Toughening and Strengthening of Alumina with Silver Inclusions

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Abstract

The fracture toughness and flexural strength of Al_2O_3/Ag composites are investigated. Both the strength and toughness of alumina are enhanced as silver inclusions are added. From microstructural observation, the crack is bridged or deflected by silver inclusions. The toughness enhancement is contributed mainly by the plastic deformation of the silver inclusions. The strengthening effect is contributed both by the matrix grain refinement and by the toughness enhancement.

1 Introduction

The application of ceramics as structural components is often limited by their brittleness. A major research objective has therefore been to improve their toughness. Previous studies have demonstrated that brittle solids can be toughened by incorporating metallic inclusions into them.[1-7] Similar to the zirconia toughened ceramics (ZTC),[8] the toughness of the metal-toughened ceramics can be described as

$$K_{IC,composite} = K_{IC,matrix} + \Delta K_{IC,metal} \qquad (1)$$

where $K_{IC, composite}$ is the toughness of the composite, $K_{IC,matrix}$ the toughness of the matrix and, $\Delta K_{IC,metal}$ the toughness increase contributed by the metallic reinforcement. The toughness enhancement for the ceramic–metal composites is believed to be contributed by the plastic deformation of metallic inclusions.[9-11]

As metallic inclusions are embedded within ceramic matrices, the metallic inclusion is constrained by the brittle and rigid matrix. The deformation behaviour of the constrained inclusion has been investigated by Ashby *et al.*[9-10] They suggested that the toughness increase, contributed by plastic deformation, $\Delta K_{IC,plastic\ deformation}$, is proportional to square root of the product of the volume fraction, F, and the inclusion size, d, as

$$\Delta K_{IC,plastic\ deformation} = C\,(F\,d)^{1/2} \qquad (2)$$

where C is a constant. The constant depends on the bonding characteristics. Deve *et al.* have suggested that the toughness enhancement is increased with the increase of the ductility of metallic inclusion.[11] Among metals, silver is superior for its ductility. Furthermore, silver is chemically stable up to temperatures higher than its melting point, 960°C. Therefore, the silver containing composites can be prepared by pressureless sintering in air.[12,13]

The mechanical properties of Al_2O_3/Ag composites have been studied recently.[12,13] In these studies, the toughness is determined by the indentation technique. As we are going to show later, this technique tends to over-estimate the toughness of Al_2O_3/Ag composites. In the present study, the toughness of the Al_2O_3/Ag composites is determined by the single-edge-notched-beam (SENB) technique. Furthermore, the effect of silver inclusions on the strength of alumina is not determined in the previous studies. In this study, the strength of the Al_2O_3/Ag composites is also measured.

2 Experimental Procedures

Alumina (AKP-50, Sumitomo Chem. Co. Ltd., Japan) and various amounts of silver oxide (Ag_2O, Johnson Matthey Co., UK) were milled in ethyl alcohol for 4 h. The slurry of the powder mixtures was dried with a rotary evaporator. The dried lumps were crushed and sieved. The powder compacts were prepared by pressing uniaxially at 56 MPa. The sintering was performed in air at 1600°C or 1700°C for 1 h. The heating and cooling rates were 5°C/min. In order to determine the toughness of alumina as a function of grain size, the alumina specimens with different grain size were prepared by sintering the alumina compacts at 1400–1700°C for 1 h. These Al_2O_3 specimens

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were prepared with the same procedures as those for the Al_2O_3/Ag composites.

The decomposition behaviour of silver oxide was determined by a differential thermal analyzer (DTA, Netsch Co., Germany). Phase identification was performed by X-ray diffractometry (XRD). The final density was determined by the water displacement method. Before submerging the specimens in water, a wax was applied to the surface to prevent water penetration. The microstructure was observed by a scanning electron microscope (SEM) and a transmission electron microscope (TEM). Both microscopes were equipped with an energy dispersive X-ray spectrometer (EDX). Polished surfaces were prepared by grinding and polishing with diamond paste to 6 μm and subsequently with silica suspension to 0·05 μm. Since the partial pressure of liquid silver at elevated temperature is high (10 mmHg/1600°C and 23 mmHg/1700°C),[14] a silver-free surface layer is formed.[13] After removing the silver-free layer by grinding, the volume fraction of silver after sintering was recalculated by counting the point fraction on the polished surfaces. The size of silver inclusions was determined by using the linear intercept technique. The dihedral angle of the silver inclusion within alumina matrix was estimated from polished microstructures by applying a stereological counting technique.[15] The polished specimens were thermally etched at 1500°C for 1 h to reveal the grain boundaries of alumina. The size of the matrix grains was also determined by the linear intercept technique. More than 200 grains or inclusions were counted.

