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Mechanical properties of Al_2O_3 particle–Y-TZP matrix composite and its toughening mechanism

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Dense Al_2O_3 particle–Y-TZP matrix (Al_2O_3 < 40 vol %) composite was prepared by pressureless sintering at 1550 °C. Composites with 10–30 vol % Al_2O_3 particles showed enhanced fracture toughness, bending strength and Vicker's hardness as compared to single-phase Y-TZP. The highest strength (1150 MPa) and highest toughness (12.4 MPa $m^{1/2}$) were obtained for the composite containing 10 vol % Al_2O_3. It was found that, in addition to the contribution by the crack-deflection effect, the enhanced phase transformation from tetragonal to monoclinic during fracture was the main toughening mechanism in operation in the composites.

1. Introduction

Y-TZP (Y_2O_3-stabilized ZrO_2 polycrystals) is a ceramic material of high toughness and strength at room temperature [1–3]. It has also been found that the material can be strengthened at high temperature by the addition of some refractory particles such as SiC and Al_2O_3, and the addition of Al_2O_3 particles can even enhance the room-temperature strength and toughness as well [4–6]. Unfortunately, the addition of secondary rigid particles may impede the densification process and thus the composite can only be densified by hot-pressing or post-hot isostatic pressing (HIPing) [6–8], and the mechanisms for the mechanical property improvement by secondary phases are not well understood [7]; some misunderstanding even exists in the literature [6]. It was the purpose of the present work to densify the composites by pressureless sintering and to find convincing evidence for the toughening mechanism of the composite.

2. Experimental procedure

Superfine Y-TZP (3 mol % yttria) powder was prepared by the coprecipitation method using zirconium oxychloride and yttrium chloride as starting materials and ammonia solution as the precipitation medium. The pH value during coprecipitation was maintained to be greater than 9. Cl^- ions were removed completely by washing with distilled water and the coprecipitate was then dispersed to prevent the formation of hard agglomerates. The precipitated precursor was then dried at 120 °C overnight and calcined at 750 °C for 2 h. The primary particle size of the obtained powder was about 20 nm [9]. The Al_2O_3 powder used was from Sumitomo Chemical Co.(AKP-50) and the average particle size of the powder was about 0.23 μm. The Al_2O_3–Y-TZP composite powders were prepared by ball milling, in distilled water, of the two kinds of powders using Al_2O_3 balls as the milling medium. The five batches of the composite powders prepared contained 0, 10, 20, 30 and 40 vol % Al_2O_3, respectively.

The composite powders were dry pressed into discs of diameter 40 mm and isostatically pressed at 250 MPa. Binders were first burnt out and the samples were then pressurelessly sintered at 1550 °C for 2 h. The densities of the composites were measured using Archimedes' method in distilled water and machined into dimensions 2.5 mm × 5 mm × 26 mm. Fracture strength was measured using the three-point method with a span of 20 mm. Fracture toughness was determined by the single-edge notched bending (SENB) method with a notch width of about 0.2 mm. All these measurements were carried out on an Instron 1195 material tester. Vicker's hardness was determined with the micro-indentation method on Asaki AVK-A micro-hardness tester with a constant load of 98 kN. The microstructures of the composites were observed on the polished surfaces thermally etched at 1300 °C for 2 h, under a Jeol electron micro-probe instrument (JCXA-733). The phase compositions on the polished surfaces and on the fractured surfaces were determined on RAX-10 X-ray diffractometer (Rigaku Denki) [10].

3. Results

3.1. Sintered densities of the composites

Fig. 1 shows the relative densities of the sintered composites at 1550 °C. The relative densities were calculated on the basis of the theoretical densities of 6.08 and 3.98 g cm^{-3} for Y-TZP and alumina, respectively. It can be seen that the relative densities are above 98.5% when the alumina content is not higher than 30 vol %, with an optimum value at 10 vol %

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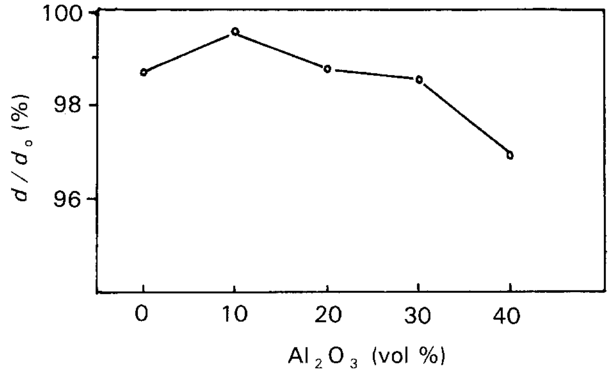

Figure 1 Sintered densities of the Al_2O_3-Y-TZP composites at 1550 °C for 2 h.

