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Short aluminosilicate fibre reinforced aluminium

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Characteristic of cheap ceramic alumina–silica (Sibral) fibres and their application in aluminium to produce composite material is given. Strengths and Young's modulus of short (average length of 148 μm) fibres are 0.8 GPa and 70 GPa, respectively and its density is 2600 kg m^{-3}. Composite materials Al–Sibral fibres were prepared by pressure infiltration of aluminium into the preform made with SiO_2 as bonding phase (by sol–gel method). Sibral fibres increase tensile strength and wear resistance of the composite. These fibres significantly reduce that coefficient of thermal expansion. Results from the interaction between fibres and matrix as well as from the fracture of composite are presented and mechanism of the interaction zone (α-Al_2O_3 in crystalline form) formation is discussed.

1. Introduction

Metal matrix composites (MMCs) reinforced by ceramic whiskers, short fibres, platelets or particulates are potential candidates for various applications and are moving to a stage of large scale production. Ceramic fibres are added to metal matrices to increase one or more of the following properties: specific strength, elastic modulus, thermal fatigue resistance, wear resistance or maximum temperature of use. The main disadvantage of today's MMCs development is the high price of these materials compared to conventionally produced materials. One of the advantages of such MMCs is that billets of the composites can be mechanically processed using the technologies developed for conventional materials (forging, extrusion). However, the performance of composites depends more than conventional alloys on the processing route. This is because a more complicated response to thermal and mechanical loading is involved as well as the large differences in thermal and elastic properties between the matrix and reinforcement. The properties of MMCs depend on matrix properties, especially when the fibre content is low or when reinforcement is achieved with short fibres or particles instead of continuous fibres. Of the many potential metal matrix systems, aluminium alloy matrix composites have been the object of much research, primarily due to the light weight, low cost and ease of fabrication of aluminium. Discontinuously reinforced aluminium alloys have been fabricated by various means, including solid state processes, such as powder metallurgy techniques (blending of metal and ceramic powders followed by hot pressing) [1] and liquid state processes, such as compocasting (blending ceramic powders or short fibres and molten aluminium, agitating and casting) [2, 3] and pressurized liquid–metal infiltration [4]. The solid state processes have been most successful to date, but are costly. Liquid–metal processes have the potential to be more economical, however the non-wetting nature of many ceramic metal interfaces and incomplete infiltration has been an obstacle [5]. For high temperature application usually ceramic aluminosilicate fibres (Al_2O_3–SiO_2 system) are used. From the Al_2O_3–SiO_2 system there are five fibre types mentioned in Table I. Chi [6] evaluated mechanical properties of Standard Oil's low-cost aluminosilicate reinforcement grade fibre Fiberfrax in aluminium. They demonstrated that the tensile and fatigue properties of aluminium alloys can be significantly improved by incorporating Fiberfrax fibres by squeeze casting. Extremely poor wear resistance is often the major drawback in wider utilization of aluminium and its alloys as the machine parts. It has been shown in previous investigations that the incorporation of hard ceramic reinforcements and soft lubricants into an aluminium alloy can improve the wear resistance [7]. The wear resistance of an aluminium alloy can be also greatly improved by incorporating as low as 5 vol % of the Fiberfrax fibres. The purpose of this work is to evaluate some properties of very cheap aluminosilicate Sibral fibres and to prepare composite Al–Sibral fibres by a liquid–metal infiltration technique. Their characteristic properties, tensile strength, Young's modulus, wear resistance, coefficient of thermal expansion and compatibility of its components are studied.

2. Experimental procedure

2.1. Fibre material and processing

The discontinuous ceramic aluminosilicate Sibral fibres with composition of 45–48 wt % of alumina and 51–54 wt % of silica were used as reinforcement materials.

For determination of tensile strength and Young's modulus of Sibral fibres longer monofilaments ($\approx$ 60 mm) were used taken from Sibral wool fibre. Tensile strength tests of Sibral fibres used in this study

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TABLE I Properties of various ceramic fibres from the Al_2O_3–SiO_2 system

Property	Fiberfrax	Fiberfrax (Sohio)	Sibral	Fibermax (mullite)	Altex (Sumitomo)
Compostion					
(wt %) Al_2O_3	49.2		47	72	85
SiO_2	50.5		53	27	15
Structure	amorphous	amorphous	amorphous	crystalline	crystalline
Mean diameter (μm)	2.5	2–5	5.4	3.0	15
Mean length (μm)	762	–	148	762	continuous
Density (kg m^{-3})	2700	2600	2600	3000	3300
Tensile strength (GPa)	1.7	1.0	0.8	0.8	1.8
Young's modulus (GPa)	105	105	73	150	222

was similar to ASTM D-3379. Monofilaments were glued on to a paper frame by epoxy resin. After clamping into the thread testing machine FM 27, the paper edges were cut off and the monofilament was tensile stressed at a cross-head speed of 0.5 mm min^{-1}. The load at fibre fracture was recorded with precision of ± 1%. The gauge length used was 20 mm. Fibre diameter was measured for each fibre at a magnification of × 1000 using a Neophot optical microscope. The fibres initially contained from about 20 to 50 wt % of shots (not fibre-like particles) which for the purposes of measuring and preform preparation were removed by a sedimentation process. Lengths of the chopped fibres were measured from their microphotographs by means of digital measuring table connected to an Atari computer. The density of the fibres was measured by a pycnometric method. The quality of fibres from the point of view of crystallinity was tested by X-ray powder diffraction method using CuK_α radiation (Fig. 1). Due to production process the aluminosilicate (Sibral) fibres are amorphous with a small amount of crystalline alumina.

