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Rheological behaviour of borosilicate composites with metallic and non-metallic dispersions

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Abstract

Silicate matrix composites are potential candidates for high-temperature applications. In the present investigation, the effect of metallic (Cu) and non-metallic (SiC particulates, platelets, short fibres and whiskers) additions on the rheological behaviour of borosilicate matrix composites has been evaluated. The hot-pressed composites were tested both in compression and tension in the temperature range of 625–725°C. SiC reinforced composites tested in compression exhibited varying degree of strengthening and strain rate sensitivity depending on the volume fraction and morphology of reinforcements. The degree of strengthening and strain rate sensitivity depends on the volume fraction and morphology of reinforcements. Strengthening effect increased with the volume fraction and aspect ratio of reinforcements. The flow behaviour of composites changed from Newtonian to non-Newtonian with strain rate sensitivity index value changing from unity to 0.48. A similar trend was seen in the rate sensitivity of copper composites. However, copper additions decreased the strength of the composites at lower temperatures because of the softer copper phase. Pre-oxidation of copper particles had certain strengthening effect on the composite. The apparent viscosity of SiC reinforced composites increased with volume fraction and aspect ratio of reinforcements. However, in particulate composites, the viscosity found to increase with particle size. The mechanical/hydrodynamic interactions among the particulates appeared to be responsible for such a behaviour. With increasing strain rate, the viscosity decreased progressively confirming the shear thinning of the composites. The tensile ductility of the composites with 40 vol% reinforcements was evaluated at 700°C. While 400% elongation was observed in SiC particulate, platelet and copper composites, in short fibre/whisker composites, the tensile elongation values were only 150%. Further, the elongation of SiC platelet and copper composites improved by decreasing temperature and volume fraction of reinforcements, and also elongation values >500% were recorded. The tensile ductility of borosilicate composites was limited by onset and growth of cavities nucleated at the reinforcement/matrix interfaces.

Keywords: Borosilicates; Composites; Cu; Inclusions; Mechanical properties; SiC

1. Introduction

In the recent past, a large number of glass and glass-ceramic matrix composites have been developed by incorporating either continuous or discontinuous reinforcements.[1–3] The main advantage of using silicate matrices is that they can be easily infiltrated through a preform in a glassy state, and in some cases, infiltrated glass can be further crystallised to achieve better mechanical properties and refractoriness. The other important advantages of silicate matrices are: their availability in wide range of compositions, thermal expansion control to match with different types of reinforcements and control over crystallisation and formability. Many refractory glass compositions based on borosilicate, lithium–, calcium–, magnesium–, and barium–aluminosilicate systems were developed as potential matrices because of the above cited advantages.[1,4] Similarly, many oxynitride and oxycarbonitride compositions are also of great interest.[5,6] Using both oxide and non-oxide matrices, a number of composite materials have been developed with metallic and non-metallic dispersions.[7–10] Many important aspects related to processing, mechanical properties and interface microstructure were studied extensively.[1,2,10–12] Although various aspects of these composites were studied in greater detail, the rheological behaviour of discontinuous particulate reinforced silicate systems remains unexplored.

Earlier investigations had studied the flow behaviour of glass and glass-ceramics without any extraneous dispersions. At very high temperatures (i.e. in the vicinity

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of glass transition temperature, T_g) glass generally exhibits a Newtonian behaviour. The Newtonian behaviour can change to non-Newtonian if the temperature is decreased. For instance, Simmons et al.[13] investigated the flow behaviour of soda-lime-silica glass and reported a transition from Newtonian to non-Newtonian at a temperature of ~710°C. At high temperatures, a viscosity shear thinning effect was also noticed due to weakening of the glass structure. Gadkaree and Chyung[3] observed an increase in processing viscosity when 30 vol% whiskers were dispersed in an aluminosilicate glass matrix (Corning 1723). The viscosity increased by two orders of magnitude when compared to aluminosilicate base matrix. Demana and Drummond[14] measured the viscosity of alumina dispersed potassia borosilicate glass (Corning 7761) composites using a beam-bending viscometer. The viscosity of the borosilicate glass started increasing with dispersion of 10 vol% alumina and it increased by several orders of magnitude (8000 times) when alumina dispersions increased to 50 vol%. While studying SiC–borosilicate system, Tewari et al.[15] arrived at a similar conclusion. They also observed a transition from Newtonian to non-Newtonian when volume fraction of particulates increased from 20 to 40 vol%. Boccaccini[16] examined the applicability of some of the existing equations to predict the viscosity of these composites. At higher volume fractions, viscosity prediction became difficult presumably due to particle–particle interactions. Rouxel and Verdier[17] observed similar trends in oxynitride glasses. In these glasses, a shear thickening effect was reported.

When compared to glass systems, the work that was carried out on glass-ceramic systems is very meagre. Wang and Raj[18] studied a fine grained lithium aluminosilicate and reported the glass-ceramic (with β spodumene as crystallising phase) to be superplastic ($m=1$ at 10^{-3} s^{-1}) with tensile elongation more than 400%. In a model proposed for explaining the superplastic behaviour, diffusion through the residual glass was found to be responsible for such high elongations.

No systematic study is available for understanding the effect of reinforcements (both metallic and non-metallic) on the flow behaviour of silicate matrix composites. In the present investigation, the effect of reinforcement morphology, volume fraction and particle size on the flow behaviour of a silicate has been studied considering borosilicate as a model system.

2. Experimental

The composites of this study consist of borosilicate matrix and discontinuous reinforcements of SiC along with copper. Copper particles were added in as received and oxidised (200°C for 10 min) condition. The size of copper particles varied from 0.6–10 μm with 23% between 6.4–8 μm. The details of various SiC reinforcements are shown in Table 1. SiC reinforcements were of various types, viz. particulate, platelet, short fibre and whiskers. SiC reinforcements in different proportions (10–40 vol%) were weighed and dry ball milled with borosilicate glass powder for 24 h. For studying the particulate size effect four different sizes (44–63, 160–210, 297–351 and 500–595 μm) of SiC particulates were used. For comparative studies with other reinforcements only 44–63 μm size particulates were used. However, whisker admixture was prepared through wet dispersion technique to ensure uniform mixing, i.e. without agglomeration of whiskers in the matrix. For studying the effect of cristobalite, borosilicate was fully crystallised by sintering at 800°C for 24 h and then crushed cristobalite was added in a controlled quantity to the glass powder. Cylindrical samples of 10 mm

Table 1
Details of borosilicate glass and various reinforcements used in this study

