness by reducing the stress at crack tips. However, the matrix may limit the long-term service performance under creep or oxidation conditions. It may then be desirable to reduce the matrix volume fraction to a minimum consistent with the above roles. The possibility of designing a new type of fibre composite microstructure has therefore been considered. By analogy with solvent and interstitial atoms in metallic solid solutions, a concept introduced in this paper is that of placing smaller matrix-coated fibres (interstitial fibres) in the interstices between the larger coated fibres (solvent fibres) in a continuous hybrid fibre composite. The potential composite microstructure and fibre packing density in such composites is described.

2. Stacking of continuous fibres

Finite element techniques have been used to study the effect of fibre distribution on residual stresses and deformation in continuous fibre composites [1, 4–6]. The fibre volume fractions were about 50 % and hence the fibres were widely spaced. The fibre stacking arrays used were a vertical (also called square or square edge) stacking or close packed (also called triangular or hexagonal) stacking. Consider the maximum packing density for long parallel cylindrical ceramic fibres in contact in these two stacking arrays.

A section normal to the fibre axes through a stack of five rows of fibres of radius R is shown schematically in Fig. 1. Rows **a b a** have the fibres in the vertical stacking sequence, with fibres vertically above each other leading to a bilayer thickness $h_{a/b} = 4R$. Rows **a c a** have the close packed stacking sequence with fibres above lying between those below, leading to a bilayer thickness $h_{a/c} = 3.73R$. The nearest and next-nearest neighbour fibres are indicated by the pairs of arrows in Fig. 1. Vertical stacked fibres in cross-section exhibit four-fold symmetry and have four nearest neighbour fibres at a distance $2R$, and four next-nearest neighbour fibres at a distance $2.83R$. Close packed layers exhibit six-fold symmetry and have six nearest neighbour fibres at a distance $2R$, and

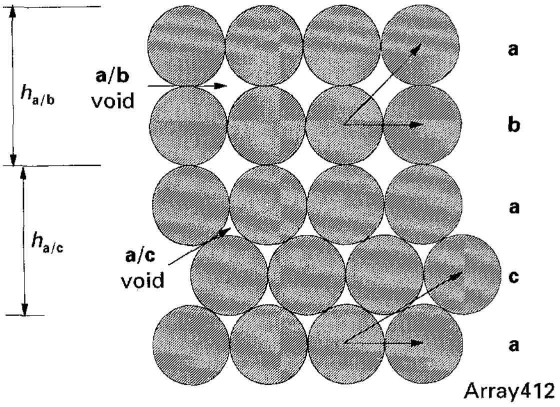

Figure 1 Schematic section through a continuous fibre composite showing vertical stacking **aba** and close packed stacking **aca** and corresponding interstitial voids.

six next-nearest neighbour fibres at a distance $3.46R$. These fibre spacings are reflected in the minimum interstitial void volume fractions, $V = 21.5\%$ and 9.3% for vertical and close packed stacking, respectively. For the same size fibres, vertical stacking leads to a layer thickness about 7.2% thicker than for close packed stacking.

The long interstitial voids between the fibres have different cross-sectional shapes and areas for the two types of layer stacking, as shown for the a/b and a/c voids in Fig. 1. In layers with vertical stacking, each fibre is surrounded by four voids, leading to one void per fibre. In cross-section each void has an area $A = 0.86R^2$ and a shape exhibiting four-fold symmetry. In the plane of the cross-section, the voids lie on a net similar to that of the fibres, with four-fold symmetry. In layers with close packed stacking, each fibre is surrounded by six voids, leading to two voids/fibre. In cross-section each void has an area $A = 0.16R^2$ and a shape exhibiting three-fold symmetry. The voids lie on a net in the cross-section similar to that of the fibres, with six-fold symmetry.

These interstitial voids represent the theoretical minimum intrinsic void size and volume fraction that can be achieved by packing arrays of cylindrical fibres of the same diameter. In an ideal composite designed for a maximum volume fraction of fibre the voids would be filled by the matrix and a thin bonding matrix layer would separate the fibres to give a composite of maximum density. In practice, the fibre spacing and the void size are usually greater than the minimum.