The sintered specimens were machined longitudinally with a 325 grit resin-bonded diamond wheel at cutting depths of 5 μm/pass. The silver-free layer was removed during the grinding procedure. The final dimensions of the samples were 3 mm × 4 mm × 36 mm. The strength of the samples was determined by the 4-point bending technique at ambient conditions. The upper and lower spans

Fig. 2. The size of silver inclusions found in the composites shown in Fig. 1.

were 10 and 30 mm, respectively. The rate of loading was 0·5 mm/min. The fracture toughness was determined by the single-edge-notched-beam (SENB) technique. The notch was generated by cutting with a diamond saw. The width of the notch was around 0·5 mm. The indentation technique was also used to determine the toughness of the composites. The load applied was 50 N.

3 Results and Discussion

The DTA analysis suggests that the silver oxide starts to decompose from 410°C. The XRD patterns indicate that there is no residual silver oxide in Al_2O_3/Ag composites after sintering. The relative density of the composites is shown as a function of silver content in Fig. 1. The size of silver inclusions is shown as a function of silver content in Fig. 2. The grain size of alumina matrix is shown in Fig. 3. Typical microstructures for the composites are shown in Fig 4. The dihedral angle of the silver inclusion within the alumina matrix is $110\pm7\cdot8°$ for the composites sintered at 1600°C and $101\pm5\cdot5°$ for the composites sintered at
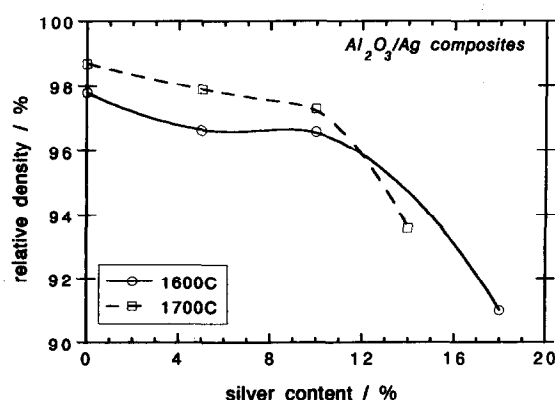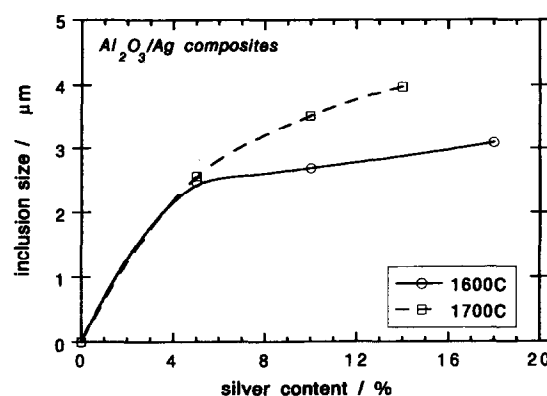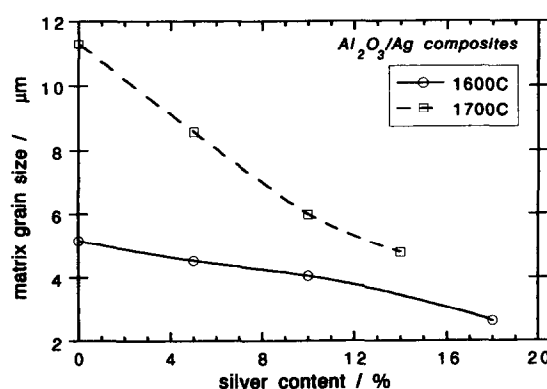

Fig. 1. The density of the composites as a function of silver content with the samples sintered at the indicated temperatures for 1 h.

Fig. 3. The size of alumina matrix found in the composites shown in Fig 1.