	Matrix	Reinforcements				
Material	Borosilcate glass	SiC				Cu
Shape	Powder	Particulate	Platelet	Short fibre	Whisker	Powder
Size range (μm)	< 53	44–63 160–210 297–351 500–595	$l=25$–30 $t=1$–2	$l=50$–100 $d=8$–10	$l=20$–30 $d=2$	6.4–10 (67%)
Composition (wt%)	SiO_2 (80%) B_2O_3 (14%) Na_2O (4%) Al_2O_3 (2%)	α + β	Mainly β (3C) with small amount of α	Mainly β (3C)	Mainly β (3C)	99.99% Pure
Density (kg/m^3)	2.2×10^3	3.2×10^3	3.2×10^3	3.2×10^3	3.2×10^3	8.9×10^3
Thermal exp. coeff. (°C^{-1})	3.3×10^6	4.3×10^6	4.3×10^6	4.3×10^6	4.3×10^6	16.7×10^6
Young's modulus (GPa)	60.0	–	440	200	440	125

diameter and 12 mm height were prepared for all compression tests. Hot pressing (DSP-6 Dr.Fritsch KG, Germany) technique was adopted to prepare these samples, where BN coated graphite die assembly was used typically at 700°C and a pressure of 10 MPa. Tensile samples of 10 mm gauge length and cross sectional area of 6 mm^2 were prepared by another hot press (Electrofuel, Canada) using the same processing parameters. The initial porosity level was very low (< 1%) for the platelet samples prepared for compression tests, whereas for tensile samples it was higher (4%). However, whisker composites are most prone high porosity levels (7–10%). Prior to high temperature tests, both compression and tensile test samples were polished to 1 μm finish. DTA (Shimadzu DTA-50) was performed on composite powder to monitor any change in the glass transition temperature (T_g) with the addition of reinforcements in the glass. X-ray diffraction (Siefert Iso Debyeflex-1002) was employed to reveal the presence of any crystallisation. SEM (JEOL JSM 840A) and optical (Leitz) metallographic observations were made on samples before and after mechanical tests. The compression and tensile tests were carried out using an MTS machine (model 810.12). Two different temperature ranges were selected for compression and tension tests; 625–700 and 675–725°C, respectively. The compression tests were carried out in the strain rate range of 10^{-5}–10^{-2} s^{-1}, while tensile tests were performed using a constant cross head velocity (starting strain rate ~10^{-3} s^{-1}). The porosity in the tensile samples before and after tensile tests were measured by water displacement technique based on Archimedes principle. After tensile tests, all the samples were cut at every one centimetre interval and the porosity was measured for each piece to get an idea about the gradient in porosity levels across the sample.

3. Results and discussion

3.1. Microstructure

The microstructures of various borosilicate composites are shown in Fig. 1. As seen from the micrographs, the reinforcement distribution is quite homogeneous in the composites. Two aspects of microstructure are important for the high temperature flow behaviour of composites. Firstly, the stability of the matrix is an important factor. During hot-pressing, if the borosilicate matrix crystallises, the formation of cristobalite influences the flow behaviour of glass. Hence X-ray diffraction analysis was employed to detect cristobalite. However, X-ray profiles did not indicate any devitrification. But when the matrix was etched with HF, very fine cristobalite crystals were seen. The total volume fraction of cristobalite is estimated to be around 5%. To understand the effect of cristobalite on the flow behaviour initial experiments were carried out by externally adding controlled quantities of cristobalite. Secondly, addition of extraneous reinforcement can affect the glass transition temperature (T_g). No change was observed in T_g when SiC added to the matrix whereas with the addition of 40 vol.% copper, T_g shifted from 750°C to 792°C. An increase in T_g tends to suppress the cristobalite formation to some extent. Further, at 700°C, no interfacial reaction was observed in SiC composites. In Cu (oxidised)-borosilicate composites a 2 μm interdiffusive layer was noticed.

3.2. Flow behaviour of composites in compression

For each composite, a load-displacement plot was obtained from cross head velocity change test in compression. Flow stress (σ) and strain rate ($\dot{\varepsilon}$) were calculated form the load-displacement plot and the data were analysed in terms of well know power law for strain rate dependent flow:

$$\sigma = K\dot{\varepsilon}^m \tag{1}$$

where K is proportionality constant, a function of temperature and microstructure and m is the strain rate sensitivity index. Flow stress–strain rate data of different composites are presented in double logarithmic plots to assess the extent and nature of strengthening with respect to filler volume fraction, morphology, size and test temperature. From this data m values have been tabulated. While considering the composite flow behaviour in terms of flow stress and strain rate, the viscosity of the composites at elevated temperatures is assessed keeping the viscous nature of the glass matrix in mind. The following relation is used for this purpose.

$$\eta = \frac{\sigma}{\dot{\varepsilon}} = \frac{K}{\dot{\varepsilon}^{(1-m)}} \tag{2}$$

The activation energy (Q) is calculated using the data available for different temperatures at a constant strain rate of 1.0×10^{-4} s^{-1}. The constitutive relationship used for this case is:

$$\dot{\varepsilon} = A\sigma^n e^{\left(-\frac{Q}{RT}\right)} \tag{3}$$

where n ($n = \frac{1}{m}$) is the stress exponent, A is a constant and R is the gas constant. From Eq. (3) the activation energy (Q) is given by

$$Q = \left(\frac{R}{m}\right)\left[\frac{\delta \ln \sigma}{\delta(1/T)}\right]_{\dot{\varepsilon}} \tag{4}$$

3.2.1. Effect of cristobalite

To start with, it is of interest to know whether the cristobalite formed during hot-pressing of glass powder

