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SiC Particle Reinforced Oxynitride Glass: Reinforcement and Rheology

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Abstract

The present work is devoted to the study of the reinforcement and flow behavior of oxynitride glass matrix composites. The main objective is to combine both good reinforcement and suitability to viscoplastic forming techniques. SiC particulate composites were synthesized with various volume fractions (up to 47 vol%) and particle sizes (from 0·5 to 150 μm). A remarkable increase of the mechanical properties is observed with increasing SiC volume fraction and decreasing particle size. The viscoplastic forming ability of the composite was then evaluated by means of creep measurements. The forming ability is found to decrease with increasing volume fraction and decreasing particle size. The rheology was also studied with relaxation tests. In this case, the kinetic is significantly higher for the glass than for the composite, consistant with the creep results. © 1997 Elsevier Science Limited.

1 Introduction

Glasses and glass-ceramics offer a unique combination of structural properties. Their advantages over ceramics are their suitability for viscoplastic forming techniques, i.e. at relatively low temperature, and their extremely wide range of compositions which allows the continuous modification of their properties.

Particle incorporation into such matrices has the potential to improve the mechanical properties and maintain their ability to undergo viscoplastic forming.

This study is devoted to the comparison of the dependence of the mechanical and rheological properties of such composites with size and volume fraction of the reinforcement particles.

2 Elaboration

The chosen matrix is an oxynitride glass with stoichiometric composition $Y_{0\cdot124}Mg_{0\cdot160}Si_{0\cdot414}Al_{0\cdot302}O_{1\cdot4}N_{0\cdot151}$. The reinforcement particles are commercial SiC powders (Table 1).

The bulk glass is milled and then mixed with the desired SiC powder by ball-milling. Dense composites are obtained by hot-pressing at a temperature higher than the glass transition temperature, but avoiding crystallization of the matrix, a complete description of the processing route is given in a previous publication.[1] Composites with various sizes (from 0·5 μm to 150 μm) and volume fractions (up to 47vol %) of particles were prepared.

3 Characterization

Young's moduli were determined by means of an ultrasonic technique, with 10 MHz piezoelectric transducers.[1,2]

Hardness and fracture toughness were measured by means of Vickers indentations, the mode I fracture toughness KIc was calculated from the equation of Marshall and Evans.[3] A load of 10 kg was selected and the dwell time at peak load was 30 s. Each specimen was indented 10 times on mirror surfaces.

The fracture strength was measured from 3 point-bend experiments using 3 specimens ($3 \times 4 \times 25\,mm^3$ rectangular bars) for each grade, with a cross-head speed of $0\cdot5\,mm\,min^{-1}$.

The creep tests were conducted with a dead-load machine. The specimens used were rectangular bars ($3 \times 4 \times 25\,mm^3$) for 3 point-bend tests and rectangular prisms ($3 \times 3 \times 4\cdot5\,mm^3$) for compression tests. The apparent viscosity (η) is expressed

Table 1. Glass and silicon carbide physical properties

	$\rho(g/cm^3)$	$\alpha(10^{-6}\,°C^{-1})$	$H_v(GPa)$	$\sigma_r(MPa)$	$Klc(MPa.\sqrt{m})$	$E(GPa)$	$G(GPa)$	ν
Glass	3·18	6·5	8·3	162	1·2	134	50	0·28
SiC*	3·21	4·0	21·6	450	3·5	420	180	0·16

*Data from *Céramiques et composites*, Tarbes, France, CCSiC-100.

according to the well-known analogy between Hookean elasticity and pure Newtonian flow by

$$\eta = \frac{\sigma}{2(1+\nu)\dot{\varepsilon}}$$

where σ is the applied stress, ν the Poisson's ratio and $\dot{\varepsilon}$ is the stationary strain rate.

The relaxation tests were conducted with a displacement imposed machine (Instron 8562) on the same type of rectangular bars as above.

4 Mechanical Properties

As shown in Fig. 1, the increase in Shear modulus, resulting from the increase in SiC volume fraction agrees well with the simple predictions for two-phase materials such as Voigt and Reuss models. The hardness increases also with increasing volume fraction of particles because of the much higher hardness of SiC.

As predicted by Evans[4] and shown by Swearingen *et al.*,[5] we observed here an increase of fracture toughness (Fig. 2) with an increasing addition of higher-modulus particles in a lower- modulus matrix and no significative dependence with particle size.