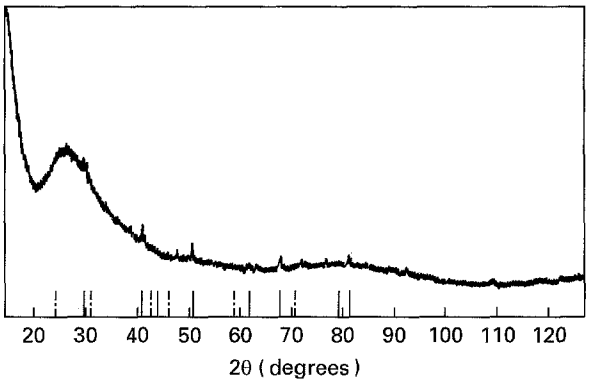


Figure 1 X-ray diffraction pattern of Sibral fibre (--- corundum, – – – quarz).

2.2. Composite fabrication

For the fabrication of the composite material sample, a gas pressure metal infiltration process was used. This technique requires a porous preform which was manufactured in-house from chopped aluminosilicate ceramic Sibral fibres. The method of preparing the preform involves dispersing the fibres in water to remove agglomerates of fibres, sedimentation of the fibres, filtration and then drying in air at about 393 K. After drying the bonding phase was introduced into fibre filtration cake. The bonding phase employed was SiO_2 gel synthesized by acid-catalysed hydrolysis of an ethanolic solution of tetraethoxysilane. The advantage of the sol–gel process for introducing a bonding phase is that the fibres can be impregnated with a liquid medium, so that the homogeneity and quantity of the deposit can be controled. Thermal treatment of samples at 873 K was used to burn off the organic portion of the binder, prior to introduction in the autoclave. The preforming of the fibre reinforcement guarantees an equal distribution and a known fibre volume fraction. The quantity of fibres in preform specimens (volume fraction V_f) varied from 0.096 to 0.245. The preforms prepared in this study were rectangular prisms 40 mm in length, 10 mm wide and 4–11 mm in height. The fibre volume fraction in specimens was controlled by pressing the prisms cut out from filtration cake with a base of 10 × 40 mm after adding the bonding phase up to the height calculated from the fibre's weight. The preform fabrication operation tends to align the fibre axes normal to the pressing direction, resulting in a two-dimensional random arrangement of fibres. The samples were prepared in an autoclave equipped with an induction melting furnace. After evacuation, pressure was applied through an influx of argon gas. The preforms were infiltrated at 973 K, at argon pressures of 4.0, 5.0 and 5.6 MPa, for 10 to 60 s. The metal used was aluminium (99.8%) the tensile strength of which was 65 MPa.

2.3. Microstructural characterization

The samples were prepared for microstructural observation by wet cutting on low-speed saw with a diamond wheel, by mechanical grinding and polishing. Composite microstructures were studied by optical microscopy, scanning (SEM) and transmission (TEM) electron microscopy. The thin foils were prepared by ion milling using a ion beam milling system RES 010. The samples were examined by means of a Jeol JEM 100C electron microscope using selected electron diffraction microscopy. The fracture surfaces from the tensile test were examined under SEM.

3. Results

3.1. Optimal microscopy

Optical microscopy was carried out on selected MMC specimens to examine qualitatively the distribution of the fibre reinforcement. The structure of the MMCs was typical of composites fabricated by the pressure infiltration of fibre preforms. During preforming the fibres are laid down in a mat-like structure and this results in a two-dimensional planar random morphology of fibres. In addition there was some clumping of fibres which gave micro-variation in fibre density ranging from high to low or even zero fibre regions. This structure and the local variation in fibre distribution are illustrated in Fig. 2. There can also be seen isolated particles which are not fibre-like.

3.2. Scanning electron microscopy

Fig. 3 shows scanning electron micrographs of Sibral fibres from the raw material (to be made into infiltration cake). Figure shows that the fibres were quite random in orientation. Fig. 4 shows scanning electron micrograph of the rectangular preform ($10 \times 40 \times 5$ mm) with 16.8 vol % fibres at the sections at a distance of 1 mm from the preform surface. The photograph of Fig. 4 is representative of all parts of the preform other than the surface. A much higher concentration of binder was present in surface area than in area shown on Fig. 4. Thus the binder mainly resided at the surface of the preform, so that the interior region had a much lower binder concentration. The origin of this effect is believed to be related to the transport of the binder toward the edge as the alcohol evaporated from the binder.

