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The preparation of Al_2O_3/Ni composites by a powder coating technique

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In the present study, nickel particles are coated onto the surface of alumina powder by an impregnation technique. The densification behaviour and the microstructural evolution of the nickel coated alumina powder during sintering are investigated. The strength and the toughness of the resulting Al_2O_3/Ni composites are determined. As the nickel content is less than 13 vol %, fully dense composites can be prepared by pressureless sintering. The matrix grain size decreases as nickel inclusions are added. The strength and the toughness of alumina can be increased by 23 and 42% by adding 5 and 8 vol % nickel, respectively. The toughening effect is attributed to plastic deformation of ductile inclusions and crack deflection by the inclusions. The strengthening effect is attributed to microstructural refinement.

1. Introduction

Brittle ceramics can be toughened by the incorporation of ductile metallic inclusions. This approach has been successfully employed in several systems [1–3]. The enhancement of toughness is attributed to a crack bridging mechanism [4–6]. However, the metallic inclusions have to be firmly bonded to the brittle matrix to start the bridging mechanism. If the ductile inclusion is weakly bonded to the matrix, the crack will propagate along the interface, and the contribution from ductility to the toughening enhancement will be negligible. If bonding between the ceramic and metal is very strong, inclusion deformation is limited to a very small section. For bonding with intermediate strength, a fraction of the interface is debonded as the crack reaches the interface; the effective bridging length is thereby increased with the improvement in toughness. Therefore, moderately weak interfaces are expected to result in high toughness composites.

Since a thermal expansion mismatch exists between the ceramic matrix and the metallic inclusions, internal stresses are set up within and around the metallic inclusions. As the thermal expansion coefficient of the ductile inclusions is bigger than that of the brittle matrix, the ceramic–metal interface is subjected to a radial tensile stress. If the size of the inclusion is bigger than the critical value, a microcrack at the interface generates spontaneously as the composite cools from the firing temperature. The detached ductile inclusions cannot be deformed plastically.

The nickel toughened alumina system has been investigated by Tuan and Brook [7, 8]. Their results suggest that the toughness of alumina can be enhanced by increasing the nickel content. However, as the inclusion is bigger than 2.5 μm, the toughness decreases dramatically [8]. It implies that by keeping the inclusion size smaller than the critical value, the toughness of alumina can be further enhanced by adding more nickel. The specimens used in their studies were prepared by reducing powder mixtures of Al_2O_3 and NiO [7, 8]. The size of nickel inclusions is thus limited by the size of NiO particles. Besides, in their studies, the effect of nickel inclusions on the strength of alumina is not determined. In the present study, Al_2O_3/Ni composites are prepared by a powder coating technique. This technique can result in smaller inclusions. The strength of the Al_2O_3/Ni composites is also determined.

2. Experimental procedure

The nickel coated powder was prepared by impregnating alumina powder in a nickel nitrate solution. The alumina powder used was α-Al_2O_3 (TM-DR, Taimei Chemical Co., Ltd, Tokyo). The zeta potential of the alumina slurry as a function of pH value was measured by the electrophoretic laser light scattering method (ELS-80, Otsuka Electronics Co., Ltd). The dispersion of alumina powder was carried out by ball milling the alumina slurry at pH 9. The grinding media used was zirconia balls. After the alumina slurry had been milled for 4 h, a suitable amount of nickel nitrate was added into the slurry. The mixture was further ball milled for 2 h. The slurry was dried slowly by constant heating and stirring at 50 °C. The dried powder was reduced in a hydrogen atmosphere at 500 °C for 12 h. The resulting powder was crushed and passed through a plastic sieve. Powder compacts

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were formed by first uniaxially pressing at 10 MPa, then by cold isostatic pressing at 250 MPa. The sintering was performed in hydrogen at 1600 °C for 1 h. The heating and cooling rates were 5 °C min^{-1}.

The sintered composites were machined longitudinally with a 325 grit resin-bonded diamond wheel at cutting depths of 5 μm pass^{-1}. The final dimensions of the specimens were $3 \times 4 \times 36\ \text{mm}^3$. The strength of the specimens was determined by the four point bending technique. The upper and lower spans were 10 and 30 mm, respectively. The rate of loading was 0.5 mm min^{-1}. The fracture toughness was determined by the single-edge-notched beam (SENB) technique.