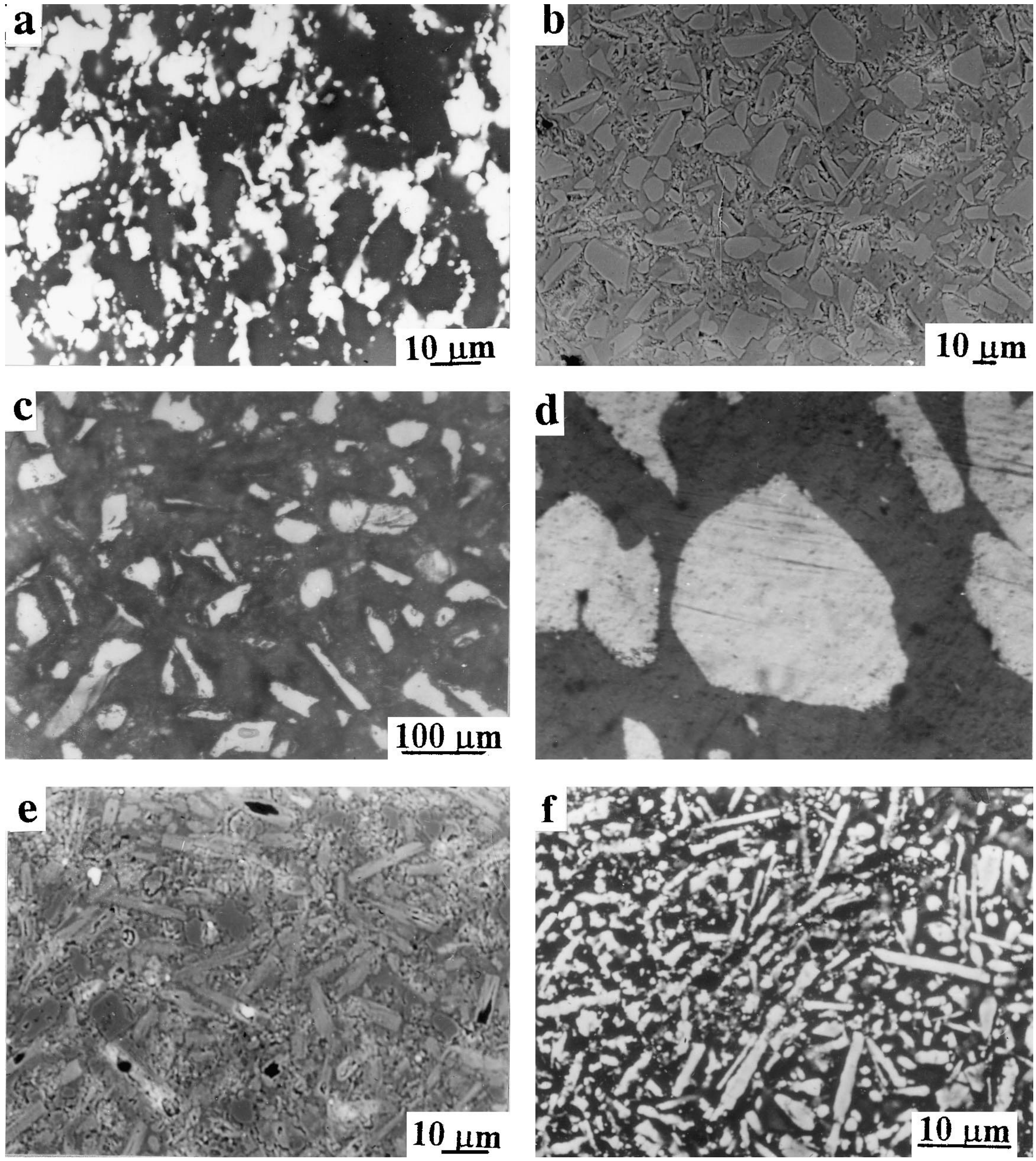

Fig. 1. Micrographs of 40 vol% composites with (a) copper, (b) platelet, (c) fine particulate, 44–63 μm, (d) coarse particulate, 500–595 μm, (e) short fibre and (f) whisker reinforcements.

modifies its flow behaviour. The data presented in Fig. 2 indicate that the flow behaviour is nearly the same with the addition of 5 and 10 vol% cristobalite at 650 and 700°C. However, there is a change in the strain rate sensitivity value which decreases from unity to ~0.7 for 5 and 10 vol% additions of cristobalite, respectively (Table 2). As the amount of cristobalite formed being less than 5 vol% in the composites, the strain rate sensitivity values would not have been affected significantly at higher temperature, i.e. 700°C.

3.2.2. Effect of copper additions

The strain rate data for different copper reinforced composites (processed with and without surface oxidation of copper particles) are presented in Fig. 3a and b. In composites with as received copper, the flow stress decreased in the temperature range of 625–675°C with the increase in copper content. On the other hand, a strengthening effect is seen with increasing addition of copper at 700°C. A similar trend is noted for composites processed with oxidised copper powder. The transition

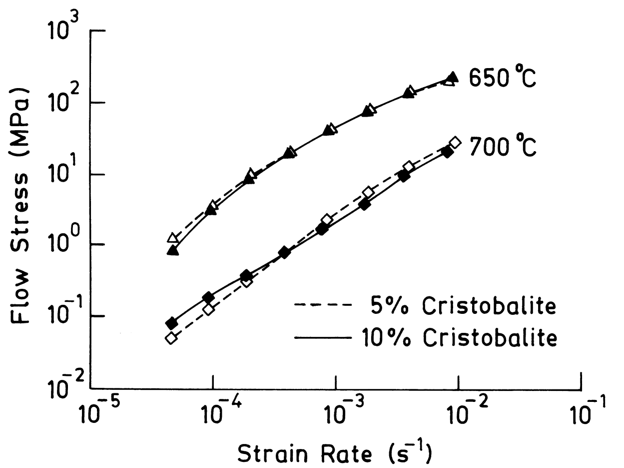

Fig. 2. Flow behaviour of borosilicate glass in the presence of varied amount of cristobalite.

Table 2
K and *m* values for cristobalite containing borosilicate glass composites

Temperature °C	Vol% of cristobalite	$\dot{\varepsilon}$ (s^{-1})	*K* (MPa s^m)	*m*
650	5	$>2\times10^{-1}$	1.7×10^3	0.67
	10	$>2\times10^{-4}$	2.2×10^3	0.72
700	5	Entire range	1.0×10^2	1.0
	10	Entire range	2.1×10^2	1.0

temperature from a reduction in strength to an increase in strength with addition of copper is, however, lowered to 675°C. The strength of copper relative to glass changes with increasing temperature. The flow behaviour shown in Fig. 3a and b suggests that while copper is soft phase below 675°C, it is the hard phase at higher temperatures in these composites.