3. Diameter of interstitial fibres

The theoretical maximum fibre diameter that can be inserted into an interstitial void in a vertical stacked fibre array is shown in Fig. 2a. It has a diameter of $0.83R$ and would occupy 63% of the interstitial void volume. Alternatively, multiple interstitial fibres may

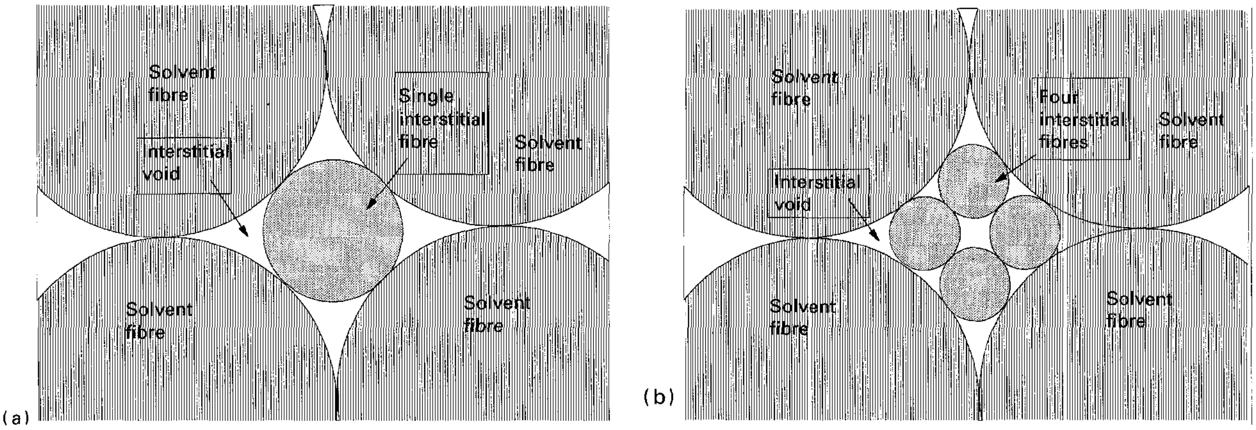

Figure 2 Diagram of interstitial fibres in vertical stacked fibre array: (a) single fibre, (b) four fibres.

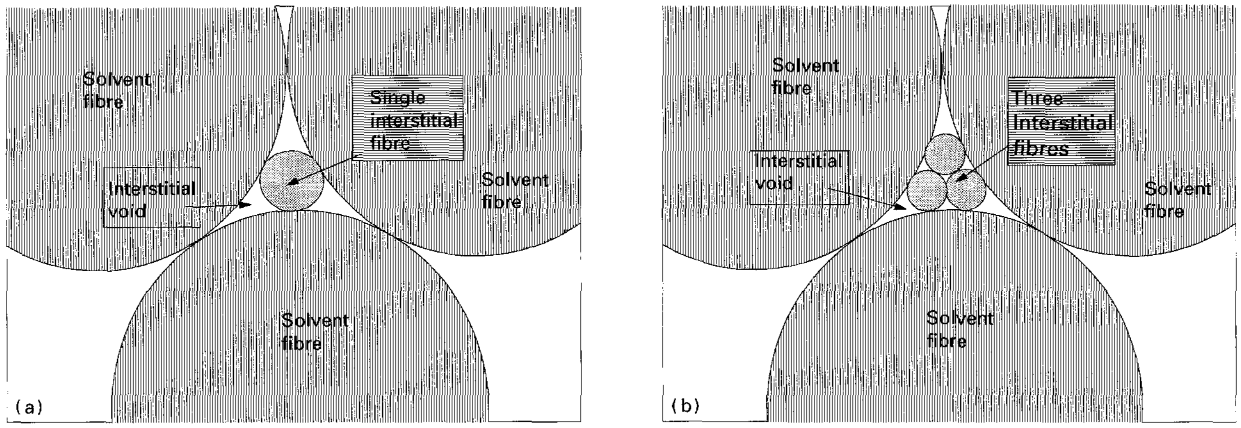

Figure 3 Diagram of interstitial fibres in close packed fibre array: (a) single fibre, (b) three fibres.