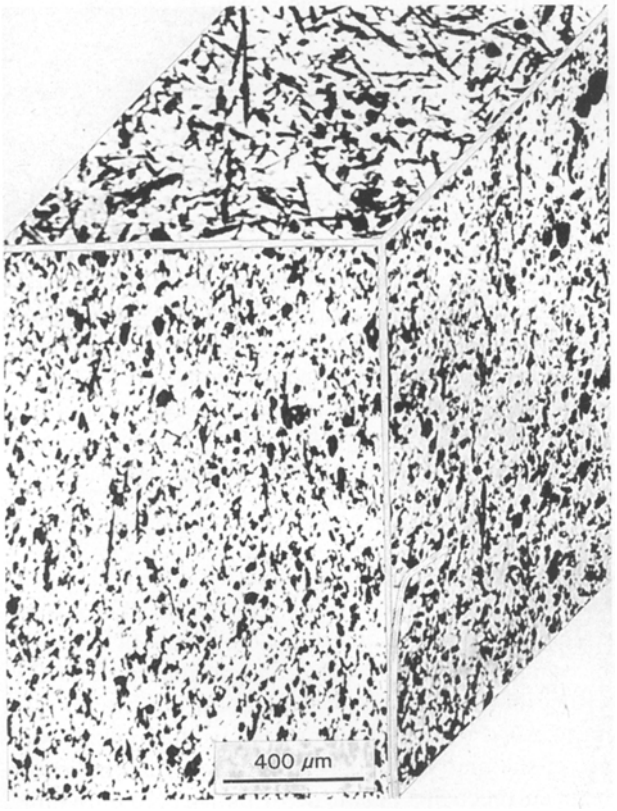


Figure 2 Microstructure of Al–9.6 vol % Sibral fibres composite fabricated by pressure infiltration.

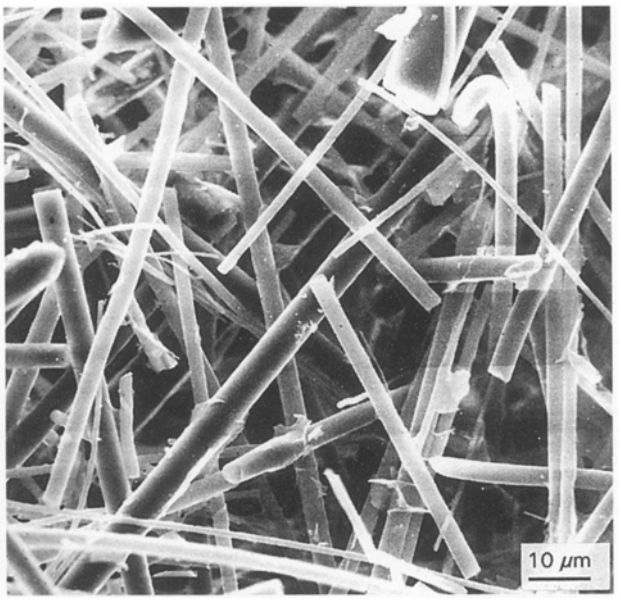


Figure 3 Electron micrograph of Sibral fibres from the raw material.

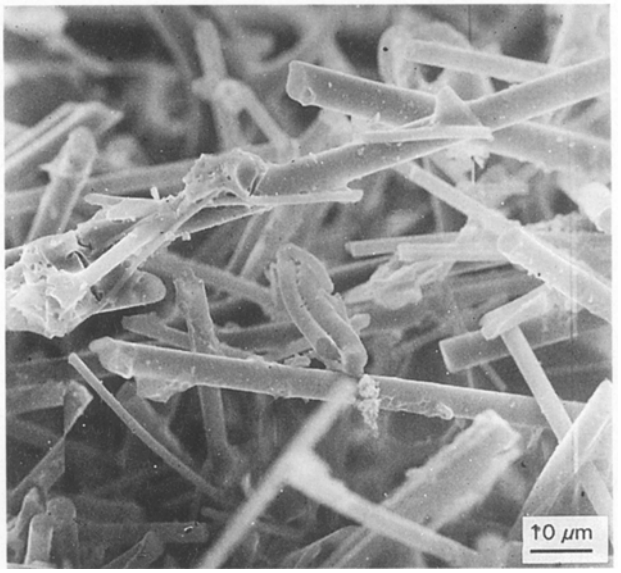


Figure 4 Scanning electron micrograph of Sibral fibres preform ($V_f = 0.168$) at a distance of 1 mm from the surface.