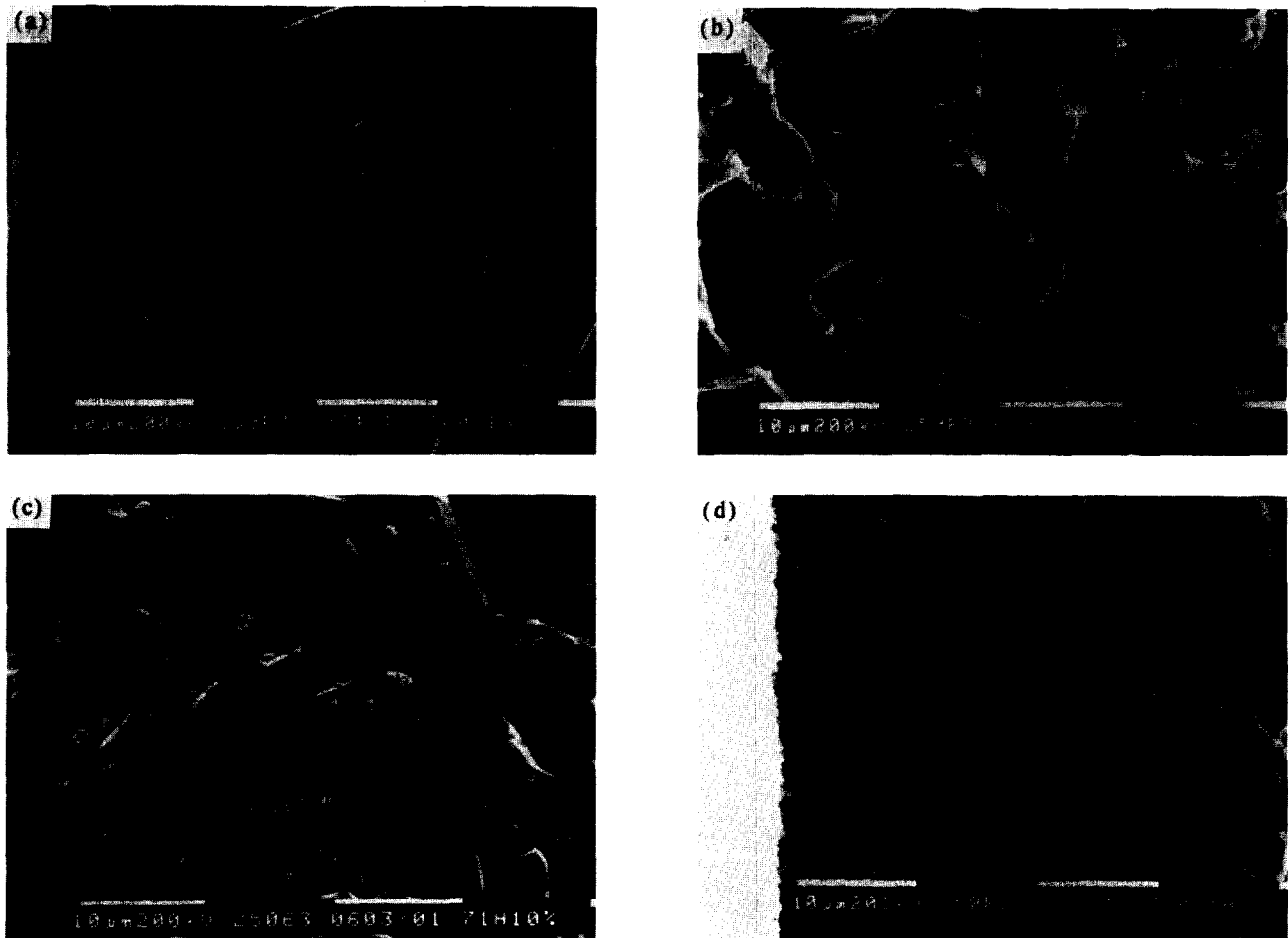**Fig. 4.** Typical microstructures of Al_2O_3/Ag composites. The vol.% of silver in the composites shown is (a) 0, (b) 5, (c) 10 and (d) 14. The composites are sintered at 1700°C for 1 h.

1700°C. These values are close to the reported values determined by the sessile drop technique.[16] Since the wetting is poor, silver melt behaves as an inclusion to the alumina matrix. The densification of the alumina matrix is thus hindered. From Fig. 4, most silver inclusions are located at the grain boundaries of alumina, the grain growth of alumina matrix is slowed down due to the presence of silver inclusions.