In addition to the effects of temperature and copper content, the differences in the flow behaviour of the composites processed with and without oxidised copper is of interest to assess the role of copper oxide. Composites processed with oxidised copper powder exhibit higher flow stress than their counterparts. The difference in their flow stress increases with volume fraction of copper and drop in test temperature. Considering the strain rate regime below 10^{-3} s^{-1}, their stress–strain rate data were analysed and the parameters of the power law relation are listed in Table 3. The strain rate sensitivity is seen to decrease with temperature and also with the increase in the copper content. From the data it is evident that the transition from Newtonian to non-Newtonian character of flow is prominent in composites with oxidised copper particles. The effect of volume fraction on viscosity is, however, not significant. The viscosity of composites with oxidised Cu powder is also higher than that of as-received Cu. This increase in viscosity of composites with oxidised copper and their

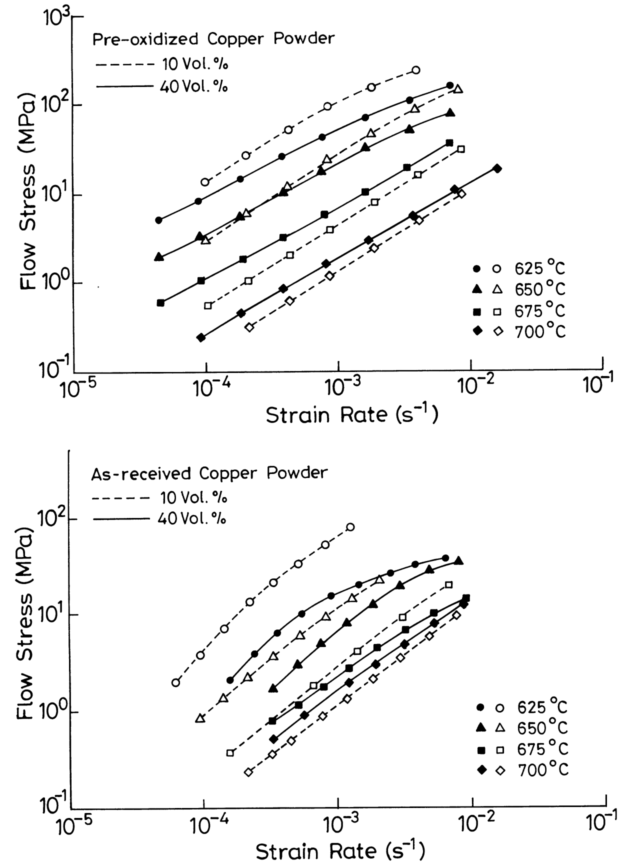

Fig. 3. Flow behaviour of copper–borosilicate composites with and without oxidation of copper particles.

high flow stress arise from a Cu_2O film on copper particles.[19]

3.2.3. *Effect of SiC additions*

The stress(σ)–strain rate($\dot{\varepsilon}$) data showing the effects of volume fraction of reinforcements, morphology of reinforcements and temperature on flow behaviour of composites are presented in Fig. 4. Borosilicate glass exhibits Newtonian viscous flow behaviour at elevated temperatures. The flow behaviour with four types of SiC–borosilicate composites can be broadly divided into two groups based on their mechanical response, viz. (a) particulate/platelet and (b) short fibre/whisker composites. The flow stress in these composites increases with the addition of SiC reinforcements and such an effect is more pronounced with increasing volume fraction and aspect ratio of reinforcements (Fig. 4a and b). Similar increase in flow stress was observed with the drop in test temperature (Fig. 4c and d). The flow behaviour of various composites relative to the borosilicate glass can be rationalised in terms of the strengthening effect of reinforcements with different aspect ratios. The borosilicate glass exhibits rate sensitive Newtonian viscous flow that is strongly temperature dependent whereas the reinforcement flow behaviour is rate insensitive and

Table 3
Various flow parameters of Cu–borosilicate composites with and without oxidation of copper

		K (MPa s^m)		m		η (MPa s) at $\dot{\varepsilon}=10^{-4}$ s^{-1}	
Composite	temperature (°C)	10 vol% Cu	40 vol% Cu	10 vol% Cu	40 vol% Cu	10 vol% Cu	40 vol% Cu
With Cu powder	625	26.3×10^3	0.4×10^3	1.0	0.63	79×10^3	35×10^3
	650	3.7×10^3	0.5×10^3	1.0	0.76	11×10^3	14×10^3
	675	1.1×10^3	0.2×10^3	1.0	0.79	4.1×10^3	4.1×10^3
	700	0.4×10^3	0.4×10^3	1.0	0.97	1.3×10^3	1.7×10^3
With oxidised Cu powder	625	2.7×10^3	0.9×10^3	0.65	0.59	210×10^3	130×10^3
	650	2.1×10^3	0.7×10^3	0.78	0.66	48×10^3	48×10^3
	675	0.7×10^3	0.7×10^3	0.88	0.83	6.2×10^3	10×10^3
	700	0.3×10^3	0.2×10^3	0.92	0.82	1.6×10^3	3.1×10^3

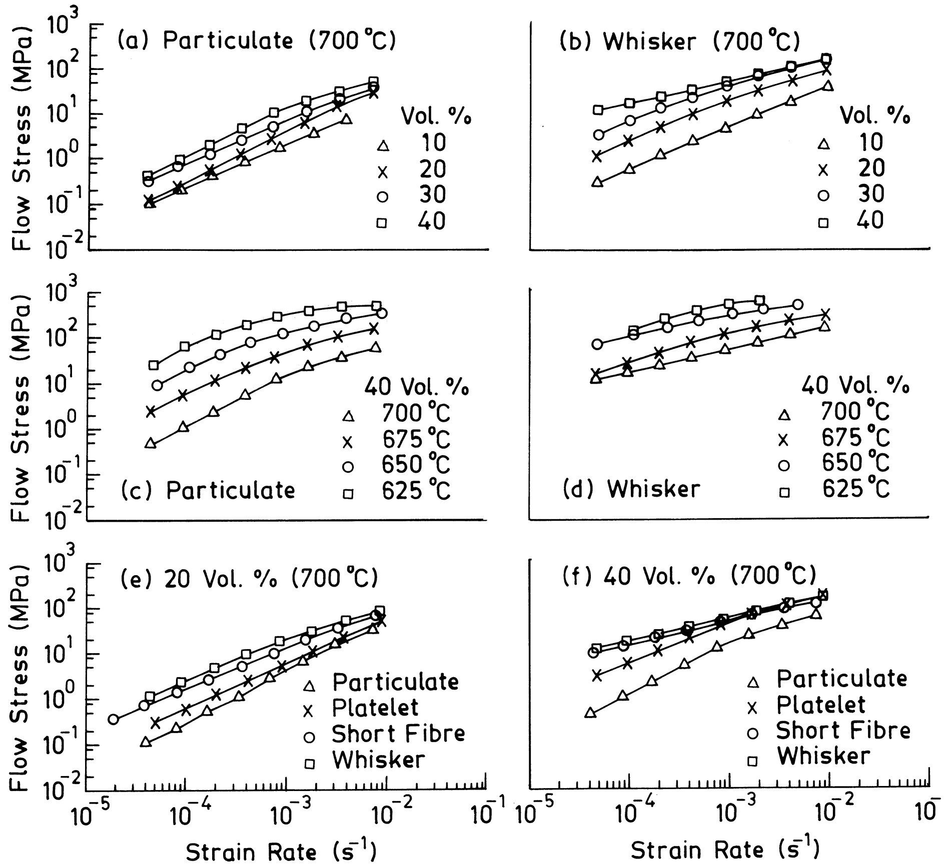

Fig. 4. Flow behaviour of SiC–borosilicate glass composites showing the effect of (a) and (b) volume fraction, (c) and (d) temperature and (e) and (f) morphology.