3.3. Fibre characterization

The diameter and length distribution of chopped Sibral fibres are shown in Fig. 5(a) and (b). The mean diameter and mean length of the fibres were 3.4 and 148 μm, respectively. In Fig. 6(a) and (b) tensile strength and Young's modulus distribution of Sibral fibres are shown. The elastic modulus of Sibral fibres is very close to the modulus of aluminium (68.5–73.8 GPa). From this statement it is clear that for improvement in elastic modulus of the Al alloys the Young's modulus of Sibral fibres is too low. The tensile strength of Sibral fibres (0.8 GPa) is about ten times higher than that of Al which would suggest a potential for increasing the strength of the Al

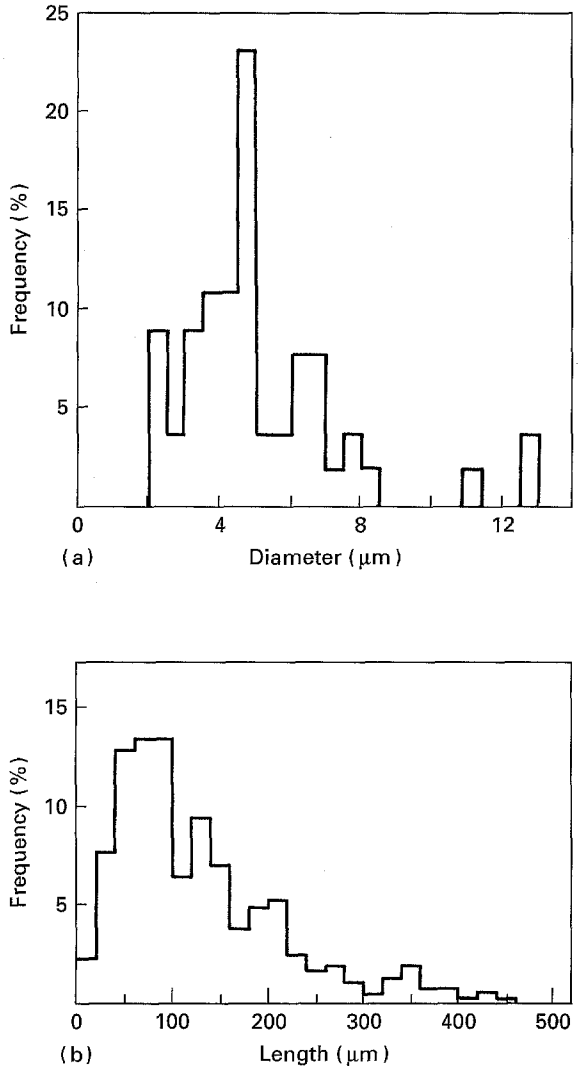


Figure 5 Distribution of the diameter (a) and length (b) of chopped Sibral fibres.

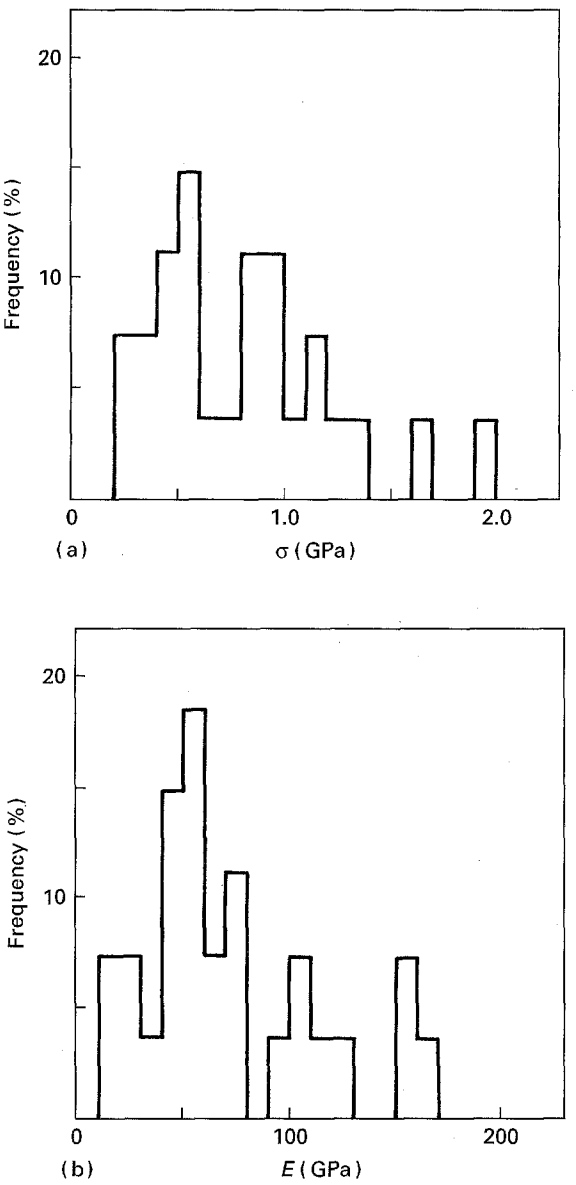


Figure 6 Distribution of the tensile strength (a) and Young's modulus (b) of Sibral fibres.

composite. This is valid only for continuous fibres. For discontinuous fibres, however, the strengthening of Al could be attained using higher volume fraction of fibres. The density of Sibral fibres is 2600 $kg\,m^{-3}$. Sibral fibres were tested by differential thermal analysis (DTA) in the temperature range 773–1673 K (500–1400 °C). At 1253 K (980 °C) a sharp exothermic peak was observed. This peak, after Zhou Yaomin *et al.* [8], corresponds to mullite formation. Expect for this peak, no other changes in DTA analysis were observed. This means that Sibral fibres are also stable at the temperature of composite material preparation (973 K).