Phase identification of the reduced powder and the sintered specimens was performed by X-ray diffractometry. The morphology of the coated powder was observed by transmission electron microscopy (TEM). The resistivity of the composite was measured by a d.c. two-probe technique, using silver paint for the contacts. The final density of the specimens was determined by a water displacement method. Before submerging the specimens in water, a wax was applied to the surface to prevent water penetration. The polished surfaces were prepared by grinding and polishing with diamond paste to 6 μm and with silica suspension to 0.05 μm. The size of nickel inclusions after sintering was determined by using the linear intercept technique. The polished specimens were then thermally etched to reveal the grain boundaries of the matrix. The thermal etching was carried out at 1450 °C for 30 min. in H_2. The size of the matrix grains was also determined by using the linear intercept technique.

3. Results and discussion

The iso-electric point of the alumina used is at pH 8. Dispersion of the alumina powder in water can thus be carried out at a pH value other than 8. In order to deposit positive nickel ion on the negatively charged alumina particles, the pH value of the alumina slurry is adjusted to 9. After the dried powder is reduced in hydrogen, only nickel and alumina can be detected by X-ray diffractometry. A TEM micrograph of the coated powder is shown in Fig. 1. On the surface of the alumina particles, small nickel particles can be observed.

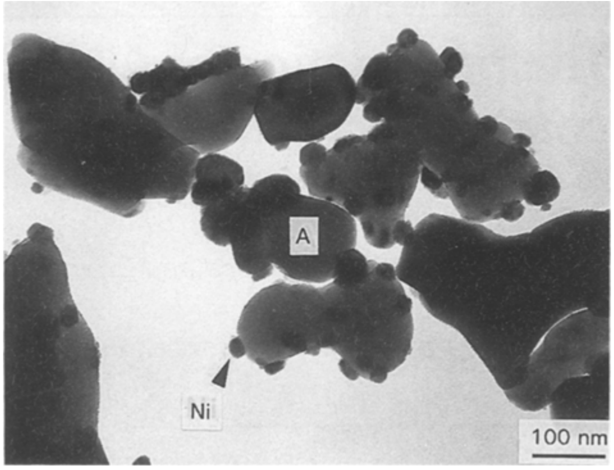

Figure 1 The morphology of the nickel (Ni) coated alumina powder. The powder contains 8 vol % Ni: (A) alumina grain (scale bar = 100 nm).

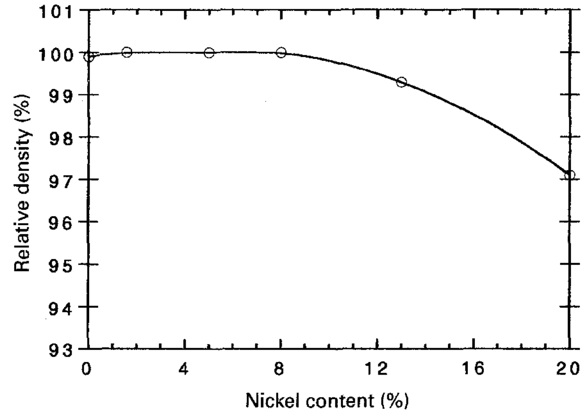

Figure 2 Relative density of the Al_2O_3/Ni composites as a function of nickel content.

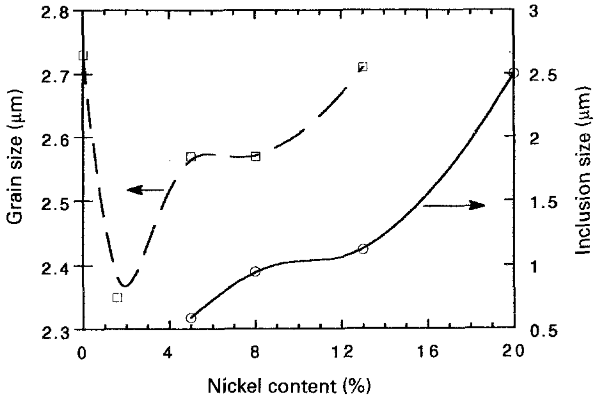

Figure 3 Inclusion size of nickel (○) and the grain size of the alumina matrix (□) as a function of nickel content.

