Toughening enhanced at elevated temperatures in an alumina/zirconia dual-phase matrix composite reinforced with silicon carbide whiskers

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Abstract

A unique temperature dependence of toughening is observed in an alumina/zirconia dual-phase matrix composite reinforced with silicon carbide whiskers. The work of fracture (WOF) of the composite is maximized at $400\,^{\circ}\text{C}$ to $130\,\text{J/m}^2$, which is about 6.5 times larger than that of monolithic alumina at room temperature. The WOF decreases sharply with an increase in temperature above $400\,^{\circ}\text{C}$. The enhanced toughening at elevated temperatures is described by the stress-induced transformation toughening of tetragonal zirconia, which is affected by the internal thermal stress owing to thermoelastic mismatch between the matrix and the whiskers. The maximum WOF is not given only by the stress-induced transformation but also by the crack-face bridging of the whiskers. The WOF was optimized at a specific zirconia volume fraction of 0.7 in the matrix, which was essentially due to the maximized tensile internal stress on zirconia in the dual-phase matrix.
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1. Introduction

A dual-phase composite consisting of alpha-alumina (α-Al_2O_3) and tetragonal zirconia (t-ZrO_2) has extremely high flexural strength over 1 GPa and is quite tough at room temperature.[1–10] The combination of α-Al_2O_3 and t-ZrO_2 is referred to as AZ hereafter. One application of an AZ composite is utilization in an artificial human hip joint due to its high resistance to fracture. The superior mechanical property of the composite is principally attributed to the stress-induced martensitic transformation from metastable t-ZrO_2 into a stable monoclinic phase (m-ZrO_2). The properties, however, can thermally degrade because the stress-induced transformation is ineffective at elevated temperatures.[3,5,11] This thermal degradation prohibits the AZ composite from structural application at elevated temperatures.

Fiber or whisker reinforcing can compensate for the thermal degradation due to duplex toughening caused by the incorporation of the crack-face bridging of fibrous reinforcements combined with the stress-induced transformation. Furthermore, toughening due to bridging is often enhanced with an increase in temperature.[12] Earlier studies on AZ matrix composites reinforced with silicon carbide whiskers[13–29] (denoted as SiC_w) have shown that the whisker reinforcement actually mitigates the thermal degradation of flexural strength.[13,19,29] The temperature dependence of the fracture toughness of the composite has not yet been examined, which is problematic because we need to know if the combination of the stress-induced transformation of t-ZrO_2 and the crack-face bridging of SiC_w is effective in terms of suppressing the thermal degradation of fracture resistance.

Thus far, the effect of the rising R-curve behavior due to the stress-induced transformation[6,8,30] as well as the bridging[31,32] on fracture toughness evaluations have ignored. In order to properly examine the duplex toughening, the fracture resistance of the composite should be measured by including the R-curve effect, which is generally difficult when working with ceramics at elevated temperatures. Work of fracture (WOF) is a measure of fracture resistance involving rising R-curve behavior.[33]

In this study, we successively measured the WOF and flexural strength of an AZ-matrix composite reinforced with SiC_w from ambient to elevated temperatures. The three-phase composite is denoted as SAZ in this paper. The phase transformation

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from *t*- into *m*-ZrO$_2$ was examined by laser Raman spectroscopy. The synergetic effect between stress-induced transformation and whisker doping on toughening is discussed through a comparison of WOF between SAZ and AZ dual-phase composites.

2. Experimental procedure

We created SAZ by hot-pressing powders in which Al$_2$O$_3$ (TM-100; Taimei Chemicals Co. Ltd., Japan) and Y$_2$O$_3$ (3 mol%)-doped ZrO$_2$ (YZ-3Y; Tosoh Co. Ltd., Japan) were mixed together and doped further with SiC$_w$ (TWS400; Tokai Carbon Co. Ltd., Japan) of 20 vol.%. The powders and whiskers were mixed in *n*-butyl alcohol with a tumbling mixer followed by quick drying with a rotary evaporator. The ZrO$_2$ volume fraction in the AZ-matrix, f_Z, was changed from 0 to 1. Hereafter, SAZs with an f_Z-value of 0 and 1 are denoted as SA and SZ, respectively. The hot-pressing was carried out with graphite dies at 1750 °C under a uniaxial pressure of 33 MPa for 1 h in an argon gas flowing atmosphere. The apparent density of all specimens, which was determined by the Archimedean method using *n*-butyl alcohol, was confirmed to be more than 99% of the theoretical density. The flexural strength of a series of the composite was measured by the four-point bending of a specimen with a dimension of 3 mm × 4 mm × 36 mm from room temperature to 1200 °C in an argon gas flowing atmosphere. The surface of the specimens for the strength measurement was finished by polishing with a paste containing abrasive grains made of a 3-μm-diameter diamond. The WOF of the composite was measured by the three-point bending of a chevron notched specimen with the same dimension in the same temperature range and atmosphere. The width of the main crack in a WOF specimen was less than 3 mm. The number of specimens for the strength and WOF measurements was four or five each for composition and temperature. The composite containing SiC$_w$ randomly distributed on horizontal plane was subjected to hot-pressing in the vertical direction, i.e., in the direction at a right angle to SiC$_w$.[34,35] We selected such orientation of specimens for the fracture tests to make crack propagation perpendicular to the orientation of SiC$_w$. The details of our fracture tests are described elsewhere.[11] Thermal expansion of the composite was measured in an Ar gas flow atmosphere with a dilatometer (DL-7000; ULVAC-RIKO Inc.). The spectroscopy of anti-Stokes Raman scattering with an ultraviolet laser (Model 2062-7S, TEM00; Spectra Physics Inc.) was carried out on a surface fractured at elevated temperatures in order to confirm the phase transformation from *t*- into *m*-ZrO$_2$. Surfaces fractured at WOF measurement were observed with SEM (SP4500, Hitachi High Technology Co., Ltd.).