temperature independent. Accordingly, the flow behaviour of composites with low volume fraction of particulate and platelets is dominated by the matrix. And also the load sharing of particulates being less compared to the whiskers, the flow behaviour of composites with lower fraction of particulates is dominated by the matrix whereas in whisker composites, it is dominated by the reinforcing phase. On the other hand, the reinforcements contribute increasingly to the flow behaviour of composites at higher volume fraction and aspect ratio (Fig. 4e and f). The rate sensitivity and temperature dependence of flow stress of composites would the decrease as the volume fraction and aspect ratio of reinforcements increase. The transition from Newtonian to non-Newtonian flow in various composites is thus a consequence of the extent of strengthening by the reinforcements and all the observed flow characteristics can be interpreted accordingly.[20,21]

The m values characteristic of strain rate regime 10^{-3} s^{-1} were evaluated and compared in Table 4. The strain rate sensitivity is seen to decrease with increasing aspect ratio of reinforcements. At 700°C, while 40 vol% particulate composites exhibited nearly Newtonian behaviour whereas a non-Newtonian behaviour with $m = 0.48$ was evident in whisker composites. Similarly, temperature also had a significant effect. As the temperature decreased, m value progressively decreased (Table 4). With a loss in rate sensitivity in the higher strain rate regime, the flow behaviour became non-Newtonian and non-linearity dominated in the double logarithmic plots. The apparent viscosity of composites increased with volume fraction and aspect ratio of reinforcements (Fig. 5a and b). Further, shear thinning or a decrease in viscosity at a higher strain rate regime was also noticed (Fig. 6). The viscosity of whisker and short fibre composites is significantly higher than that of particulate and platelet composites. A rapid rise in viscosity may be noted at the lowest temperature and highest volume fraction of reinforcements. The effects of volume fraction (V_f) of reinforcements and temperature on viscosity (η) have been analysed in terms of the following empirical relation:

$$\eta = a \exp\left(bV_f + \frac{c}{T}\right) \quad (5)$$

where a, b and c are constants and T the absolute temperature. The values of a, b, c obtained by regression analysis are listed in Table 5.

Table 4
The strain rate sensitivity index (m) of different composites in the lower strain rate ($< 10^{-3}$ s^{-1}) regime

Volume fraction	Composites			
	Particulate	Platelet	Short fibre	Whisker
700°C				
0.1	0.90	1.00	0.90	0.90
0.2	0.97	1.00	0.90	0.90
0.3	0.95	0.93	0.71	0.84
0.4	1.00	0.85	0.49	0.48
675°C				
0.1	1.00	0.94	0.96	0.93
0.2	1.00	1.00	0.77	0.90
0.3	1.00	0.87	0.80	0.66
0.4	0.95	0.82	0.61	0.58
650°C				
0.1	1.00	0.89	0.84	0.82
0.2	1.00	0.86	0.85	0.84
0.3	1.00	0.80	0.75	0.70
0.4	0.90	0.70	0.70	0.50
625°C				
0.1	0.80	0.80	0.80	0.70
0.2	0.80	0.70	0.70	0.70
0.3	1.00	0.70	1.00	0.60
0.4	0.80	0.60	0.50	0.50

Making use of the flow stress data at various temperatures for a strain rate of 1.0×10^{-4} s^{-1}, the activation energy is calculated and listed for various composites in Table 6. Activation energy generally decreases with increasing volume fraction and aspect ratio of reinforcements. The differences in the activation energy for flow in various composites can be analysed in terms of the temperature dependence of their flow stress. The temperature dependence of flow stress is expected to be different depending on the dominance of matrix or reinforcement in the flow behaviour of composites. In composites with low concentration of reinforcements, the flow behaviour corresponds to that of the matrix; thereby stronger temperature dependence of flow stress and activation energy would follow. On the other hand, low activation energy in the composites with a high concentration of reinforcements is indicative of milder temperature dependence of the flow stress and it follows from an increasing degree of load shared by the reinforcing phase.

3.2.4. SiC particulate size effect

In SiC particulate composites, in addition to strengthening, addition of reinforcements led to non-Newtonian behaviour especially at higher volume fractions. The flow became increasingly non-Newtonian with decreasing temperature, increase in volume fraction and increase in strain rate. Flow behaviour of the composites for reinforcements of four different size ranges and volume fraction is found to be strongly dependent on temperature. For a given temperature and volume fraction the extent of strengthening depends on reinforcement size (Fig. 7a and b). The flow stress is higher for larger particles. The strain rate sensitivity index values calculated are presented in Fig. 8 for two extreme cases of reinforcement size. The modification of microstructural features as well as load bearing capacity of the material with deformation in progress are responsible for the change in rate sensitivity. In particulate reinforced glass composites, a gradual loss in rate sensitivity is also probably due to greater frequency of collisions between the particles at higher volume fractions. For all the composites the apparent viscosity increased with volume fraction of reinforcements and drop in temperature. The size dependency of viscosity is shown in Fig. 5c and d. An increase in viscosity of nearly five fold is seen for two extremes of particle size at 40 vol% loading. The observed particle size effects may be rationalised by considering the colloid-chemical and hydro-dynamic/mechanical forces acting on the particles in suspension. For small particle size (< 5 μm), colloid-chemical interactions contribute significantly towards an increase in the viscosity. However, in coarser particle size ranges of this study and their inertness to the matrix exclude the possibility of any colloidal-chemical activity. The Vand[22] model suggests that particles