3.4. Tensile properties of composite material

Tensile strength at room temperature was performed on an Instron tensile testing machine at a constant cross-head speed of 0.2 $cm\,min^{-1}$. An electronic extensometer with gauge length of 10 mm (magnification of × 1250) was connected to the specimen in order to record the exact strain in the first part of the stress–strain curve. Young's modulus at room temperature was determined from the linear part of stress–strain curve recorded during tensile testing of composite. Fig. 7(a, b) show the variation of the tensile strength and Young's modulus with the volume fraction of fibres, respectively. As shown in this figure strength and modulus at room temperature are increasing, compared with base metal. The strength of this material containing 24.5 vol % of fibre is greater than that of pure aluminium by about 100%.

3.5. Fracture surfaces

The fracture surfaces of all tensile specimens were examined in order to determine the nature of failure of the composite. The surface were macroscopically brittle in appearance. Examination of the fractographies led to the following results: the fracture surface of the matrix is slightly dimpled which indicates the occurrence of a plastic deformation. No fibre pull-out phenomenon was observed. Numerous fibres were fractured longitudinally (Fig. 8). It is apparent from

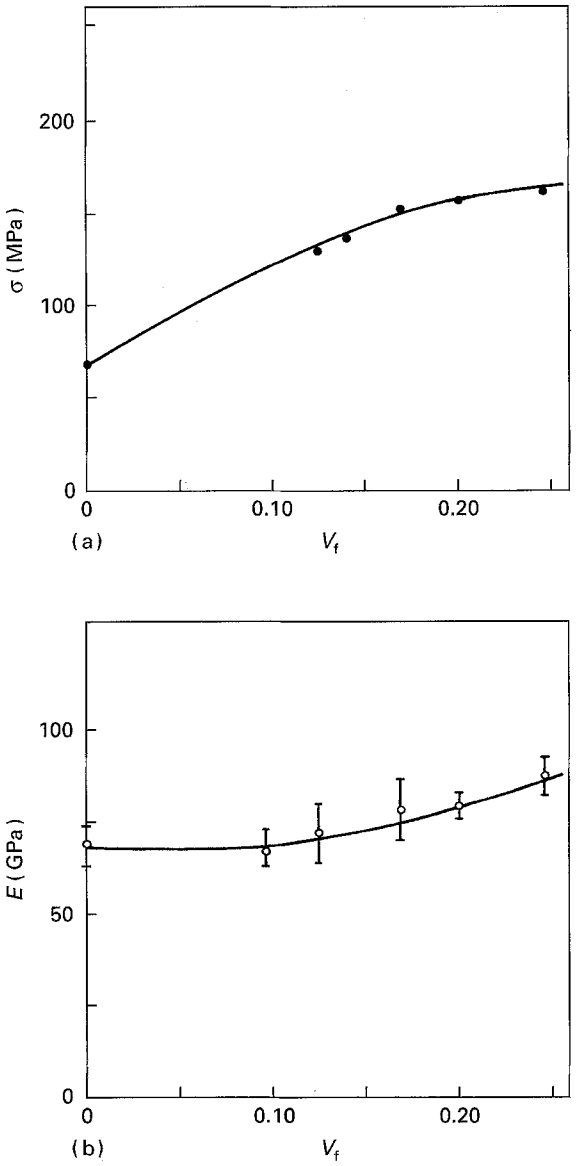


Figure 7 Variation of the tensile strength (a) and Young's modulus (b) with volume fraction (V_f) of Sibral fibres in composites.

the previous remarks that the fibre–matrix bonding is very strong, allowing an efficient load transfer.

3.6. Wear

The abrasive wear tests were carried out at room temperature on an apparatus similar to that used by Nathan and Jones [9]. All experiments were carried out under dry conditions, sliding the samples for 2 min under relative speed of 0.1 m s^{-1} and applied load 5 N, on a number 120 abrasive paper. Prior to, and after, each measurement, the specimen was cleaned and weighed with a precision of 0.01 mg. The rate of wear of investigated samples of composite materials containing 9.6–24.5 vol % Sibral fibres have been plotted as a function of volume fraction of fibres in Fig. 9. The results indicate that rate of wear of the fibre reinforced samples increases with content of fibres. The rate of wear of the fibre reinforced sample containing 24.5 vol % is reduced by 30% compared with aluminium.