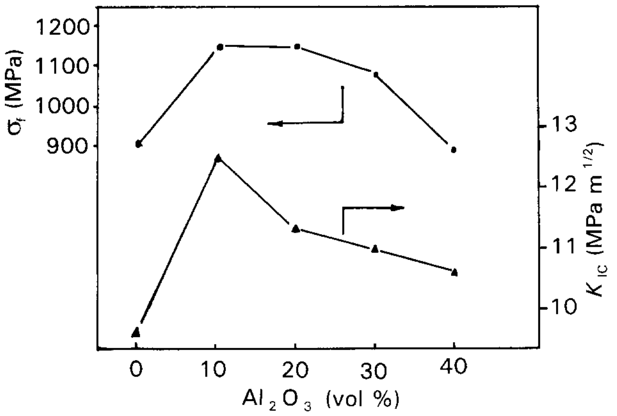

Figure 2 (●) Fracture strength, σ_f and (▲) fracture toughness, K_{1c} versus Al_2O_3 content.

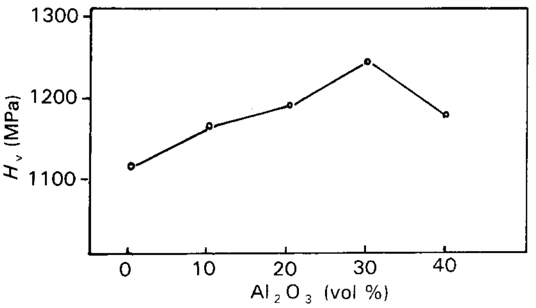

Figure 3 Vicker's hardness versus Al_2O_3 content.

Al_2O_3, but an obvious decrease is found when the Al_2O_3 content is increased to 40 vol %.

3.2. Mechanical properties of the composites

The fracture strength, (σ_f), and toughness, (K_{1c}), of the composite ceramics are shown in Fig. 2. Both fracture strength and toughness increase for the composites after addition of appropriate contents of the secondary alumina particles. The maximum values of strength and toughness were obtained for the composite material containing 10 vol% Al_2O_3, showing an increase of 28% and 30%, respectively, when compared with pure 3Y-TZP. Beyond 30 vol % alumina content in the composite material, the strength decreases sharply while the toughness decreases slowly.

The Vicker's hardness values also increase almost linearly up to 30 vol % Al_2O_3 as shown in Fig. 3. The highest hardness was obtained at 30 vol % Al_2O_3 with a value of 1249 MPa. The hardness increase is probably due to the higher hardness of Al_2O_3 than Y-TZP in a well-sintered material.

4. Discussion

4.1. Effect of sintered density on the mechanical properties

Although the densification rate of the composites is reduced by the rigid Al_2O_3 particles, as reported in a previous paper [11], the densities of the composites sintered at 1550 °C for 2 h are equal to or higher than 98.5% when the alumina content is not higher than 30 vol %. The well-sintered high-density material ensures good mechanical properties are obtained. The much decreased value of strength with an Al_2O_3 content of 40 vol % is associated with its much lowered relative density value.

4.2. Effect of thermal expansion mismatch

The thermal expansion mismatch between particlates and matrix may lead to thermal residual stress. According to the report by Taya *et al.* [12] in the system of TiB particle–SiC matrix, the thermal expansion mismatch between TiB and SiC leads to the development of internal thermal residual stresses and the variation of the mechanical properties. In the TiB(p)–SiC(m) system, the thermal expansion coefficient of the matrix is lower than that of the TiB particles, the thermal expansion coefficient difference in such a system, according to the analysis by Taya *et al.*, leads to an average compressive stress in the matrix and, therefore, an apparent increase of the fracture toughness. While in the Al_2O_3 particles–Y-TZP system, the thermal coefficient of the Y-TZP matrix is obviously higher than that of Al_2O_3 particles, in the present system, the thermal residual stress status is also in contrast with the TiB(p)–SiC(m) system. In fact, the determination of the lattice parameters (plane distances) of the Y-TZP matrix in the composites shows no or very little alterations of the lattice parameters of the Y-TZP matrix with change in composition by the addition of Al_2O_3 particles, as illustrated in Table I.