TABLE I Minimum size of interstitial voids and maximum diameter of interstitial fibres in stacks of continuous cylindrical solvent fibres. R = radius of solvent fibre

Type of stacking	Minimum interstitial void volume fraction and area, A	Single interstitial fibre		Multiple interstitial fibres	
		Maximum diameter	Volume fraction in void	Maximum diameter	Fibre volume fraction in void
Vertical stacking	21.5% $A = 0.86R^2$	0.83 R	63%	4 fibres each 0.41 R	4 fibres 63%
Close packed	9.3% $A = 0.16\,R^2$	0.31 R	47%	3 fibres each 0.22 R	3 fibres 71%

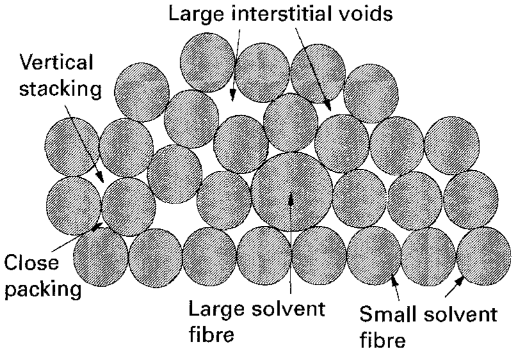

Figure 4 Effect on interstitial void size and shape of introducing a larger solvent fibre.

be used. For example, four smaller fibres of half this diameter, $0.41R$, also occupying 63% of the interstitial void volume (Fig. 2b).

The maximum fibre diameter that can be inserted into an interstitial void in a close packed fibre array is much smaller than for the vertical packed array, as shown in Fig. 3a. It has a diameter of $0.31R$ and would occupy 47% of the interstitial void volume. Alternatively, three smaller fibres of diameter $0.22R$ may be used (Fig. 3b), and they will occupy about 71% of the interstitial void volume. These data are summarized in Table I.

4. Substitutional solvent fibres

Again by analogy with atoms in solid solution, a fibre of different diameter may be substituted for a solvent fibre. The effect of a substitutional solvent fibre of larger diameter than the solvent fibres is shown in Fig. 4. As expected, the packing of the solvent fibres is seriously disrupted. Normal packing is only possible at some distance from the substitutional fibre and much larger interstitial voids are produced. This suggests that substitutional fibres of selected size may be used to generate areas containing larger interstitial voids for the introduction of larger diameter or greater numbers of interstitial fibres.

TABLE II Volume fractions of solvent and interstitial fibre in a continuous cylindrical fibre composite

		Volume fraction			
Type of stacking	Hybrid fibre combination	Solvent fibre	Interstitial fibre	Total fibre	Matrix
Vertical stacking	Solvent fibres only	78.5% maximum	zero	78.5%	21.5% minimum
	Solvent fibres plus single interstitial fibre per interstitial site	78.5%	13.54%	92%	8%
	Solvent fibres plus 4 interstitial fibres per interstitial site	78.5%	13.54%	92%	8%
Close packed stacking	Solvent fibres only	90.7% maximum	zero	90.7%	9.3% minimum
	Solvent fibres plus single interstitial fibre per interstitial site	90.7%	4.4%	95.1%	4.9%
	Solvent fibres plus 3 interstitial fibres per interstitial site	90.7%	6.6%	97.3%	2.7%

TABLE III Calculated maximum interstitial fibre diameters for two sizes of solvent fibre

Type of stacking	Solvent fibre radius, R (μm)	Maximum single interstitial fibre diameter (μm)	Maximum multiple interstitial fibre diameter (μm)
Vertical stacking	70.0	58	4 fibres 29
	50.0	41.4	4 fibres 20
Close packed stacking	70.0	21.7	3 fibres 15
	50.0	15.5	3 fibres 11