Figure 8 SEM of the fracture surface of composite containing 20.0 vol % Sibral fibres.

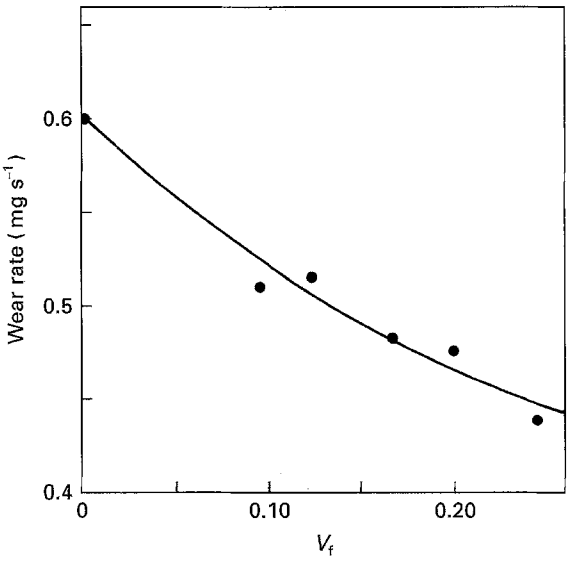


Figure 9 Effect of volume fraction of reinforcement on wear behaviour.

3.7. Thermal expansion

Materials with tailorable coefficients of thermal expansion (CTEs) can be obtained with composite materials. As shown by Turner [10], the CTE is a function of the CTEs, the bulk moduli and the volume fraction of the components.

$$\alpha_C = \frac{\alpha_m V_m E_m + \alpha_f V_f E_f}{V_m E_m + V_f E_f}$$

where α is the CTE, V is the volume fraction, E is Young's modulus and the subscripts c, m and f refer to the composite, matrix and fibre, respectively. Generally, the thermal expansion of a composite is

dominated by the component with the highest value of bulk modulus. In this case, where elastic modulus of aluminium and that of Sibral fibre are approximately the same, the CTE of the composite depends only on the fibre fraction in the composite and on the CTE of the components. The CTE of Al–Sibral composite was measured with dilatometer on rectangular specimens $2 \times 9.5 \times 35$ mm containing 9.6–24.5 vol % short Sibral fibres. The specimens were heated to 600 K and then without any delay cooled to room temperature (300 K). This cycle was repeated twice in order to eliminate the influence of differing thermal histories of the investigated materials. All testing was done in argon. In Fig. 10 the dependence of CTE on fibre volume fraction in temperature interval 300–600 K can be seen.

The dotted line represents the theoretical dependence of CTE of composite after Turner's expression and the solid line gives the experimental results. The addition of 24.5 vol % Sibral fibres into the Al matrix results in almost 35% CTE reduction compared to the initial value of CTE of Al (24.3×10^{-6} K^{-1}). The experimental results show that decrease of CTE with fibre volume fraction is larger than that from Turner's relation.

For calculation of theoretical dependence of CTE on V_f (dotted line, Fig. 10) the partial transformation of Sibral fibres to crystalline Al_2O_3 during the composite preparation (discussed elsewhere) was not taken into account. Young's modulus of Al_2O_3 fibres [11] is approximately three times higher than that of Sibral fibres. This higher value of Young's modulus can lower the CTE of a composite in Turner's relation. It is possible therefore to explain quantitatively the experimental results in terms of the change in the Sibral fibres.

3.8. Compatibility of the fibres and the matrix

The local chemistry of the interface can have a major impact on strength of the bonding at the interface. Differences in the coefficient of thermal expansion across the interface also have an impact on the mechanical response of the composite. Many researchers have investigated the reaction between molten aluminium and alumina [12–14]. At temperatures below 2000 K the reaction has been reported as

$$4Al(l) + Al_2O_3(s) = 3Al_2O(g) \quad (1)$$

Eustathopoulos *et al.* [13] reported that for this reaction the partial pressure of the volatile Al_2O is approximately 1.3×10^{-3} Pa at 1133 K. The reaction (Equation 1) does not give rise to any stable bonding between the Al and alumina fibres. The interface reaction between Al and SiO_2 (also contained in the Sibral fibres) can be expressed as

$$4Al(l) + 3SiO_2(s) = 2Al_2O_3(s) + 3Si(s) \quad (2)$$

This reaction is thermodynamically possible because the free energy change is negative between 200 and 2000 K [14].