3. Results and discussion

3.1. Effect of ZrO$_2$ volume fraction in AZ matrix on the mechanical properties of SAZ at room temperature

The WOF and flexural strength of SAZ at room temperature were measured as a function of f_Z, as shown in Fig. 1. The WOF
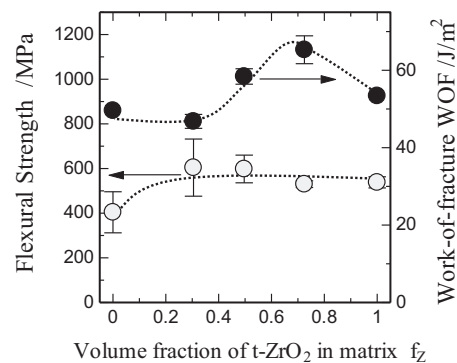

Fig. 1. Work of fracture (WOF) (closed circles) and flexural strength (open circles) of SAZ as a function of *t*-ZrO$_2$ volume fraction in the matrix f_Z.

was optimized at a specific f_Z value of about 0.7. The composite with the optimized WOF is referred as S3A7Z hereafter.

It is important to examine the grain size of ZrO$_2$ because the stability of the tetragonal phase strongly depends on the size.[3] In the case of the AZ composite,[11] the grain size is in the submicron range regardless of f_Z. In the case of the AZ matrix of the SAZ, the grain size of the matrix is also in the submicron range regardless of f_Z according to the observation of the fractured surface (Fig. 2), although the sintering temperature of the SAZ is much higher than that of the AZ. The rather small difference in the grain size between the AZ composite and the AZ matrix is attributed to the suppression of grain growth by SiC$_w$ through the pinning of the grain boundary. The submicron size of ZrO$_2$ grains with 3 mol% Y$_2$O$_3$ indicates the stable existence of *t*-ZrO$_2$ at room temperature.[3] This is why peaks assigned to the *m*-ZrO$_2$ are not found in the Raman spectra of the AZ composite.[11] However, peaks assigned to the *m*-ZrO$_2$ are observed in the Raman spectra of the SAZ on a polished surface (see Figs. 3 and 4), although the grain size of the AZ matrix is in the submicron range. The phase transformation from *t*- to *m*-ZrO$_2$ is attributed to tensile residual thermal stress owing to thermoelastic mismatch between the AZ matrix and the SiC$_w$, which is discussed in detail in the following paragraph.

The optimization in WOF is very similar to that of an AZ dual-phase composite,[11] although the residual tensile stress on *t*-ZrO$_2$ in SAZ is much larger than that in the AZ composite due to the significant thermoelastic mismatch between the AZ-matrix and SiC$_w$. The tensile stress on *t*-ZrO$_2$ in SAZ is so large that transformation from *t*- into *m*-ZrO$_2$ spontaneously progresses during the cooling process of the hot-pressing without any assistance from external stresses. The occurrence of spontaneous *t* → *m* transformation in SZ and S3A7Z is confirmed by Raman spectra assigned to *m*-ZrO$_2$ on a polished surface (see Figs. 3 and 4). No such spontaneous transformation was detected in the AZ composite.[11] The large residual tensile stress on *t*-ZrO$_2$ in SAZ is counteracted to some degree at room temperature due to the volume expansion of ZrO$_2$-grains caused by the spontaneous transformation. The average residual tensile stress on the matrix of SAZ due to the thermoelastic mismatch between AZ-matrix and SiC$_w$ is roughly calculated on the basis of micromechanics with an assumption of thermoelastic and geometrical isotropy[11]