Comparison of fracture toughness evaluating methods in 3Y-TZP ceramics reinforced with Al$_2$O$_3$ particles

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3Y-TZP/Al$_2$O$_3$ composites containing 0 to 30 vol% alumina were fabricated by sintering at 1550 °C for 2 h. The fracture toughness was measured using a range of techniques. The fracture toughness measured using the single edge V-notched beam (SEVNB) method, the controlled surface flaw and indentation strength in bending methods decreased with increasing Al$_2$O$_3$ content. This can be explained by the increased critical transformation stress, decreased volume fraction of transformable t-ZrO$_2$ and increased residual tensile stress. In contrast, the value obtained by the indentation fracture (IF) method showed an increasing tendency. Accordingly, the IF method is unsuitable for evaluating the fracture toughness of 3Y-TZP/Al$_2$O$_3$ composites. The SEVNB method is considered one of the most reliable methods for determining the fracture toughness of ceramic materials.

Key words: 3Y-TZP/Al$_2$O$_3$ composites, Fracture toughness, Single edge V-notched beam, Residual stress.

Introduction

Zirconia (ZrO$_2$) ceramics are used widely in dentistry owing to their excellent mechanical properties [1]. It is believed that a stress-induced, tetragonal zirconia to monoclinic zirconia phase transformation can increase the fracture toughness. This phase transformation is accompanied by a substantial increase in volume (~ 4%), which can cause a compressive stress on a ground surface or in the vicinity of a crack tip. This clamping constraint on the crack tip needs to be overcome for an advancing crack to propagate [2]. 3 mol% Y$_2$O$_3$ stabilized ZrO$_2$ (3Y-TZP) ceramics have attracted particular attention due to the large range of solid solubility for yttria in tetragonal zirconia and the low eutectoid temperature (~ 550 °C), which avoids the decomposition of both tetragonal and cubic solid solutions during cooling from the sintering temperature [3].

The fracture toughness is an important mechanical property for the utilization of 3Y-TZP as dental ceramics. On the other hand, the toughness of 3Y-TZP ceramics can vary widely (from 4.25 to 10 MPam$^{1/2}$) depending on the fracture toughness evaluation methods used [4-8]. Moreover, confusing results concerning the effect of Al$_2$O$_3$ on the fracture toughness of Y-TZP ceramics have been reported. Santos et al. [9] reported that the addition of Al$_2$O$_3$ had no significant effect on the fracture toughness, whereas Choi and Bansal [10] stated that the fracture toughness increased with increasing Al$_2$O$_3$ content. Nevarez-Rascon et al. [11] found that 3Y-TZP containing 20 wt% Al$_2$O$_3$ exhibited the maximum fracture toughness of the samples examined. These discrepancies might be due to different microstructures, such as grain size, porosity, purity, Al$_2$O$_3$ particle size. Therefore, it is necessary to clarify the effect of an Al$_2$O$_3$ addition on the fracture toughness of ZrO$_2$/Al$_2$O$_3$ composites.

The single edge V-notched beam (SEVNB) method is considered one of the most reliable methods for evaluating the fracture toughness of ceramics. In the present study, alumina-reinforced zirconia composites, containing 0 to 30 vol% alumina, were fabricated by sintering. This study evaluated the fracture toughness of 3Y-TZP/Al$_2$O$_3$ ceramics using the SEVNB method, and compared the results with those obtained using different methods, such as indentation fracture (IF), indentation strength in bending (ISB) and controlled surface flaw (CSF) methods.

Experimental procedure

Preparation of materials

3 mol% Y$_2$O$_3$ stabilized ZrO$_2$ (> 99.7%, 90 nm, Tosoh, Japan) and Al$_2$O$_3$ (99.99%, 0.33 µm, Sumitomo, Japan) were used as the starting materials. Different amounts of Al$_2$O$_3$ ranging from 0 to 30 vol% were added to the ZrO$_2$ powders. The mixed powders were ball-milled for 24 h in an ethanol medium using a plastic jar and ZrO$_2$ balls. The mixed powders were dried on a hot plate and stirred to avoid gravity-induced segregation. After drying and sieving, the mixed powders were pressed uniaxially at 100 MPa, and cold isostatically
pressed at 150 MPa. The green bodies obtained were sintered at 1550 °C for 2 h in air. After grinding (220 grit diamond wheel) and polishing (1 µm diamond paste), rectangular specimens with dimensions of 3 × 4 × 40 mm were obtained. After polishing, some specimens were annealed at 1200 °C for 1 h in air to estimate the residual stress generated during polishing.

