JOURNAL OF MATERIALS SCIENCE 26 (1991) 6453–6457

Discontinuous metallic-glass ribbon reinforced glass–ceramic matrix composites

R. U. VAIDYA, K. N. SUBRAMANIAN
Department of Metallurgy, Mechanics and Materials Science, Michigan State University, East Lansing, Michigan 48824-1226, USA

Discontinuous metallic-glass ribbons of varying lengths and widths were used to reinforce a brittle glass–ceramic matrix. The fracture strength and toughness of such composites as a function of ribbon volume fraction and geometry were measured by three-point bending. The mechanical properties were found to be relatively isotropic in the plane of compaction (without significant loss of strengthening achieved with unidirectional reinforcement). The higher composite strength exhibited in a direction perpendicular to the plane of compaction was attributed to the higher percentage of ribbons oriented with their short transverse faces perpendicular to the opening crack front.

1. Introduction

The potential of using metallic-glass ribbons as reinforcements for brittle glass–ceramic matrices has been demonstrated [1–5]. Such composites were produced by relatively simple, and cost effective, pressing and sintering techniques. The studies carried out so far have indicated that significant improvements in the mechanical properties of the glass–ceramic matrices can be achieved by the use of relatively small volume fractions of metallic-glass ribbon reinforcements.

Some of the advantages of metallic-glasses which make them attractive as reinforcements is their high strength, toughness and corrosion resistance. The high cooling rates required in their manufacture imparts the ribbon geometry. This ribbon geometry not only provides for a large surface area to bond with the matrix, but has also been found to be useful in improving the off-axis properties, when used as reinforcements for brittle polymer matrix composites [6, 7].

The studies carried out so far [1–5] have focussed on the reinforcement of brittle glass–ceramic matrices with continuous metallic-glass ribbon reinforcements. These continuously reinforced composites are, however, unsuitable for structural applications, because of the high degree of directional strengthening they provide. Most structural applications demand a good degree of strength isotropy. Fabrication of these continuously reinforced composites is relatively difficult (since they involve ribbon lay up, etc.). The extremely small thickness of the ribbons also makes it difficult to incorporate large volume fractions of reinforcements into the matrix. In order to overcome these problems, discontinuously reinforced metallic-glass ribbon/glass–ceramic matrix composites were fabricated. The mechanical properties of these composites were measured and compared with those of continuously reinforced composites. The load transfer behaviour in such composites was also studied, and compared to the theoretical model proposed earlier [8].

2. Experimental procedure

Corning glass 7572 and Metglas 2605S-2 were used as the matrix and reinforcement, respectively. Details of the physical properties and chemical compositions of the matrix and reinforcement are provided elsewhere [1].

Three different ribbon dimensions were used in the study, namely $0.5 \times 0.5\ \text{cm}^2$ (length × width), $1.0 \times 0.5\ \text{cm}^2$ and $1.0 \times 0.25\ \text{cm}^2$. The ribbon thickness was kept constant (25 μm). The ribbons were precoated with the glass powder prior to composite fabrication. The precoated reinforcements were mixed with the glass powder (containing 8 to 10% by weight of amyl acetate binder). The ribbons were distributed as randomly as possible so as to prevent the ribbons from orienting preferentially with respect to one particular face (Fig. 1). Pressing was carried out at 3000 p.s.i. in a steel die. After compaction, the specimens were heated to 250 °C for 4 to 5 hours to drive off the organic binder. Sintering was carried out at 400 °C for 3 h and was followed by a crystallization step at 450 °C for 30 min in order to convert the matrix into a glass–ceramic.

The specimens were cut from the fabricated rectangular bars, and were tested in three-point bending in an Instron machine, using a cross-head speed of $0.05\ \text{cm min}^{-1}$. The three-point bend tests were

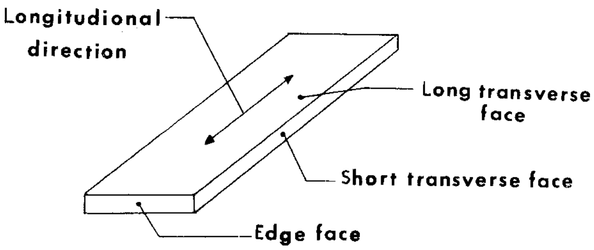

Figure 1 Illustration of a ribbon reinforcement indicating the terminology used to represent the various surfaces and directions.