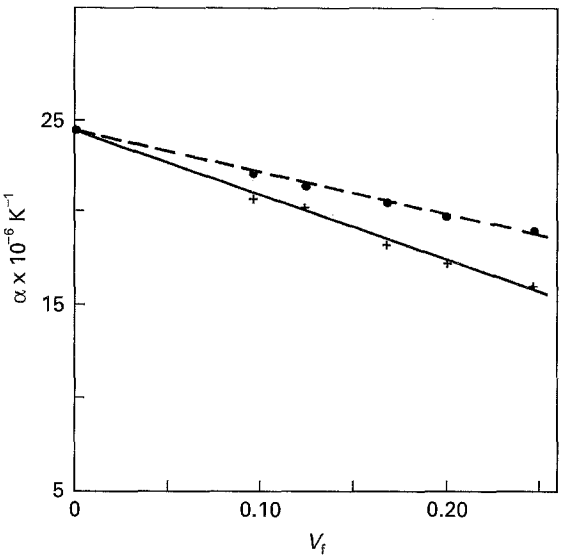


Figure 10 Dependence of CTE on fibre volume fraction in temperature interval 300–600 K. --- theoretical; —— experimental.

Figure 11 Optical micrograph showing the reaction zone in Sibral fibres.

In the process of Al–Sibral fibre composite material preparation the amorphous fibre reacts with aluminium (Fig. 11). The concentration profile (Fig. 12) across the fibre and fibre–matrix interface indicates that in the inner (amorphous) part of the fibre there is no change in the composition. In the outer part of the fibre due to interaction of the fibre with aluminium, crystalline aluminium oxide is formed. Silicon diffuses from this crystallized zone to the periphery of the fibre and aluminium diffuses into the fibre, reacting with oxygen from silica and gradually making alumina (α-Al_2O_3). This is confirmed by electron diffraction

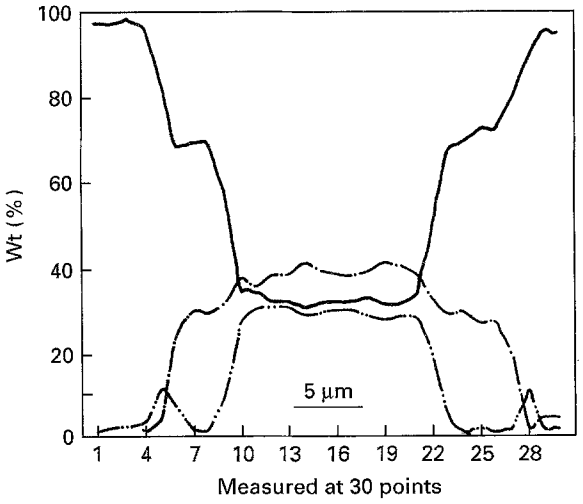


Figure 12 Concentration profiles of Si, Al and O across the boundary. —— Al; —·— O; —··— Si.

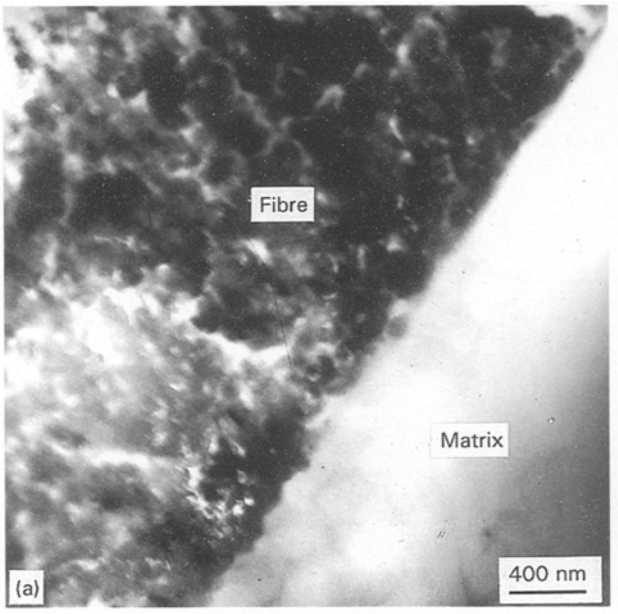
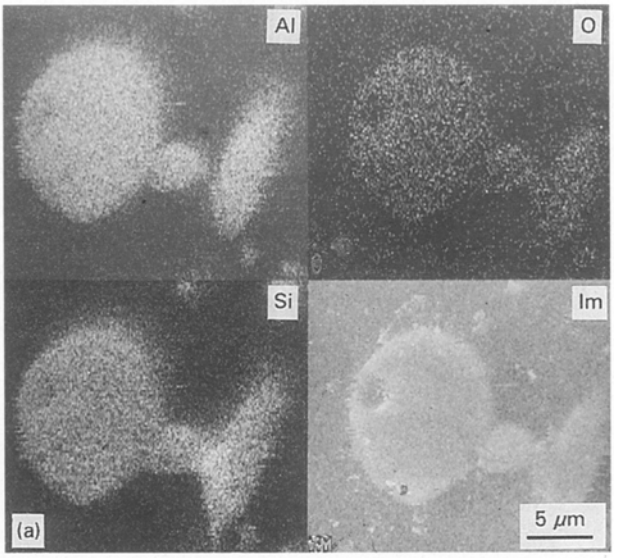


Figure 13 TEM of Al–Sibral fibres composite (a). Electron diffraction pattern (b).