Concerning the fracture strength there is an important effect stemming from the particle size and volume fraction, with a remarkable increase in strength with increasing volume fraction (Fig. 2) and decreasing particle size (Fig. 3). Changes in the second phase characteristics might affect the composite Young's modulus, the critical flaw size which initiates fracture and the residual stresses that develop upon cooling.

The increase in strength with increasing volume fraction of reinforcement particles can be principally attributed to the resultant increase of Young's modulus.[6,7]

According to different studies on such composites,[7,8] particles introduce flaws in proportion to their size. Evaluation of the critical flaw size in our composites[1] showed that the flaw size increases with the particle size but does not show any significant dependence with the volume fraction, which corroborates these studies.

Compiling the results obtained by different authors concerning glass matrix composites, Nadeau and Dickson[9] concluded that internal stresses resulting from contraction mismatch could vary with the particle size. When the coefficient of thermal expansion of the glass is higher than that of the particles, three regimes of behaviour exist. For 300 μm particles the strength can be reduced, while for particles around 10 μm the best reinforcement possibilities are obtained. The transition between these two regimes occurs for particles of 20 to 40 μm in size. The reinforcement effects measured in our composites correspond well to these three size regimes.

5 Rheological Properties

Figure 4 shows that the apparent viscosity, obtained from compression creep tests at 930°C

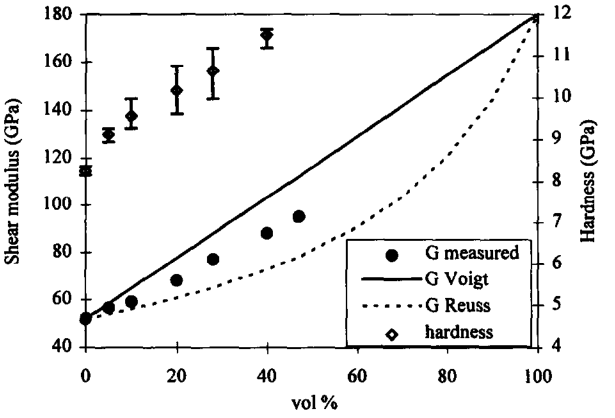

Fig. 1. Shear modulus and hardness as a function of SiC volume fraction.

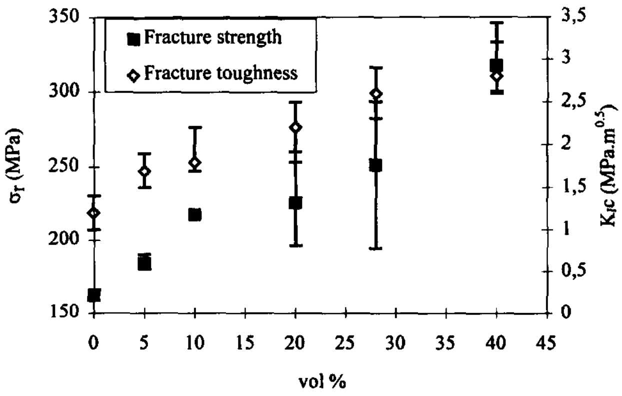

Fig. 2. Fracture strength and fracture toughness as a function of SiC volume fraction.

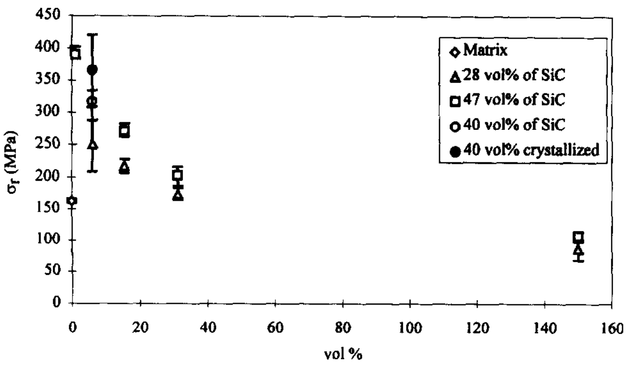

Fig. 3. Fracture strength as a function of particle size.