Measurement of fracture toughness

The SEVNB method was carried out using both as-polished and annealed specimens. The surface of the specimens (3 × 40 mm in size) were pre-notched using a diamond cutting wheel (thickness: approximately 250 µm). The depths of the notch were approximately 0.5 mm according to the ISO standard [12]. Fig. 1 shows the apparatus for forming a groove using a resin bonded diamond cutting wheel. The apparatus was designed specially to introduce sharp V-notches, as shown in Fig. 2(a). The notches were then sharpened with a razor blade by polishing the notch tip with diamond paste (1 µm) placed into the notch. The V-notch root radii were measured by scanning electron microscopy (SEM) of the notch tip of each specimen. Fig. 2(b) shows an example of a V-notch. The sharpened notch root radius of the specimen in the micrograph was approximately 17.5 µm. For a valid $K_{IC}$ measurement, the notch root radii and depths of specimens were kept to less than 20 µm and 0.8 mm, respectively. The specimens with sharpened notches were finally loaded in a four-point bending test jig (outer span: 30 mm, inner span: 10 mm) under a cross head speed of 0.5 mm\textpermin. The equation is described as follows:

$$K_{IC} = \sigma \sqrt{a Y} = \frac{P}{b w} \times \frac{S_1 - S_2}{w} \times \frac{3 \sqrt{\alpha}}{2(1-\alpha)^{1.5}} Y$$

(1)

For four-point bending test, $Y$ is calculated by the following equation:

$$Y = 1.9887 - 1.326 \alpha - \frac{(3.49 - 0.68 \alpha + 1.35 \alpha^2) \alpha (1-\alpha)}{(1+\alpha)^2} \alpha = \frac{a}{w}$$

(2)

where $K_{IC}$ is the fracture toughness, $\sigma$ is the fracture strength, $P$ is the fracture load, $b$ is the specimen thickness, $w$ is the specimen width, $S_1$ and $S_2$ are support spans ($S_1 > S_2$), and $Y$ is the stress intensity shape factor.

For comparison, the fracture toughness was also measured using indentation based methods, such as the IF [13], ISB [14] and CSF [15]. In the IF method, Vickers indentations were introduced to the mirror-like sample surface using a hardness tester (Akashi, Model AVK-C0) under a 98 N load with a dwell time of 15 s. The equation proposed by Evans [16] was used to calculate the fracture toughness.

For the ISB method, Vickers indentations were produced at the center of the specimen’s mirror-like surface. The specimens were then fractured using a 3-point bending test. The crosshead speed was 0.5 mm\textpermin. The fracture stress ($\sigma_f$) was calculated using the following equation [17]:

$$\sigma_f = \frac{3WL}{2BD^2}$$

(3)

where $W$ is the breaking load, $L$ is the span, $B$ is the specimen’s width and $D$ is the specimen’s thickness.

The fracture toughness was calculated using the equation proposed by Chantikul et al. [14] as follows:

$$K_{IC} = 0.59 \left( \frac{E}{H} \frac{1}{\alpha} \right)^{0.5} \left( \frac{\sigma_f}{\alpha} \right)^{0.5}$$

(4)

where $E$ is the elastic modulus, $H$ is the hardness and $P$ is the indentation load.