TABLE IV Properties of a commercial large-diameter fibre (solvent fibre) and potential interstitial fibres

Property	SiC fibre β-SiC	Nicalon NL-200 β-SiC	HI-Nicalon β-SiC	silica	Altex γ-Al_2O_3	Nextel 480 mullite	Fibre FP α-Al_2O_3	Tyranno amorphous
Supplier	(BP)	Nippon Carbon	Nippon Carbon		Sumitomo Chemicals	3M	DuPont	Ube Industries
Composition (wt %)	CVD SiC on 15 μm W-wire	Si–C–12%O	Si–C–0.4%O	SiO_2	85 Al_2O_3 15 SiO_2	70 Al_2O_3 28 SiO_2 2 B_2O_3	99%	A Grade Si–C–O < 5% Ti
Diameter (μm)	100	14/12	14	9	12/17	oval 10 × 13	19	11 or 8.5
Relative density	3.4	2.55	2.74	2.21	3.25	3.05	3.92	2.29
E (GPa)	400	220	270	74	210	220	385	200
Tensile strength (GPa)	3.75	2.8	3	4	1.8	2	1.6	3
Fracture strain (%)		1.4	1	5.4	0.85	1	0.4	1.8
TC λ ($W m^{-1} K^{-1}$)	110	12		11				
Specific heat ($J kg^{-1} K^{-1}$)	820	1,140		840				795/60 °C 1172 at 400 °C
CTE (10^{-6} °C^{-1})	4.5	3.1		0.54	8.8	4.38	7.6	3.1
Electrical resistivity (Ω m)	1.5×10^4	10^5–10^6	1.4×10^2	10^{12}				10^4–10^5
Maximum temperature (° C)		+ 1000	1500		1000	1000	1000	1500 after 1 h in air

5. Potential hybrid fibre MMCs

A consequence of introducing interstitial fibres into a metal matrix composite to produce an interstitial fibre composite (IFC) is that the volume fraction of fibres is increased at the expense of the matrix volume fraction. The minimum matrix volume fraction between solvent fibres is given in Table II, together with the respective solvent and interstitial fibre and matrix volume fractions in IFCs. The introduction of one large or four small interstitial fibres into the vertical stacked array leads to a reduction in the matrix volume fraction of about 60%. In a close packed array, one large or three small interstitial fibres reduces the matrix volume fraction by 47% and 71%, respectively. The interstitial fibre volume fraction varies from about 13% in vertical packing to about 4%–7% in close packing. Although this is small compared with the solvent volume fraction, it increases the total theoretical fibre volume fraction to about 97% (Table II) without affecting the spacing or distribution of the solvent fibre in the IFC.

The calculated maximum fibre diameters required to fit into interstitial sites in current commercial MMCs are listed in Table III for two solvent fibres of diameter 100 μm (typical of Sigma SiC fibre and Saphikon alumina fibre) and 140 μm (typical of Textron SiC fibre). The maximum interstitial fibre diameters are in the range 11–58 μm. The diameters and properties of a solvent fibre (Sigma SiC) and of some potential continuous interstitial ceramic fibres are summarized in Table IV. The latter have diameters in the range 9–19μm, making them suitable for single or multiple interstitial fibres.

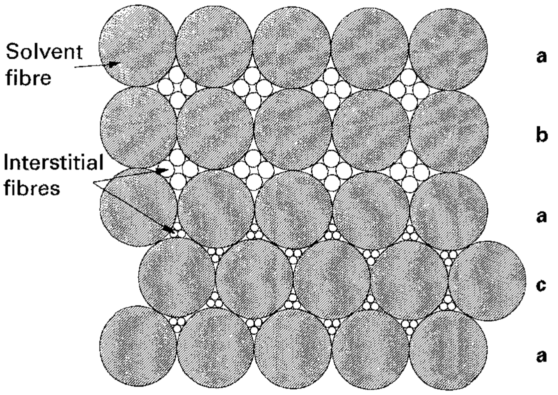

Figure 5 Schematic diagram of a section through a continuous interstitial fibre composite (IFC) showing the solvent and interstitial fibre distribution.