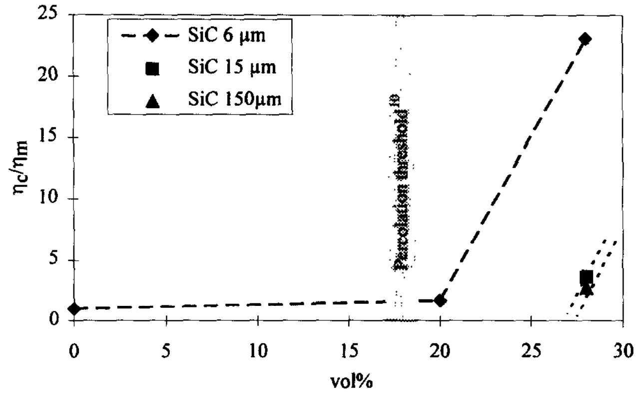

Fig. 4. Apparent viscosity as a function of SiC volume fraction.

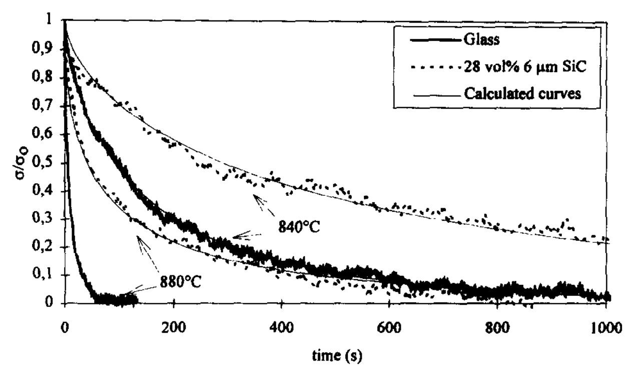

Fig. 5. Examples of experimental and calculated relaxation curves.

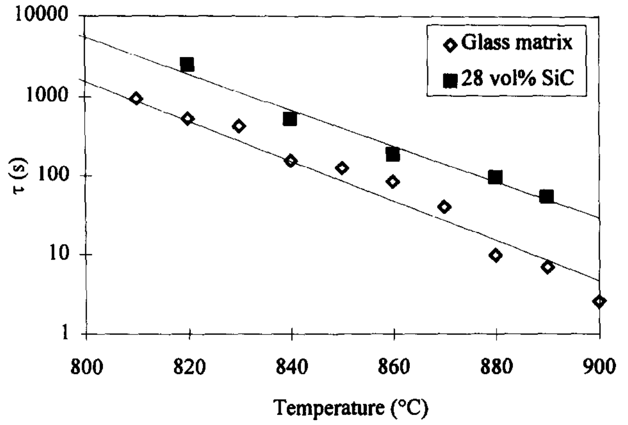

Fig. 6. Comparison of the dependence of the relaxation times with temperature.

under 50 MPa, increases with increasing volume fraction and decreasing size of the particles. The same type of behaviour was observed from 3 point-bend tests.

For the 6 μm SiC, a rigidity threshold is observed at approximately 20 vol%. It corresponds to the value predicted by Scherer[10] and observed by Dutton and Rahaman[11] or Tewari *et al.*[12] As observed by Dutton and Rahaman, the volume fraction of particles corresponding to the rigidity threshold increases with the increasing particle size. We can remark here that this increase is significant for increasing particle size up to 15 μm and very low for higher sizes.

A preceding study[13] of these materials showed that the dependence of the apparent viscosity on the temperature is the same for the glass and for the composite. Then the knowledge of the apparent viscosity for a given composition at a given temperature allows to predict the viscosity of the composite for the whole viscoplastic forming range.

The viscoplastic forming range corresponds to viscosities of less than 10^{12} Pa.s and lies between 870 and 1040°C where crystallization occurs.

Displacement-imposed 3 point-bend tests were conducted on the glass and on the 28 vol% 6 μm SiC composite to compare their mechanical relaxation behaviour. The experimental curves obtained could be fitted (Fig. 5) using the Kohlraush and Watt formula

$$\sigma/\sigma_0 = \exp(-t/\tau)^b$$

where τ is the relaxation time and b is close to 0·5. The comparison of the relaxation times (Fig. 6) confirms the results obtained with the creep study.

6 Conclusion

Because of the direct contradiction that exists between the best possible mechanical properties and an easy viscoplastic forming for particle-reinforced glass composites, a compromise must be found based on their dependence with the volume fraction and size of the particles.

The viscoplastic forming was tested on a 40 vol% 6 μm SiC composite.[13] A parabolic shell was shaped at 980°C within 30 min, starting from a $40 \times 40 \times 2\,\text{mm}^3$ plate. The possibility of a post-forming crystallization of the matrix was shown and gave a 15% increase in the fracture strength of the composite (Fig. 3).

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