For the CSF method, the specimens were precracked

![Fig. 1. Apparatus to form a groove using a resin bonded diamond cutting wheel (thickness: approximately 250 µm).](image1)

![Fig. 2. Apparatus to introduce sharp V-notch using a razor blade (a) and an example of a sharpened V-notch in a Al₂O₃/3Y-TZP specimen (b).](image2)
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... by introducing a 98 N Vickers indentation at the center of the polished surface. The indented surface was then polished with diamond paste to remove a ~0.06 mm layer to eliminate the effect of residual stress on the fracture toughness caused by the indentation [18]. The specimens were tested using a 4-point bending method. The fracture toughness was calculated using the equations reported by Govila as follows [19]:

$$K_I = \sigma M (\pi a/Q)^{1/2}$$  \hspace{1cm} (5)

where $\sigma$ is the fracture stress, $a$ is the flaw depth, and $M$ and $Q$ are numerical factors related to the flaw and specimen geometry, respectively. For semicircular flaws, $M = 1.03$. $Q$ was obtained using the following equation:

$$Q = \Phi - 0.212(\sigma/\sigma_y)^2$$  \hspace{1cm} (6)

$$Q \approx \Phi^2$$

where $\sigma_y$ is the yield stress, $0.212(\sigma/\sigma_y)^2$ is a plastic-zone correction factor (this factor is negligible for brittle failure in ceramics), and $\Phi$ is an elliptic integral of the second kind:

$$\Phi = \int_0^{\pi/2} \sin^2 \theta + (a/c) \cos^2 \theta)^{1/2} d\theta$$  \hspace{1cm} (7)

where $2a$ is the depth of a semielliptical crack and $2c$ is the total crack length at the free surface.

**Results and discussion**

Fig. 3 presents the fracture toughness of the as-polished and annealed specimens as a function of the Al$_2$O$_3$ content. The fracture toughness decreased with increasing Al$_2$O$_3$ content. The fracture toughness of the polished specimens was higher than those of the annealed specimens due to the residual compressive stresses introduced during polishing. This decreasing tendency could be attributed to the increased critical transformation stress ($\sigma_t$), the decreased volume fraction of transformable t-ZrO$_2$ and the increased residual tensile stress [20].

Table 1 lists the critical transformation stress, fracture toughness increment (AK), transformed zone depth (h) and volume fraction ($V_m$) of the monoclinic phase. The critical transformation stress increased with increasing Al$_2$O$_3$ content, suggesting that extra stress should be applied to induce a t→m transformation with increasing Al$_2$O$_3$ content, which resulted in a decreased transformation zone and fracture toughness. The depth of the transformed zone decreased with increasing Al$_2$O$_3$ content, suggesting that less t→m transformation occurred with increasing Al$_2$O$_3$ addition: the decreasing t→m transformation would result in a decrease in fracture toughness [20].

The thermal residual stresses developed in the composite ceramics play an important role in the fracture toughness of composites. Thermal residual stresses can be introduced due to thermal expansion mismatch between ZrO$_2$ (10.3 × 10$^{-6}$/°C) and Al$_2$O$_3$ (8.1 × 10$^{-6}$/°C) [21]. A radial compressive stress and hoop tensile stress were developed in the 3Y-TZP/Al$_2$O$_3$ composite. The hoop tensile stress increased with increasing Al$_2$O$_3$ content. Crack deflection could not be expected under such a tensile residual stress circumstance. Therefore, a residual tensile stress may be another reason for the decreased fracture toughness [20].

For the CSF method, the fracture surfaces were examined by SEM to observe the semi-elliptical median cracks of the composites, as shown in Fig. 4. The lengths (2a and 2c in Eq. 5) of the semi-elliptical surface cracks in the 3Y-TZP/Al$_2$O$_3$ composites were measured to calculate the fracture toughness.

Fig. 5 shows the fracture toughness as a function of the Al$_2$O$_3$ content tested using the CSF and ISB methods. The fracture toughness measured using the CSF and ISB methods showed a decreased tendency with increasing Al$_2$O$_3$ content, even though the decreasing fracture toughness was not as pronounced as observed in the ISB method.

![Fig. 3. Fracture toughness (SEVNB method) of the 3Y-TZP/Al$_2$O$_3$ composites.](image)

**Table 1.** Critical transformation stress ($\sigma_t$), fracture toughness increment (AK), transformed zone depth (h) and volume fraction of each composite.