The relative density of the composites is shown as a function of nickel content in Fig. 2. Fully dense Al_2O_3/Ni composites can be prepared as long as the nickel content is lower than 13 vol %, despite wetting of nickel melt on alumina being poor [9]. Furthermore, the solubility of alumina in the nickel melt is negligible [10]; the nickel melt behaves as a solid inclusion instead of as a liquid-phase sintering aid. The presence of the inclusions can thus prohibit densification of the matrix [11]. The inclusion size of nickel and the grain size of the alumina matrix are shown as a function of nickel content in Fig. 3. The inclusion size increases with the increase of nickel content. With a small amount of nickel addition, the grain size of matrix decreases. Typical microstructures of the composites are shown in Fig. 4. The inclusion size in the composite containing 1.6 vol % nickel is too small to be observed. For the composite containing 20 vol %, nickel, the thermally etched surface is covered by nickel spheres. The grain size of the composite is thus not determined. From the micrographs, most of the nickel inclusions are located either at the grain boundaries or at the grain junctions. Movement

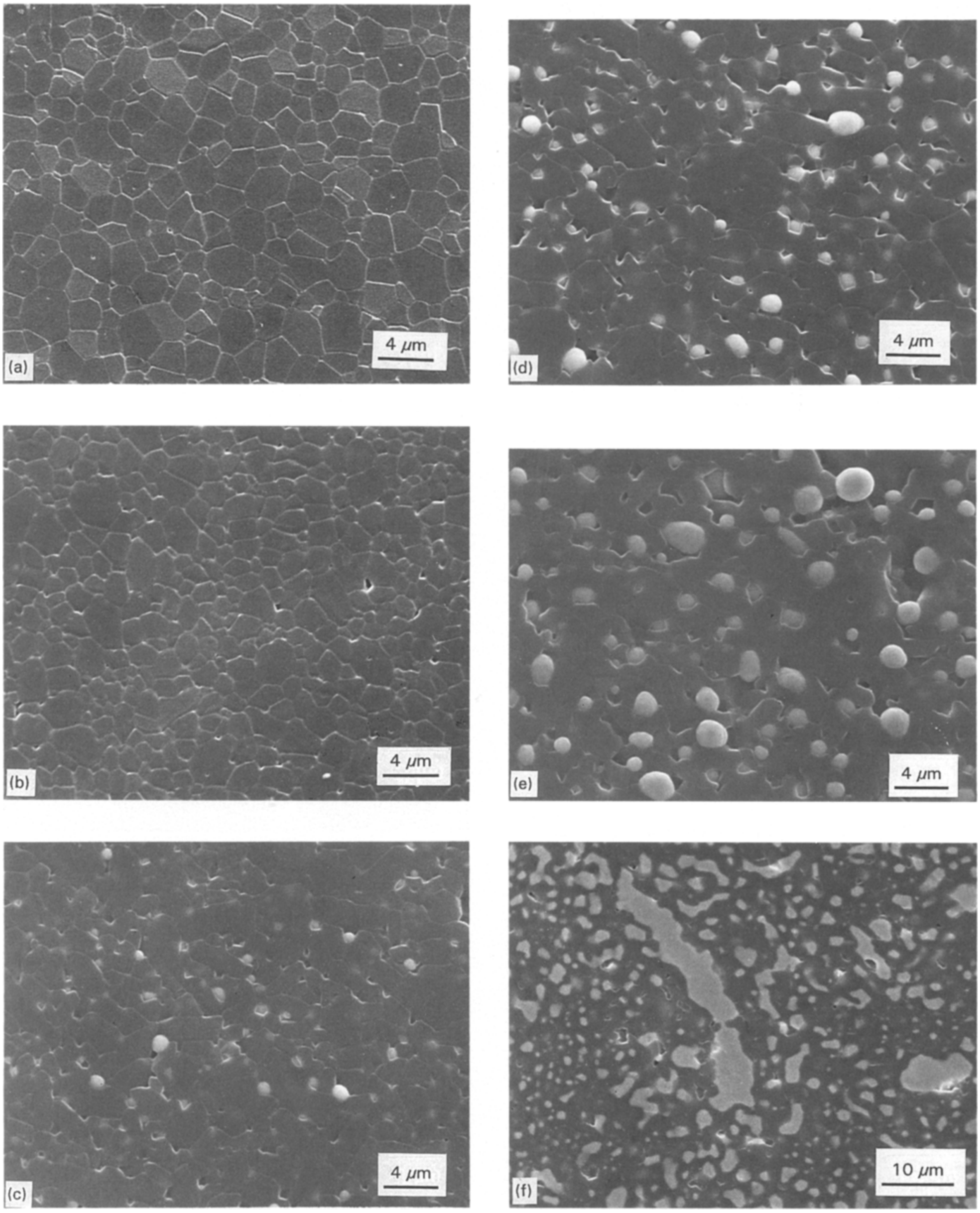

Figure 4 Typical microstructures of the Al_2O_3/Ni composites. The nickel content in the composites is (a) 0, (b) 1.6, (c) 5, (d) 8, (e) 13, and (f) 20 vol %, respectively. The composite containing 20 vol % Ni is not thermally etched.