TABLE I Relation between lattice parameters (plane distance *d*), and composition

Al_2O_3 content (%)	*d* (Å)	
	ZrO_2 (1 1 1)	ZrO_2 (2 2 0)
0	2.977	1.821
10	2.976	1.821
20	2.978	1.821
30	2.976	1.821
40	2.976	1.821

If a stress-free status is assumed for the present system before cooling from the sintering temperature (this may be true when differential shrinkage between Y-TZP and Al_2O_3 during densification is not considered because of the superplastic flow nature of Y-TZP at high temperature), the Y-TZP matrix in the present system should be subjected to a radial compressive stress and a tangential tensile stress, due to the differential shrinkage arising from the different thermal expansion coefficient when cooling to room temperature. This is in contrast to the situation analysed by Evans [13] for a system with differential shrinkage during heating. However, the measured lattice parameters for Y-TZP do not suggest the obvious presence of such a stress status. The tension and compression stress may cancel each other out, on the one hand, and, on the other, the superplasticity of Y-TZP [14] at above about 1350 °C will play a role in the stress relaxation.

The stress status analysed above suggests that the thermal residual stress in the present system may not contribute at all to the toughening of the composites.

4.3. Toughening contribution from crack deflection effect

Fig. 4 shows the microstructures on the polished surfaces of Y-TZP and 20 vol % Al_2O_3–Y-TZP ceramics and the mode of crack propagation in them. The crack propagation path is rather straight and no crack deflection effect can be clearly seen in Y-TZP; however, in the composite the crack is deflected by secondary Al_2O_3 particles (darker ones in the microstructure), which results in a somewhat rough fracture surface and leads to a higher fracture energy. As the fracture toughness, K_{1c}, is proportional to the square root of the fracture energy, γ_f, according to the Griffith equation

$$K_{1c} \propto \gamma_f^{1/2} \tag{1}$$

so the crack-deflection effect leads to enhanced toughening of the composite ceramics. It is known that the fracture strength, σ_f, is proportional to K_{1c} and inversely proportional to the square root of the defect dimension (defined as the half-length, c, of the defects)

$$\sigma_f \propto K_{1c}c^{-1/2} \tag{2}$$

therefore if the defect population and size are relatively constant, the material is also strengthened when toughened.

However, according to the theoretical analysis by Faber and Evans [15], the contribution to increasing toughness by the crack-deflection effect is somewhere around 10%, so the greater part of the total increase of 30% in fracture toughness on the addition of 10 vol % Al_2O_3 must be from other causes.

4.4. Toughening effect by phase transformation

According to the analysis by Lange [16] and Gao *et al.* [17] and many other authors, the energy for crack propagation absorbed by the stress-induced martensitic phase transformation depends on the amount transformed from tetragonal to monoclinic in the wake of the crack tip, thus the crack can be effectively stopped and the fracture energy be increased. A higher volume fraction of phase transformation will result in a stronger toughening effect. However, the addition of rigid secondary particles such as Al_2O_3 in Y-TZP was believed by Tsukuma *et al.* [6] to decrease the effect of phase transformation, and the toughening phenomenon of the composites by Al_2O_3 is attributed to the thermal expansion mismatch effect [7].