<table>
<thead>
<tr>
<th>Samples</th>
<th>$V_m$ (%)</th>
<th>h (µm)</th>
<th>AK (MPam$^{1/2}$)</th>
<th>$\sigma_t$ (GPa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>3Y-TZP</td>
<td>22.11</td>
<td>3.89</td>
<td>1.21</td>
<td>1.271</td>
</tr>
<tr>
<td>3Y-TZP/10A</td>
<td>20.97</td>
<td>3.66</td>
<td>1.16</td>
<td>1.051</td>
</tr>
<tr>
<td>3Y-TZP/20A</td>
<td>12.86</td>
<td>2.12</td>
<td>0.57</td>
<td>1.437</td>
</tr>
<tr>
<td>3Y-TZP/30A</td>
<td>8.04</td>
<td>1.28</td>
<td>0.29</td>
<td>1.451</td>
</tr>
</tbody>
</table>
Fig. 6 presents the fracture toughness of 3Y-TZP/Al₂O₃ measured using the IF method. The test was carried out at 98 N. In general, ceramics are finished by conventional grinding followed by diamond polishing or lapping. Conventional polishing employs high loads and diamond abrasives, which can cause a residual stress on the surface of the ceramics. The effect of residual stress on the fracture toughness obtained by the IF method was assessed by testing both polished and annealed specimens as well as ground and polished specimens. The fracture toughness of the polished and annealed specimens increased slightly with increasing Al₂O₃ content, whereas the fracture toughness of the ground and polished specimens appears to be sensitive to Al₂O₃ addition. The value increased dramatically when the Al₂O₃ content was 10 vol% and reached saturation at 6.9 MPam¹/². The specimens without heat treatment showed higher fracture toughnsses that the annealed ones. To explain the increase in fracture toughness, the residual compressive stress generated during grinding and polishing were estimated using the following equation [22]:

\[
\sigma_r = \frac{K_p - K_a}{2(c/\pi)^{1/2}}
\]

where \(\sigma_r\) is the residual stress (MPa), \(K_p\) is the fracture toughness (IF) of the ground and polished sample, \(K_a\) is the fracture toughness (IF) of the annealed sample and \(c\) is the crack length (µm). Fig. 7 shows the estimated residual compressive stresses generated during the grinding and polishing procedures in the 3Y-TZP/Al₂O₃ composites. The residual stress was also sensitive to the addition of Al₂O₃ and presented a dramatic jump at 10 vol%. This result is consistent with the fracture toughness of the polished and annealed specimens. The results also suggest that the residual stress generated during polishing has a significant effect on the measured fracture toughness.

The effects of the indentation load on the fracture toughness (IF method) in the 3Y-TZP/Al₂O₃ composites were also investigated (Fig. 8). The fracture toughnesses were measured using ground and polished specimens. For all the composites, the fracture toughness decreased with increasing load. The 3Y-TZP/Al₂O₃ composites...
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showed higher values than the 3Y-TZP ceramics until 196 N. However, there was no appreciable difference in fracture toughness at 294 N between 3Y-TZP/Al₂O₃ composites and 3Y-TZP ceramics. Based on this study, the IF method is unsuitable for evaluating the fracture toughness of 3Y-TZP/Al₂O₃ composites. Overall, the SEVNB method is believed to be the most reliable method for determining the fracture toughness of ceramic materials.

Conclusions

3Y-TZP/Al₂O₃ composites containing 0 to 30 vol% of Al₂O₃ were fabricated by pressureless sintering, and the fracture toughness was evaluated using a range of methods. The fracture toughness measured by the SEVNB method decreased with increasing Al₂O₃ content, which was explained by the increasing critical transformation stress, decreasing volume fraction of transformable t-ZrO₂ and increasing residual tensile stress. The fracture toughness tested using the CSF and ISB methods showed a similar tendency to that observed using the SEVNB method. The IF method was found to be unsuitable for measuring the fracture toughness of 3Y-TZP/Al₂O₃ composites. Overall, the SEVNB method is considered the most reliable method for determining the fracture toughness of 3Y-TZP/Al₂O₃ ceramic composites.

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References