of grain boundaries is retarded due to the presence of the nickel inclusions. The grain size of alumina matrix is thus smaller than that of pure alumina. Furthermore, the inclusion size is proportional to the matrix grain size, Fig. 3; coarsening of the nickel inclusions is likely a coalescence process [11]. Occasionally, small nickel inclusions can be observed within alumina grains, Fig. 5. The resistivity of the composites is shown as a function of nickel content in Fig. 6. The resistivity drops, as the nickel content increases above 13 vol %, suggesting that an interconnected nickel network is formed.

The strength and the toughness of the composites are shown as a function of nickel content in Figs 7 and 8. Each point in the figures represents the average value of three to four specimens. The error bars

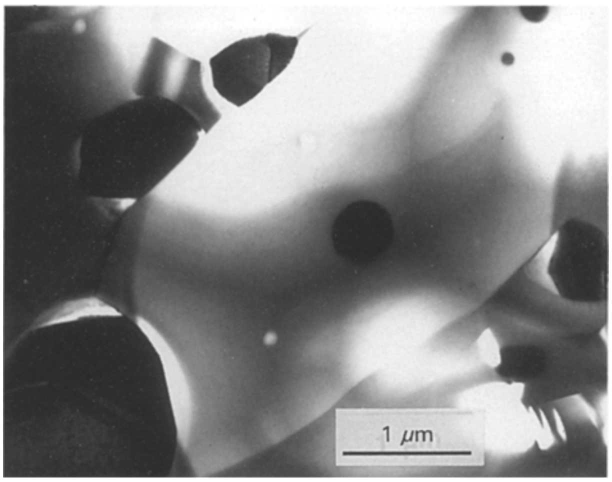

Figure 5 TEM micrograph for the composite containing 13 vol % nickel.

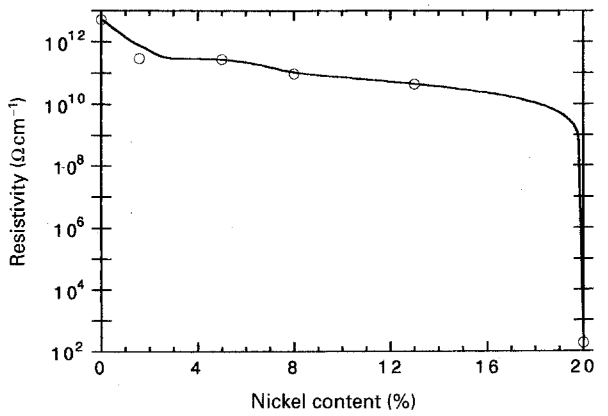

Figure 6 Resistivity of the Al_2O_3/Ni composites as a function of nickel content.

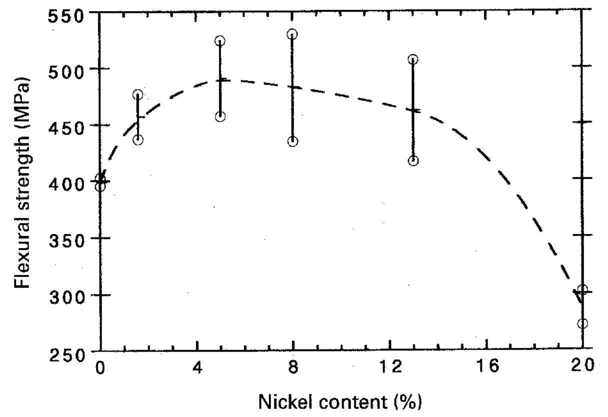

Figure 7 Flexural strength of the Al_2O_3/Ni composites as a function of nickel content.

indicate one standard deviation. Except the composite containing 20 vol % nickel, both the strength and the toughness of alumina increase with increasing nickel content. With 5 vol % nickel addition, the strength of alumina increases by 23%. With 8 vol % nickel addition, the toughness of alumina is enhanced by 42%. For the composite containing 20 vol % nickel, the nickel inclusions are interconnected to each other, Fig. 6. For the composite, the possibilities of the crack interacting with the inclusions is very high. The ductility of the nickel inclusions can thus be fully employed. However, the mechanical properties of the composite are very disappointing. Since the inclusion size of the composite is bigger than the critical value, Fig. 3, a microcrack is formed at the interface. Furthermore, the density of the composite is low, Fig. 2.