0022–2461/91 $03.00 + .12 © *1991 Chapman & Hall*

carried out in accordance with ASTM Standard C-203/85. Fracture toughness measurements were carried out using single-edge notched beam specimens (in three-point bending). The tests were carried out in accordance with ASTM STP 678. A total of three to five specimens were used to obtain the average value for each data point. In all the specimens failure occurred at the midpoint.

3. Results and discussion

The results of the experimentally measured bending strength as a function of increasing volume fraction of reinforcements (for different ribbon dimensions) are given in Table I. From the data it is evident that increasing the volume fraction of reinforcements increases the strength. The experimentally measured strength of the discontinuous composites was about 80% of the strength of the continuously reinforced composite having the same volume fraction of reinforcements. The strength of the discontinuously reinforced composites can be compared to the theoretical composite strength (continuously reinforced) in Fig. 2.

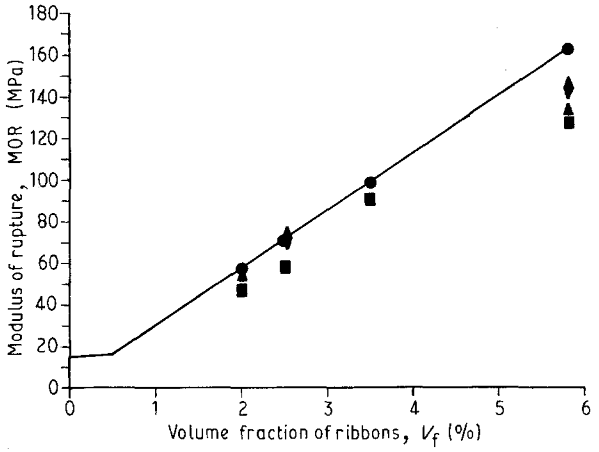

Figure 2 Plot of the composite bending strength as a function of increasing volume fraction of metallic-glass reinforcements. The solid line drawn through (●) indicates the theoretically predicted composite strength (using $\sigma_c^* = \sigma_f^* V_f$). The other legends represent three different ribbon dimensions as (■) length 0.5 cm, width 0.5 cm, (▲) length 1.0 cm, width 0.5 cm, (◆) length 1.0 cm, width 0.25 cm. Ribbon thickness was 25 µm.

The strength of the composite was measured in three different directions; longitudinal denoted by 'L', transverse denoted by 'T' and in a direction perpendicular to the plane of compaction denoted by 'CT'. These three orientations are shown in Fig. 3. All the specimens were cut from the same block of composite material in order to avoid any discrepancies induced due to processing. The strength values obtained for the three different orientations are given in Table II. The strength of the composite in the transverse direction (T) is about 95% of that in the longitudinal direction (L), which in turn is about 80% of the strength of a continuously reinforced composite in the longitudinal direction. From the strength values given in Table II it is evident that the composite exhibits strength isotropy in the plane of compaction. These results are illustrative of the advantages provided by ribbon reinforcements over fibres. Continuous fibre-reinforced composites are extremely weak in the transverse direction (5 to 25% of the longitudinal strength). This drawback is overcome by using multiple plies (containing ribbons oriented in different directions), but at the expense of a reduction in the strength in the longitudinal direction. On the other hand, continuous ribbon reinforced composites have been shown to exhibit transverse strengths as high as 50% of their longitudinal strengths [7]. By using discontinuous ribbon reinforcements of appropriate dimensions, an even higher degree of strength isotropy can be obtained.

In addition to the ribbon geometry, the strength of the bond between the ribbon and the matrix is also significant. Studies carried out on metallic-glass ribbon–polymer matrix composites (which exhibit relatively weak interfacial bonding) [6, 7], and Nichrome ribbon–slide glass matrix composites [5] have indicated a lower degree of strength isotropy as

TABLE I Experimentally measured composite strength as a function of ribbon volume fraction, for three different ribbon dimensions

Volume fraction of ribbons (%)	Ribbon dimensions		(l/w)	Average composite fracture strength (MPa)	Variance (%)
	length, l (cm)	width, w (cm)			
5.8	0.5	0.5	1	127.75	6.8
5.8	1.0	0.5	2	141.99	8.4
5.8	4.0[a]	0.5	8	162.4	
5.8	1.0	0.25	4	132.12	10.5
3.5	0.5	0.5	1	91.5	3.7
3.5	4.0[a]	0.5	8	98.0	
2.5	0.5	0.5	1	58.37	2.7
2.5	1.0	0.5	2	73.41	6.9
2.5	4.0[a]	0.5	8	70.0	
2.0	0.5	0.5	1	47.18	5.5
2.0	4.0[a]	0.5	8	56.0	
2.0	1.0	0.25	4	55.0	11.8

[a] Indicates ribbon length for a continuously reinforced composite, based on maximum die size.