from TEM of the foil of Al–Sibral fibre composite (Figs 13(a, b)). Electron diffraction is taken from the interface of fibre and matrix using the matrix data for the evaluation of diffraction of the fibre. This is also



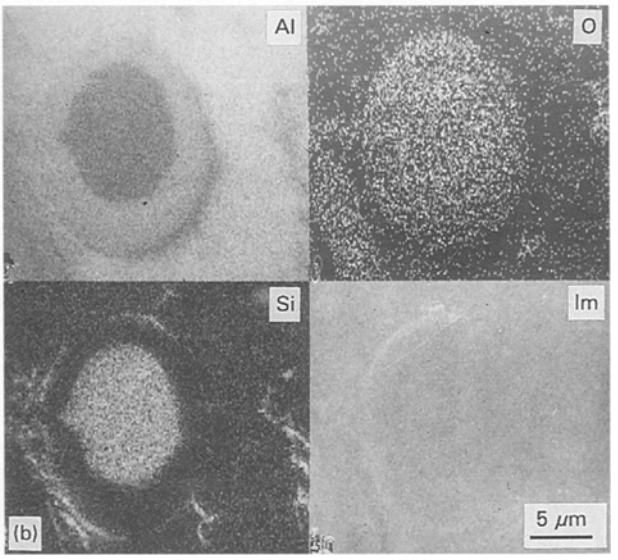


Figure 14 Two-dimensional dot maps of Al, Si and O on polished cross-section of the original Sibral fibres (a) and composite material with Al matrix (b).

confirmed comparing two-dimensional scans of Al, Si and oxygen on polished cross-sections of the original Sibral fibres (Fig. 14(a)) and composite material with Al matrix (Fig. 14(b)). Silica acts as an oxygen source which causes oxidation of liquid aluminium. The reaction zone thus consists of the crystalline form of α-Al_2O_3. The thinner fibres are, at the given conditions, all transformed to crystalline Al_2O_3; the thicker fibres also consist of some amorphous silica and alumina. The results obtained can be explained by the reaction mechanism Equation (2).

4. Conclusions

With the results obtained from this investigation the following conclusions are drawn.

1. It was demonstrated that the tensile properties of aluminium can be mildly improved by incorporating low cost aluminosilicate Sibral fibres.

2. The wear resistance of the aluminium matrix can be improved by incorporating Sibral fibres. This effect increases with the volume fraction of fibre, and for 24.5 vol % fibre the wear resistance increased by 30%.
3. The incorporation of Sibral fibres into aluminium matrix brings about a significant reduction of the thermal expansion coefficient (35% for Al–24.5 vol % Sibral fibres).
4. At the processing temperature the reaction between molten aluminium and solid SiO_2 which is contained in the Sibral fibres was verified and is governed by the reaction of (Equation 2.)

Acknowledgements

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References

1. I. J. TOTH, W. D. BRENTNALL and G. D. MENKE, *J. Mater. Sci.* **24** (1972) 19.
2. J. W. MCCOY, C. JONES and F. E. WARNER, *Sampe Q.* **19** (1988) 37.
3. S. DAS, T. K. DAN, S. V. PRASAD and P. K. ROHATGI, *J. Mater. Sci. Lett.* **5** (1986) 562.
4. S.-Y. OH, J. A. CORNIE and K. C. RUSSELL, Ceram. Engng. Sci. Proc. **8** (1987) 912.
5. G. P. MARTINS, D. L. OLSEN and G. R. EDWARDS, *Met. Trans.* **19B** (1988) 95.
6. F. K. CHI, R. D. MAIER, T. W. KRUCEK and P. M. BOYMEL, in Proceedings of the 6th ICCM and 2nd ECCM conference on composite materials, London, July 1987, edited, by F. L. Mathews, N. C. Buskell, J. M. Hodgkinson and J. Morton, (Elsevier Applied Science Publishers Ltd., London, 1987) Vol. 2, p 2. 449.
7. C. S. LEE, Y. H. KIM and K. S. HAN, *J. Mater. Sci.* **27** (1992) 793.
8. ZHOU YAOMIN, LI XIANGAN, CHEN MEIYI, *J. Mater. Sci.* **29** (1994) 1707.
9. G. K. NATHAN and W. J. D. JONES, *Wear* **9** (1966) 300.
10. P. S. TURNER, *J. Res. Natn. Bureau Standards USA* **37** (1946) 239.
11. U. MADELENO, H. LIU, T. SHINODA, Y. MISHIMA and T. SUZUKI, *J. Mater. Sci.* **25** (1990) 3273.
12. L. F. MONDOLFO, "Aluminium alloys: structure and properties" (Butterworths, London, 1976) p. 344.
13. N. EUSTATHOPOULOS, J. C. JOUD, P. DESRE and J. M. HICTER, *J. Mater. Sci.* **9** (1974) 1233.
14. K. J. BRONDYKE, *J. Amer. Ceram. Soc.* **36** (1953) 171.

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