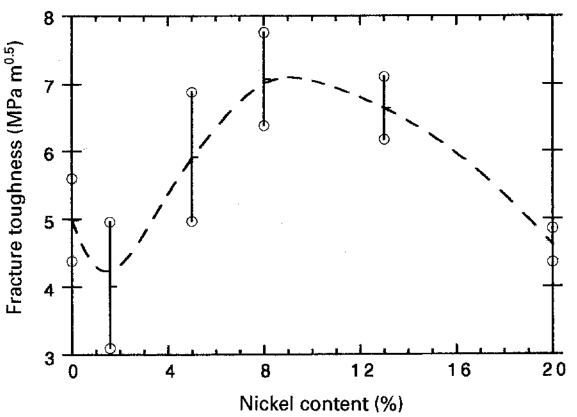

Figure 8 Fracture toughness of the Al_2O_3/Ni composites as a function of nickel content.

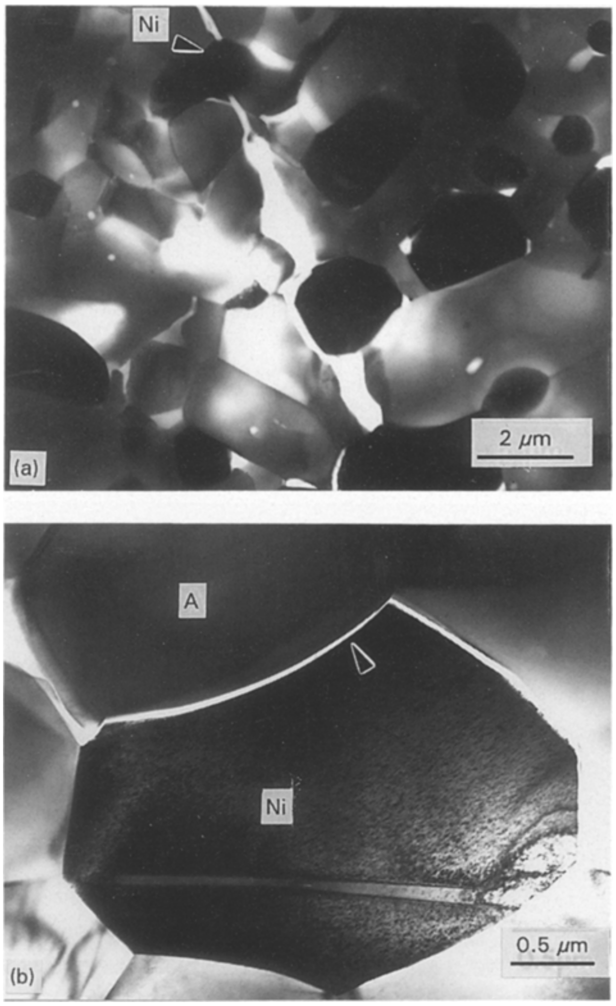

Figure 9 (a) Bridging nickel particle, and (b) microcrack observed in the composite. The bridging particle and the microcrack are indicated by arrows.

From microstructural observation, Fig. 9, the nickel inclusions are either stretched or intact, suggesting that the crack is either bridged or deflected by the inclusions. A microcrack can be observed at the interface of the coarse inclusion, Fig. 9b. The major crack is deflected due to the presence of the microcrack. The toughness enhancement is attributed to crack bridging and crack deflection. As far as the strength of the composites is concerned, since the alumina matrix grain size decreases as nickel is added, the composites are strengthened due to microstructural refinement.

4. Conclusions

In the present study, nickel particles are coated on the surface of alumina particles by an impregnation technique. The Al_2O_3/Ni composites are prepared by sintering pressurelessly the coated powder. As the nickel content is less than 13 vol %, fully dense composites with an inclusion size smaller than the critical value, 2.5 μm, are prepared. Both the strength and toughness of alumina increase due to the addition of nickel inclusions. Toughness enhancement is attributed to crack bridging and crack deflection; strengthening is attributed to matrix grain refinement. As 20 vol % nickel is added to alumina, the nickel inclusions form interconnected networks. However, due to this, the inclusions are larger than the critical size and the density is low; the toughness and the strength of the composite are low.

Acknowledgements

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