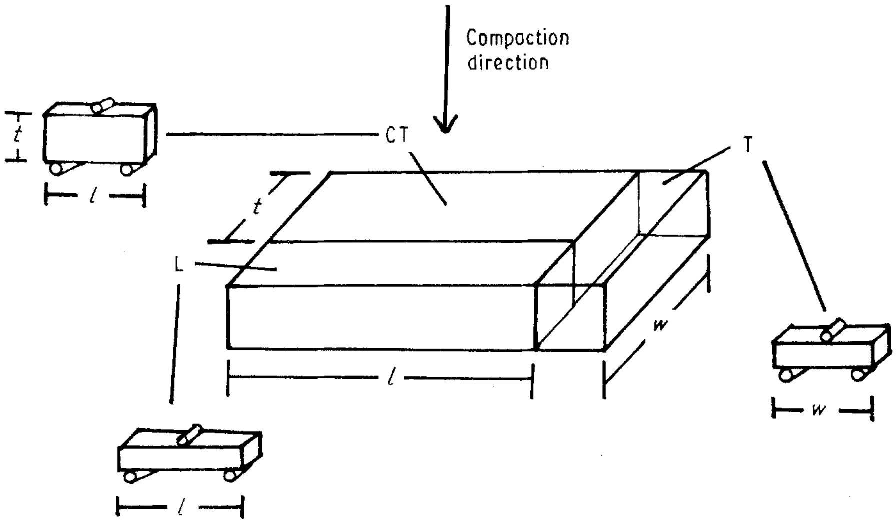

Figure 3 Figure illustrating the various orientations in which the composite specimen was tested.

TABLE II Experimentally measured composite strength in three different directions. The three directions used are identified in Fig. 2. Volume fraction of ribbons 5.8%, dimensions of ribbons length 0.5 cm, width 0.5 cm, thickness 25 μm, theoretical composite strength 162.4 MPa

Direction	Average composite fracture strength (MPa)	Variance (%)
L	132.3	5.8
T	125.8	10.1
CT	146.2	3.8

compared to the system investigated. Pullout tests carried out on the metallic-glass/glass–ceramic system have indicated the presence of a very strong bond between the matrix and the ribbons. The nature of this bond can be observed in Fig. 4. This strong nature of the bond is primarily responsible for the high level of strength utilization of the metallic-glass ribbons, and also the isotropy in the system.

Thus the high transverse strength and in-plane isotropy in such metallic-glass ribbon/glass–ceramic matrix composites can be attributed to three important factors.

1. High strength of the metallic-glass ribbons,
2. Unique ribbon geometry, and
3. Good bonding between the metallic-glass ribbons and the matrix.

Specimens with the 'CT' configuration exhibited the maximum strength. Although the ribbons were distributed as randomly as possible, a larger fraction of the ribbons reoriented themselves with their long transverse faces (Fig. 1) perpendicular to the direction of compaction during the compaction and/or sintering stage. Scanning microscopy carried out on the fractured surfaces confirm this effect (Fig. 5). The higher strength obtained for this orientation can be primarily attributed to the higher moment of inertia of the ribbons in that orientation. Details of the orientation effects in such metallic-glass ribbon/glass–ceramic matrix systems have been discussed elsewhere [4, 5].

The results of the fracture toughness tests carried out are presented in Table III. The fracture toughness of the composite specimens was significantly isotropic

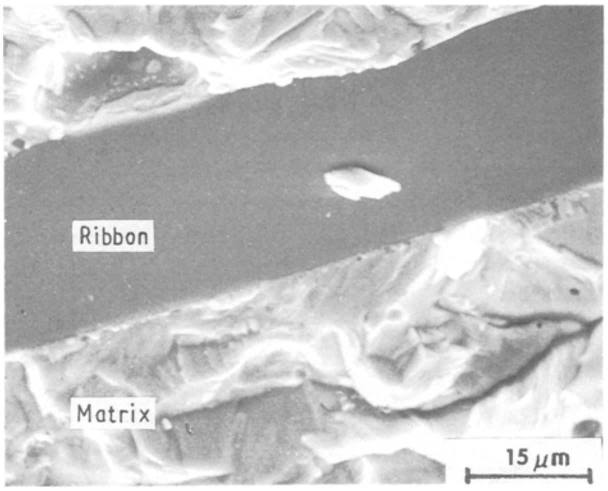

Figure 4 Scanning micrograph illustrating the strong bond between the matrix and the ribbon.