Since the thermal expansion of silver (22/MK) is bigger than that of alumina (9/MK), thermal stress results after the cooling step. Twins can be observed within the silver inclusions, (Fig. 5). Due to the fact that the twins are lens-shaped, they are deformation twins.[17] It suggests that the silver inclusions are highly constrained due to the presence of residual stress. Occasionally, small particles can be observed near the Al_2O_3/Ag interface, (Fig. 5). The small particles analyzed by EDX are mainly composed of Cr and Fe. It may be from the contamination from the steel mold used for forming. From the TEM study, no reaction layer can be observed at the interface and no Al detected within the silver inclusions. Since the solubility of alumina in Ag is negligible, it further confirms that the silver melt can not act as an effective liquid

phase to promote the densification.

The fracture toughness of the composites is shown as a function of silver content in Fig. 6. The interactions between a crack and silver inclusions are shown in Fig. 7. In the figure, the crack is bridged by, or deflected by, the silver inclusions. It suggests that the toughening mechanisms for the composites are plastic deformation and crack deflection. Both the plastic deformation[9–11] and crack deflection[18] mechanisms can contribute to the toughness enhancement. The toughness of Al_2O_3/Ag composites determined by the SENB and indentation techniques is thus higher than that of Al_2O_3 alone. However, the toughness determined by the indentation technique results in higher values. The crack opening introduced by the indentation is much smaller than the notch width produced by cutting with a diamond saw. Since the crack surfaces with a small opening are bridged by silver inclusions (Fig. 7), the propagation of the crack with a small opening is more difficult. Therefore, the toughness determined by the indentation technique is much higher than that determined by the SENB technique.

In order to determine the toughness of alumina matrix, $K_{IC,matrix}$, the toughness of the alumina

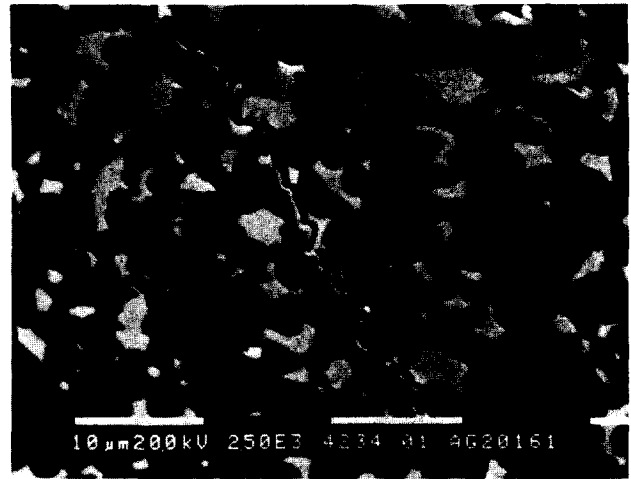Fig. 5. A highly constrained silver inclusion found within the alumina matrix.

Fig. 7. The interactions between silver inclusions and a crack introduced by indentation.
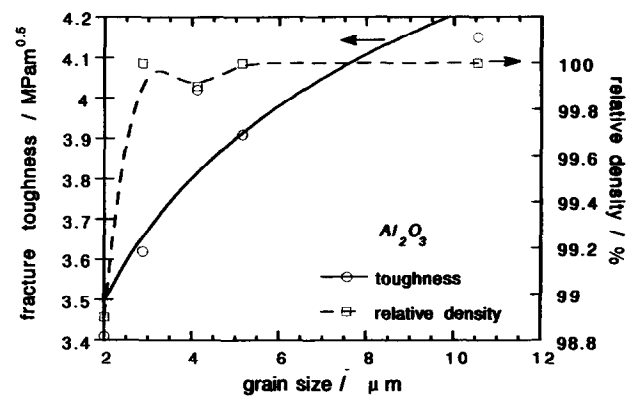

Fig. 8. The fracture toughness determined by the SENB technique and the density of Al_2O_3 specimens as a function of grain size.