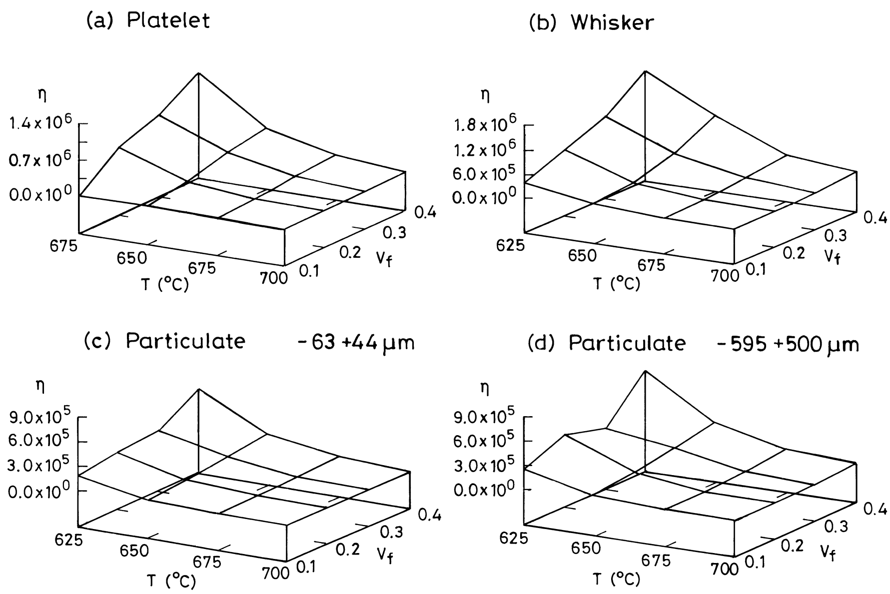

Fig. 5. The variation of viscosity with volume fraction and temperature for (a) platelet, (b) whisker and particulate (c) 44–63 μm and (d) 500–595 μm composites.

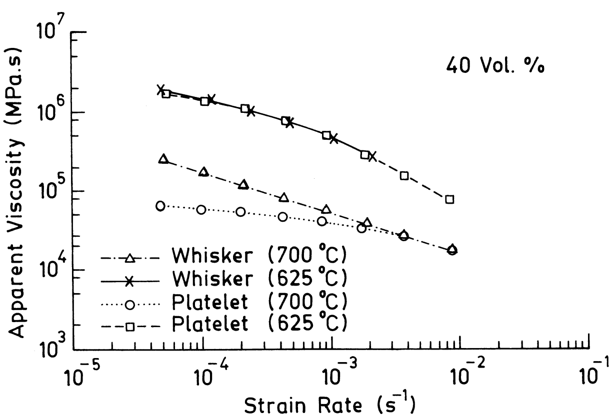

Fig. 6. Shear thinning behaviour of 40 vol% platelet and whisker composites.

Table 5
Values of constants in Eq. 5

Composites	*a*	*b*	*c*
Particulate	1.5×10^{-18}	5.8	4.7×10^{4}
Platelet	3.3×10^{-18}	7.4	4.7×10^{4}
Short fibre	3.5×10^{-15}	7.5	4.1×10^{4}
Whisker	4.7×10^{-14}	7.9	3.8×10^{4}

Table 6
Activation energy values (kJ mol^{-1}) for flow calculated at a constant strain rate of 1.0×10^{-4} s^{-1}

Vol%	Particulate	Platelet	Short fibre	Whisker
10	544	468	500	457
20	403	480	436	408
30	335	350	360	378
40	345	391	387	426

of adjacent layers sliding past each other form a doublet or triplet and rotate in the suspension as single entity for a finite period of time. In the process due to immobilised liquid entrapped in between and around the particles, an effective increase in volume fraction will occur and thereby an increase in viscosity follows. Similarly, collisions between the particles dissipate energy which can also intern give rise to an increase in viscosity.[23,24] Further, the physical phenomena responsible for the rise in viscosity with larger particle and higher degree of particle loading also intern affect the strengthening behaviour and rate sensitivity of the composites. The effect these parameters on viscosity and other related parameters is discussed at a greater length else where.[25] Further, the combined effect of temperature (T), volume fraction (V_f) and average particle size (s) on the apparent viscosity (η) has been analysed in terms of the following relation:

$$\eta = a\exp\left(bV_{\mathrm{f}} + \frac{c}{T} + ds\right) \qquad (6)$$

where a, b, c and d are constants. T is the absolute temperature. From the experimental data, the values of these four constants calculated by regression analysis are 4.6×10^{-19}, 5.8, 4.8×10^{4}, 1.1×10^{-3}, respectively.

The activation energy values were found to vary in the range of 335–540 kJ/mol depending on the particle size and volume fraction. The activation energy generally

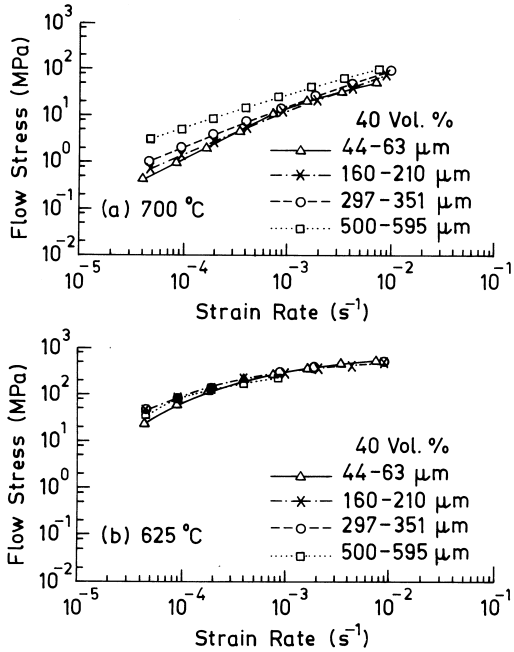

Fig. 7. Effect of different size ranges of reinforcements on strengthening of 40 vol% composites at different temperature (a) 700°C and (b) 625°C.

decreased with increasing particle size and volume fraction of particulate.

3.3. *Flow behaviour of composites in tension*

It may appear that good ductility can be expected both in tension and compression in rate sensitive materials. While plastic instability known as necking affects tensile ductility, such instability does not occur in compression. In materials exhibiting viscous flow, strain rate hardening stabilises uniform flow along the gauge length of a tensile specimen and flow localisation and neck growth are suppressed. Hence a material exhibiting viscous flow would neck down to a point and high tensile ductility can be expected. However, rate sensitivity of flow stress is only a necessary but not sufficient condition for large ductility. The onset of fracture events involving initiation and propagation of cracks/voids become significant in tensile mode of testing and the tensile ductility may be limited by fracture. Compressive stresses would not favour initiation and propagation of cracks/voids and thus compressive ductility may be better than that in tension. Glass matrix composites generally exhibit lower ductility in tension than in compression at temperatures around glass transition temperature where the flow becomes rate sensitive. The tensile ductility of these composites is of interest from the view point of their deformation processing. Accordingly, the tensile ductility of various composites is explored in this study at different temperatures. The results and the origin of the differences in the tensile ductility of different composites are considered below.

The tensile ductility of 40 vol% copper and SiC composites at different temperatures is illustrated in Fig. 9 and Table 7. Two general trends can be noted from these data of different composites. At a given temperature, the ductility of particulate/platelet and copper reinforced composites is significantly greater (400%) than in short fibre/whisker composites (150%). Further, the tensile ductility of copper and platelet composites is higher at lower temperatures.