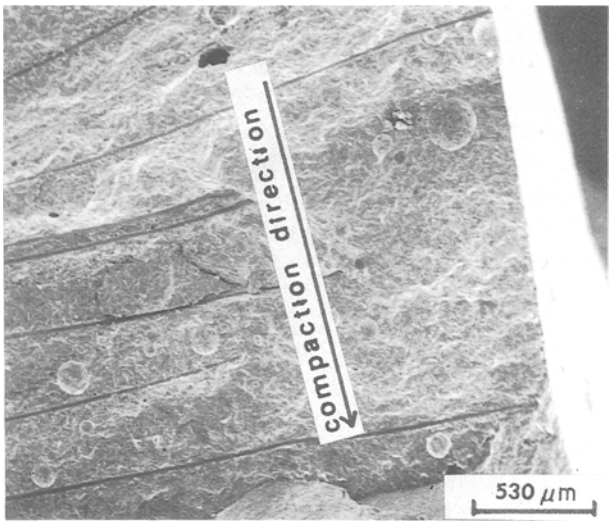

Figure 5 Micrographs of the discontinuously reinforced composites illustrating a large fraction of the reinforcements oriented with their long transverse faces perpendicular to the direction of compaction.

TABLE III Experimentally measured composite fracture toughness in two different directions. These directions are identified in Fig. 2. Volume fraction of ribbons 5.8%, dimensions of the ribbons length 0.5 cm, width 0.5 cm, thickness 25 μm

Direction	Average composite fracture toughness K_{IC} (MPa m$^{1/2}$)	Variance (%)
L	2.67	8.9
T	2.16	11.5

(though slightly lower in the transverse direction), and could possibly be attributed to the non-uniform ribbon distribution.

In order to characterize such discontinuous metallic-glass ribbon reinforced composites, it is necessary to examine the load transfer mechanism in such systems. Previous studies [8] carried out on such composites have indicated that both the ribbon length and width affect the load transfer in such composites. The two important equations developed as a result of that analysis are

$$\sigma_c^* = \left(\frac{2\tau V_f(w + t)l_c}{wt}\right)\exp[-(l_c - x)] + (\sigma_m^* V_m) \quad (1)$$

and

$$\sigma_{cm}^* = \sigma_f^* V_f = \left(\frac{2\tau V_f(w + t)l_c}{wt}\right) \quad (2)$$

where σ_c^* is the composite fracture strength, σ_{cm}^* the maximum composite strength, σ_f^* the ribbon fracture strength, σ_m^* the matrix fracture strength, V_f the volume fraction of the ribbons, V_m the volume fraction of the matrix, x the ribbon length, l_c the critical ribbon length (for this geometry $l_c = 2/w$), w the ribbon width, and t the ribbon thickness.

These equations were developed assuming perfect bonding between the matrix and the reinforcements, and assuming load transfer from the matrix to the ribbons occurred by shear. The exponential term in Equation 1 was obtained by experimental data fitting and satisfying the boundary conditions; for $x = 0$ (no ribbon), $\sigma_c^* = \sigma_m^* V_m$, and for $x = l_{c'}$, $\sigma_c^* = \sigma_{cm}^*$.

According to Equation 1, increasing the ribbon length (till l_c) causes an increase in the strength of the composite. The maximum composite strength is obtained for a ribbon length of l_c (Equation 2). The critical ribbon length depends on the width of the ribbon. Increasing the ribbon width decreases the critical ribbon length required for maximum load transfer.

A comparison between the theoretically predicted strength and experimental results can be seen in Fig. 6. A, B, C, D, E and F represent curves for different volume fractions and critical lengths of reinforcements. These curves were plotted by using Equation 1. From this equation it can be seen that the maximum load transfer for a given ribbon width is obtained for the same critical ribbon length. The maximum load carried, however, depends on the volume fraction of the reinforcements (and increases with increasing volume fraction of reinforcements). A, C, D and E represent curves plotted by using the critical length corresponding to a ribbon width of 0.5 cm, and different volume fractions of ribbon reinforcements. B and F represent curves corresponding to a ribbon width of 0.25 cm (but with different volume fractions of ribbon reinforcements). It can be seen that the ribbon width affects the critical ribbon length (compare curves A with B and curves E with F) but not the maximum load carried. The experimental results obtained are in agreement with the theoretically proposed model [8].