specimens sintered at various temperatures is determined (Fig. 8). The toughness is determined by the SENB technique. The density of the Al_2O_3 specimens is varied from 98·9 to 99·9%; therefore, the effect of porosity on the toughness can be ignored. The toughness of alumina is increased with the increase of grain size. It is similar to the

results reported by Tuan et al.[19] According to eqn (1), the toughness increase, $\Delta K_{IC,metal}$, contributed by the metallic inclusions which can be determined as $K_{IC,matrix}$ is known. The toughness increase of the composites is shown as a function of the square root of the product of volume fraction, F, and inclusion size, d, in Fig. 9. Since the enhancement from the deflection around spherical inclusions is limited,[18] more significance is to be given to the
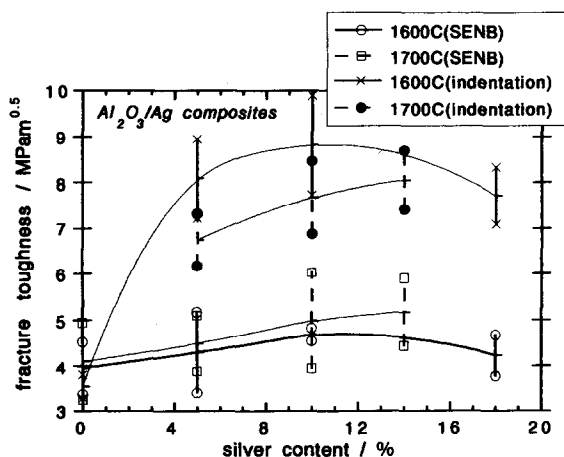

Fig. 6. The fracture toughness determined by the SENB and the indentation techniques for the composites shown in Fig. 1 as a function of silver content.
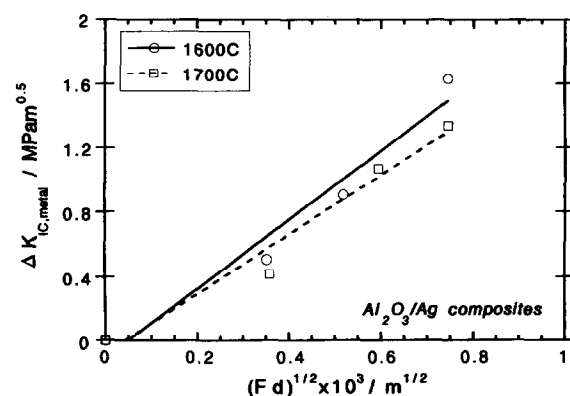

Fig. 9. The fracture toughness increase as a function of the square root of the product of volume fraction and inclusion size. The sintering temperatures are indicated.
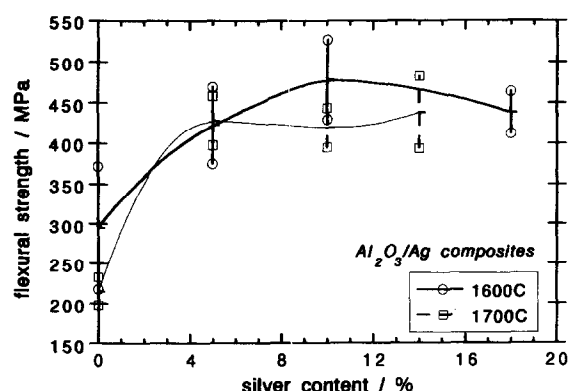
Fig. 10. The flexural strength for the composites shown in Fig. 1 as a function of silver content.

plastic deformation mechanism. This is supported by the linear relationship between the toughness increase and the square root of the product of volume fraction and inclusion size (Fig. 9).

The flexural strength of the composites is shown as a function of silver content in Fig. 10. With the enhanced toughness and the reduced grain size of alumina matrix as silver is added, the strength of alumina is improved substantially.

4 Conclusions

Dense Al_2O_3/Ag composites are prepared by pressureless sintering at 1600°C and 1700°C for 1 h. The densification behaviour, microstructure development, and mechanical properties of the composites are investigated. Even though the silver inclusions are melted during sintering, due to the fact that the wetting of silver melt on alumina is poor and the solubility of alumina in silver is limited, the presence of silver inclusions hinders the densification. The matrix alumina grains are refined due to the grain boundaries being dragged by the silver inclusions. The addition of ductile inclusions not only toughens but also strengthens the ceramic matrix. The toughening is contributed mainly by the plastic deformation of silver inclusions. Due to the increase in toughness and the refinement of matrix grain size induced by the presence of the inclusions, the strength of the alumina is also significantly increased.

Acknowledgements

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