The reinforcements in glass matrix composites not only reduce the rate sensitivity of flow stress but also promote early fracture in tension. Both of these factors influence the tensile ductility of composites. The observed variation in tensile ductility of composites with different reinforcements may have its origin in the aspect ratio of reinforcements. The aspect ratio of short fibres/whiskers is considerably higher than that of particulate/platelets. Although the short fibre/whisker reinforcements are randomly distributed in the matrix, some of the interfaces will be transversely aligned with respect to the tensile direction and such interfaces are potential sites for initiating voids/cracks. Similar events can occur to a lesser degree in particulate/platelet composites. The

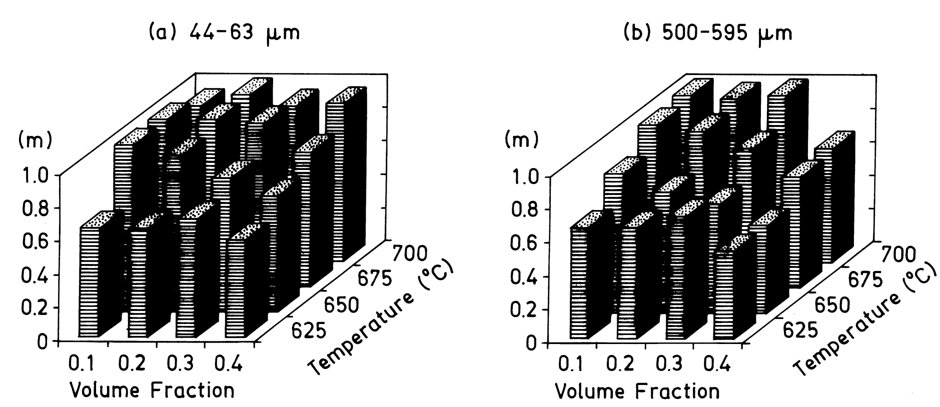

Fig. 8. Variation in strain rate sensitivity index (m) values with volume fraction and temperature for (a) 44–63 μm and (d) 500–595 μm particulate composites.

Table 7
Tensile elongations of different borosilicate composites

Composites	Volume fraction	% Elongation at		
		675°C	700°C	725°C
Particulate	0.4	–	400	–
Platelet	0.2	> 600	> 650[a]	> 650
	0.4	500	400	250
Whisker	0.2	200	400	250
	0.4	–	150	–
Short fibre	0.4	–	200	–

[a] When tested beyond the uniform hot-zone, an elongation of > 800% was recorded without failure.

local stress concentration at the matrix/reinforcement interfaces is also expected to be more severe in the case of reinforcements of higher aspect ratio. Moreover, the strength of composites with short fibres/whiskers is higher than the others and thereby the magnitude of tensile stress is higher than in case of reinforcements with smaller aspect ratios. Because of these reasons, fracture would set in earlier in reinforcements of high aspect ratio. Thus the overall effect of the reinforcement geometry is a lower tensile ductility in composites with reinforcements of high aspect ratio relative to those with reinforcements of low aspect ratio.

The other trend is the enhanced tensile ductility in copper and platelet reinforced composites at lower test temperature. From the data shown in Fig. 9b and c and Table 7, higher ductility at 675°C can be noted than at 700 and 725°C in both platelet and copper reinforced composites. The higher ductility seems to occur despite the increase in tensile flow stress at lower temperature. Thus it appears that the temperature dependent fracture events dominate the flow behaviour over the level of tensile stress. Diffusion controlled growth and linkage of voids may be the rate-controlling factor in the fracture of these composites. These processes are thermally activated and occur more readily at high-temperature and limit the tensile ductility. Further, the change in the relative hardness of the two phases of the composite with temperature may also play an important role in copper composites. Copper is softer than the matrix at 675°C, whereas it is the harder phase at 700°C. Initiation of voids at particle/matrix interfaces may occur to a greater extent in the case of harder particles (Fig. 10). Accordingly the observed trend in ductility variation with temperature may be expected.

Another noteworthy observation in the tensile flow behaviour of composites is the extensive swelling of deformed specimens as a result of formation of voids/pores. Bulk density measurements were carried out at different locations along the gauge length of tensile specimens to estimate their pore content and the data are presented in Fig. 11. A maximum of 60% pore content was observed and it varied depending on the nature and volume fraction of reinforcement in the composites. A

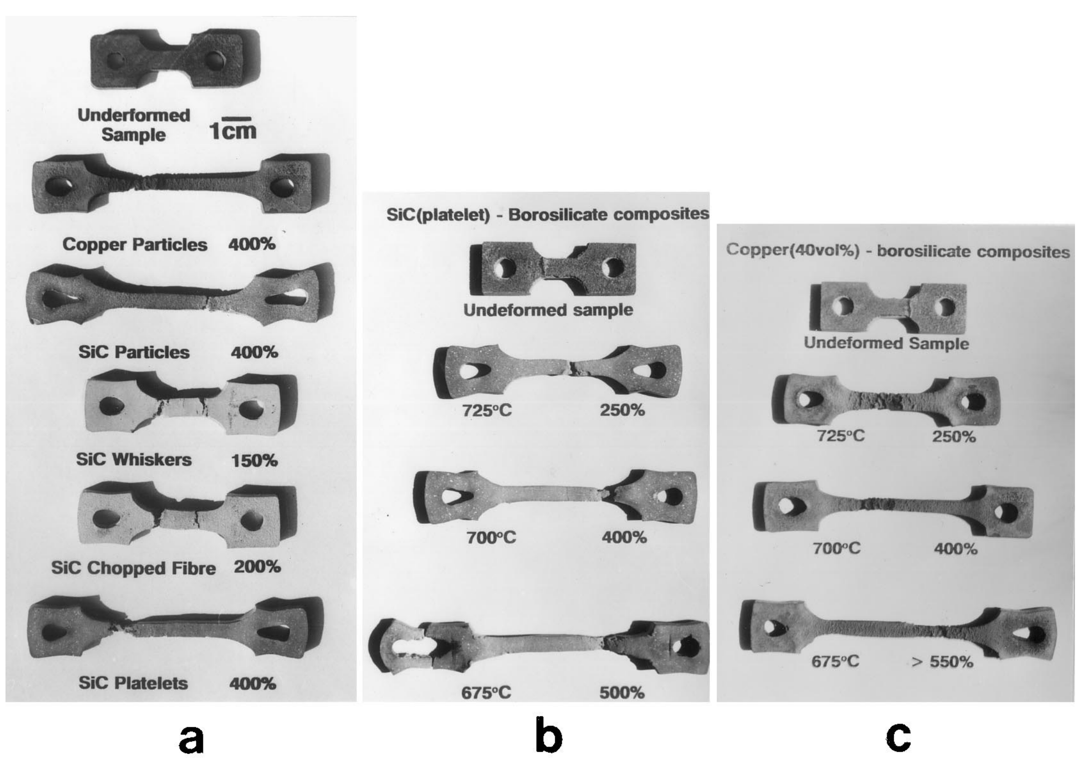

Fig. 9. (a) Tensile elongations of 40 vol% copper and SiC composites. (b) Tensile specimens of 40 vol% SiC composites at three different temperatures and (c) tensile specimens of 40 vol% copper composites at different temperatures.