6. Discussion

The microstructure of a section through an IFC with the two types of packing is shown schematically in Fig. 5. The two-dimensional distribution of the solvent fibres and the interstitial fibre clusters in this section can be described by nets. In vertical stacking, the solvent and interstitial fibre clusters lie at the nodes of two superimposed square nets. In close packed stacking, the solvent and interstitial fibre clusters lie at the nodes of two superimposed face centred hexagonal and hexagonal nets, respectively. It is clear from comparing Figs 1 and 5 that the interstitial fibres have no effect on the solvent fibre net and that the interstitial fibre array in the IFC represents a new type of composite microstructure. Only maximum packing densitiy has been considered in this paper, but the principle of two superimposed nets is also applicable to IFCs with lower packing densities.

Many of the interstitial fibres listed in Table IV have good high-temperature properties and may be more resistant to degradation at elevated temperatures than the matrix they replace. The use of the very small diameter fibres such as Tyranno or carbon fibre, would allow many more fibres to occupy some interstitial sites. With interstitial fibres, the matrix-dependent toughness may be replaced by toughness dependent on crack deflection at the many interstitial fibre interfaces. The higher strain to fracture obtained with some fibres (Tyranno and C-fibres) and possible changes in the transverse fracture behaviour [5] may further enhance the toughness.

Manufacturing composites using the MCF technique and containing interstitial fibres depends upon coating a thin film of the matrix on these fibres and on the solvent fibres. The coating thickness must be sufficient to fill the remaining interstitial void space and to provide the required fibre spacing during consolidation. Success in coating continuous fibres has been reported using electron beam vapour evaporation and deposition and magnetron sputtering techniques [1]. Precise positioning of the fibres will be required during the layup stage. In addition to the possible improvements in mechanical properties, the use of interstitial fibres may be particularly useful when some changes in the physical properties such as electrical or thermal conductivity are required without affecting the properties obtained by the solvent fibres alone. Another advantage may be the high fibre volume fraction that can be achieved using interstitial fibres, which approach the properties of a high-density monolithic ceramic fibre composite.

7. Conclusion

In a stack of continuous cylindrical fibres in contact, one or more small-diameter (interstitial) fibres may be introduced into the interstices between larger diameter (solvent) fibres. For typical solvent fibres of about 100–140 μm diameter, the maximum interstitial fibre diameter is in the range 11–58 μm, which covers the size range now available in high-strength commercial ceramic fibres. In metal matrix composites, interstitial fibres can increase substantially the fibre volume fraction at the expense of the matrix. This may help to reduce adverse effects due to the matrix at elevated temperatures. Similarly, interstitial fibres may allow improvements in mechanical properties or changes in the physical properties such as electrical or thermal conductivity. These effects could be achieved without affecting the volume fraction or distribution of the solvent fibre.

Acknowledgements

The author thanks Drs C. M. Ward-Close and M. Pikethley, DRA(F), and Professors J. W. Steeds and M. N. R. Ashfold, University of Bristol, for many helpful discussions on the composite research programme, and EPSRC for financial support.

References

1. P. G. PARTRIDGE and C. M. WARD-CLOSE, *Int. Mater. Rev.* **38** (1993) 1.
2. C. M. WARD-CLOSE and P. G. PARTRIDGE, *J. Mater. Sci.* **25** (1990) 4315.
3. A. R. BUNSELL, (ed.), "Composite Materials", Vol. 2 (Elsevier, 1988).
4. J. R. BROCKENBROUGH, S. SURESH and H. A. WIENECKE, *Acta Metall.* **39** (1991) 735.
5. M. R. WISNOM and D. S. LI, Final contract, 2034/93 University of Bristol (1991).
6. J. F. DURODOLA and R. DERBY, *Acta Metall.* **42** (1994) 1525.

Received 19 January
and accepted 21 February 1995