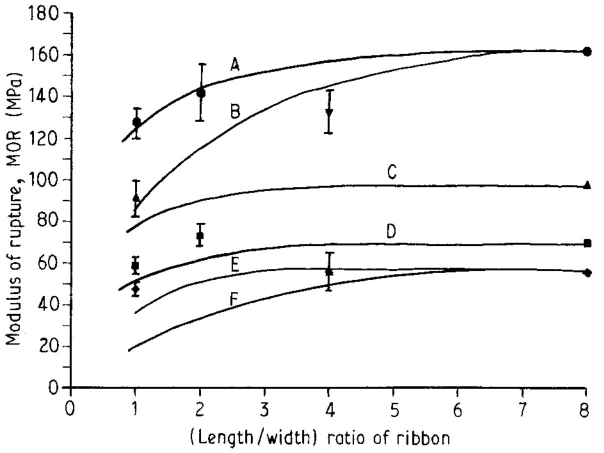

Figure 6 Plot of the composite strength as a function of the ratio of ribbon length to width. Solid curves A, B, C, D, E and F are obtained by using Equations 1 and 2. (V_f is the volume percent of the ribbon reinforcements and w is the ribbon width.) (A, $V_f = 5.8\%$, $w = 0.5$ cm; B, $V_f = 5.8\%$, $w = 0.25$ cm; C, $V_f = 3.5\%$, $w = 0.5$ cm; D, $V_f = 2.5\%$, $w = 0.5$ cm; E, $V_f = 2.0\%$, $w = 0.5$ cm; F, $V_f = 2.0\%$, $w = 0.25$ cm) The experimentally measured strength values are indicated by (●, $V_f = 5.8\%$, $w = 0.5$ cm; ▼, $V_f = 5.8\%$, $w = 0.25$ cm; ▲, $V_f = 3.5\%$, $w = 0.5$ cm; ■, $V_f = 2.5\%$, $w = 0.5$ cm; ◆, $V_f = 2.0\%$, $w = 0.5$ cm; ✱, $V_f = 2.0\%$, $w = 0.25$ cm)

The studies have indicated the feasibility of developing discontinuous metallic-glass ribbon reinforced glass–ceramic matrix composites with structural capabilities. Although some information on the load transfer in such composites has been obtained, further studies are necessary to understand the mechanism completely. It is also necessary to test scaled up components in order to investigate the 'size effect' before such composites can be incorporated into structural applications.

4. Conclusions

From the studies carried out so far it is evident that discontinuous metallic-glass ribbon reinforcements provide an unique method of obtaining isotropic properties in brittle matrix composites. These characteristics are as a result of a combination of factors which are (1) the high strength of the ribbons, (2) the high interfacial bond strength, and (3) the unique ribbon geometry. The stress transfer in such composites is complex, and depends on both the ribbon length and width. Specifying both these quantities is essential, as they both affect the maximum load carrying capabilities of the system.

Acknowledgements

The authors would like to thank the Composite Materials and Structures Center at Michigan State University for supporting and funding this project.

References

1. R. U. VAIDYA and K. N. SUBRAMANIAN, *J. Mater. Sci.* **25** (1990) 3291.
2. *Idem, ibid.* **26** (1991) 1391.
3. *Idem*, ASM/ACCE/ESD Conference Proceedings, Detroit, October 89 (American Society for Metals, Metals Park, OH, 1989) p. 227.
4. *Idem, J. Amer. Ceram. Soc.* **78** (1990) 2962.
5. R. U. VAIDYA, T. K. LEE and K. N. SUBRAMANIAN, Fifth Annual American Society for Composites (ASC) Conference Proceedings, East Lansing, June 1990 (Technomic, Lancaster, PA, 1990) p. 902.
6. J. R. STRIFE and K. M. PREWO, *J. Mater. Sci.* **17** (1982) 359.
7. *Idem*, AFWAL Report TR-80-4060 (1980).
8. R. U. VAIDYA and K. N. SUBRAMANIAN, *Comput. Sci. Technol.* in press.

Received 14 September 1990
and accepted 7 March 1991