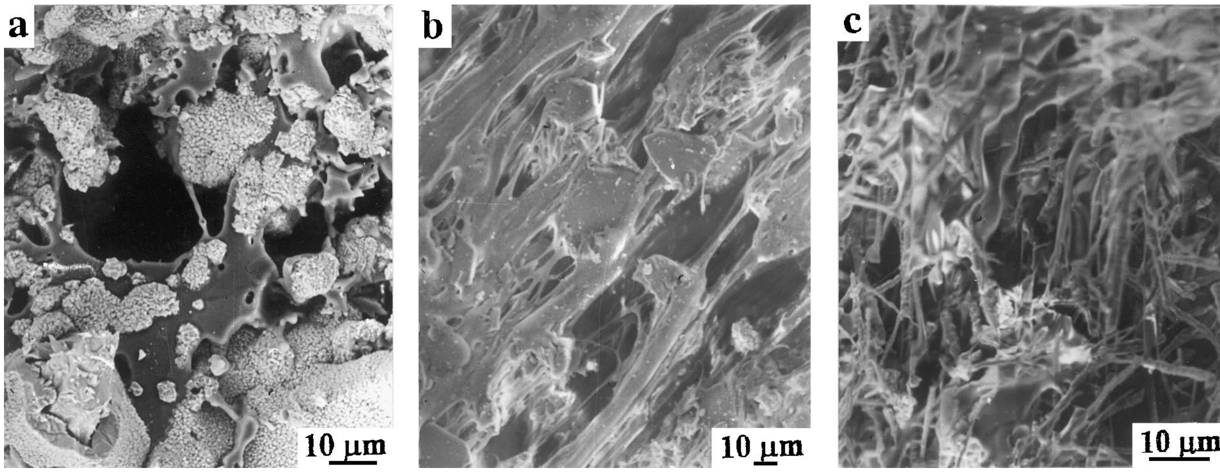

Fig. 10. Cavitation in (a) copper, (b) platelet and (c) whisker composites.

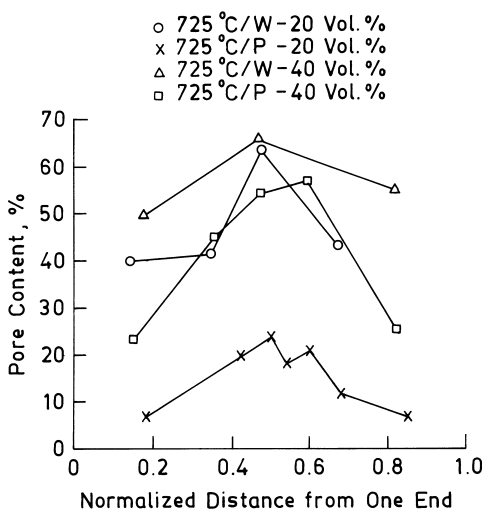

Fig. 11. Variation of porosity across the tensile specimens of different borosilicate composites.

porosity gradient from either end toward the centre of the gauge length can be noted. The triaxiality of stress state in the neck zone of the tensile specimen promotes pore formation to a greater degree in the final fracture zone than the ends of the gauge. Thus the porosity gradient is related to the extent of triaxiality of stress state and the non-uniformity of specimen cross-section along the gauge.

4. Conclusions

1. At higher temperature (700°C), the presence of cristobalite (up to 10 vol%) did not influence the flow behaviour of borosilicate glass. However, at lower temperatures the effect appeared to be significant. Although there was not much influence on flow stress values the strain rate sensitivity value decreased from unity to 0.7 at 650°C.
2. Addition of increasing amounts of copper resulted in a lower strength of the composites due to the softer copper particles relative to the matrix. However, at 700°C, strengthening was observed due to softening of glass matrix relative to metallic particles. Composites processed with oxidised copper exhibited a higher flow stress. The strain rate sensitivity decreased as the copper content increased.
3. All SiC–borosilicate composites exhibit strengthening in response to increasing strain rate and drop in temperature. While the flow stress increases with the volume fraction and aspect ratio of reinforcements, the rate sensitivity gradually decreases. In case of SiC reinforcements, the strain rate sensitivity index decreased from unity to 0.50, whereas in copper dispersed composites it decreased to 0.63. With a loss in rate sensitivity in the higher strain rate regime, the flow behaviour tends to become more non-Newtonian and non-linearity of double logarithmic stress-strain rate plots predominates.
4. The apparent viscosity of copper dispersed composites varied with temperature and is almost independent of copper. Composites with oxidised copper exhibited a slightly higher viscosity compared with as-received copper containing composites. In SiC dispersed composites, the apparent viscosity increases with volume fraction and aspect ratio of reinforcements. The greatest resistance to matrix flow is offered by the reinforcements having highest aspect ratio, which results in a significant increase in viscosity of short fibre/whisker composites. However, in particulate composites viscosity increased with higher volume fraction and larger

size of the particles. The mechanical/hydrodynamic interactions among the particles are probably responsible for the rise in the viscosity in the composites with larger particles. In SiC composites empirical relationships between apparent viscosity and various processing parameters have been developed by regression analysis. With increase in strain rate, the viscosity decreases progressively confirming to the shear thinning behaviour of the composites.

5. At 700°C, copper and SiC platelet/particulate composites exhibited higher tensile elongations (400%) than in whisker/short fibre composites. 20 vol% composites offered higher tensile ductility compared to its 40 vol% counterpart. In 40 vol% platelet composites tensile ductility increased with decrease in temperature. The tensile ductility of composites is limited by the onset and growth of cavities/cracks at the reinforcement/matrix interface as a result of stress concentrations of varying degree depending on the reinforcement morphology. Across the gauge length of all the tensile samples, a variation in the cavitation is observed. The cavitation generally increased from shoulder to the centre of the samples.

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