The present work provides some different information from that indicated above [6, 7]. Fig. 5 shows the monoclinic phase contents on the fractured surfaces and the polished surfaces for the composites. It indicates that the amount of phase transformation from tetragonal to monoclinic is promoted most effectively at an alumina content in the composite of 10 vol %, which corresponds to the highest fracture toughness and strength of the material. This agrees well with what would be expected from Equation 1. Because the densities of the materials remain high when the Al_2O_3 content is kept below 30 vol %, the strength of the composites are simultaneously strengthened. Up to an

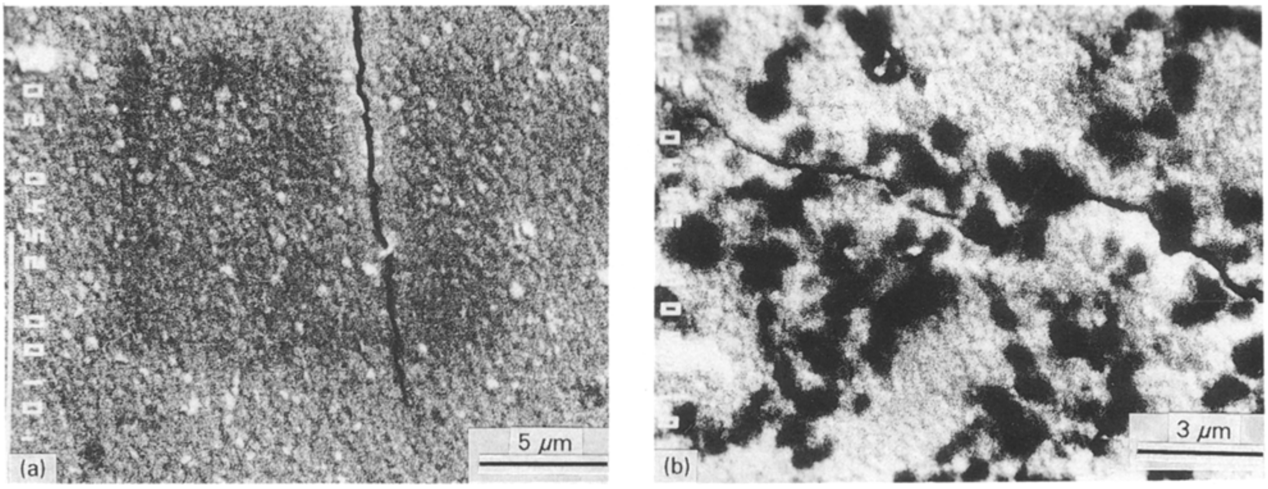

Figure 4 Crack propagation in (a) single-phase Y-TZP and (b) 20 vol % Al_2O_3–Y-TZP composites.

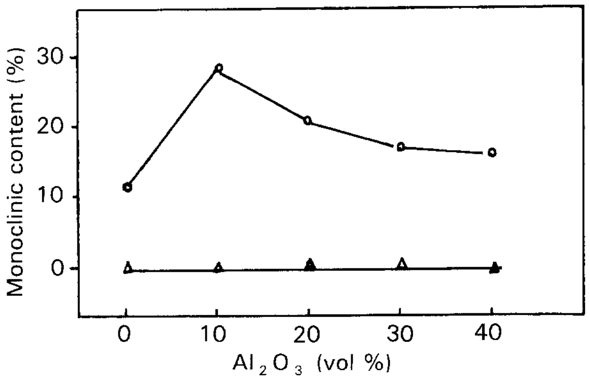

Figure 5 Monoclinic contents on (△) the polished surfaces and (○) fractured surface versus Al_2O_3 content.

Al_2O_3 content of 40 vol % in the composite, the decrease in density leads to a decrease in strength, as can be estimated from Equation 2. Therefore, it can be concluded that, in addition to the crack-deflection effect, the main contribution to the toughening of Al_2O_3–Y-TZP composites is the enhanced phase transformation from tetragonal to monoclinic on the addition of secondary Al_2O_3 particles to the Y-TZP. The underlying mechanisms for the increased phase transformation by Al_2O_3 particles are not well understood, but may be related to the thermal expansion mismatch and the crack-deflection effect.

5. Conclusions

1. The fracture toughness, strength and Vicker's hardness of Y-TZP materials can be effectively increased by the addition of secondary Al_2O_3 particles for the pressurelessly sintered dense Al_2O_3–Y-TZP composites.

2. It is proposed that the important mechanism for toughening and strengthening of the composites, in addition to the crack-deflection effect, is the enhanced phase transformation from tetragonal to monoclinic during crack propagation in the composites.

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