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Simulation Design for the Composition of Zirconia Composite Ceramic Tool

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The relationship between the fracture toughness increment (ΔK_{IC}) resulting from toughening mechanisms, such as phase transition, residual stress, geometry effect, and grain bridging, and the volume fraction of zirconia was established to simulate and design the composition of a zirconia-matrix composite tool, thereby avoiding "trial-and-error" experiments. The composition of the ZrO_2/Al_2O_3 ceramic tool was simulated in accordance with the requirement for fracture toughness. It was shown that the simulated result was in agreement with experiment and that the established simulation model was to some extent valid in predicting the composition of the zirconia-matrix composite ceramic tool with dispersed α-Al_2O_3. Thus, a new type of ceramic tool material, a ZrO_2/Al_2O_3 composite, was developed by adding α-Al_2O_3 to ZrO_2 on the basis of the results of the computer simulation.

Keywords ceramics, composites, mechanical properties, phase transitions, thermal expansion

1. Introduction

New materials, especially ceramics, are usually developed through a "trial-and-error" approach, which wastes time and labor. With the advancement in computer technology and material science knowledge, material design can be carried out based on the known properties of and fabrication experience with different materials with the aid of computer modeling and simulation.[1] The compositions of the materials to be developed may be designed and simulated in accordance with the requirement for the physical or mechanical properties. The development process of a new material can be carried out based on the simulated information of the material composition. Many researchers have investigated the problem of material simulation and have achieved some success, but, as the complexity of the microstructure-mechanical property relationship for ceramic materials increases, research on the ceramic material simulation continues to evolve past the exploratory stage.[2] In addition, some existing simulation models are available only for certain practical applications, and the error associated with the calculations is a little high. Therefore, it is necessary to study the fundamental problem of ceramic material simulation and optimization by examining the composition design of a zirconia (ZrO_2) matrix ceramic with the addition of alumina (Al_2O_3), for example.

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2. Simulation Model for Designing Composition of ZrO_2/Al_2O_3

2.1 Modeling Purpose

To improve the comprehensive mechanical properties and the fracture toughness (K_{IC}) of the ceramic tool material 3Y-PSZ (i.e., 3 mol% Y_2O_3 partially stabilized ZrO_2), a new ceramic composite tool material, ZrO_2/Al_2O_3, will be developed by adding α-Al_2O_3 to the 3Y-PSZ matrix. The relationship between the volume fraction of Al_2O_3 in the composite and the K_{IC} of the ZrO_2/Al_2O_3 was investigated to study the effect that the Al_2O_3 volume fraction had on the K_{IC} increment (ΔK_{IC}). The approach used simulation modeling as a way to determine the Al_2O_3 fraction and to avoid trial-and-error experiments. To accomplish this, a function relating the ΔK_{IC} due to the phase transition of ZrO_2 and the addition of Al_2O_3 to ZrO_2 was developed.

2.2 Relationship Between ΔK_{IC} and Fraction of Zirconia

The assumptions for building the model are as follows: the alumina grain is approximately spherical and uniform in size; the alumina is dispersed uniformly in the 3Y-PSZ powder (i.e., the distance is the same between any of the two alumina grains); the yttria (Y_2O_3) volume fraction in 3Y-PSZ is small and may be ignored; and the ΔK_{IC} caused by various reinforcing mechanisms may be linearly superposed.

Research has shown that the reinforcing mechanisms in 3Y-PSZ with alumina additions are phase transition toughening by ZrO_2, and grain dispersion toughening through crack deflection and grain bridging.[3-6] Thus, the ΔK_{IC} of ZrO_2/Al_2O_3 compared with 3Y-PSZ without alumina (Al_2O_3) can be expressed as follows:

$$\Delta K_{IC} = \Delta K_{ICPT} + \Delta K_{ICDR} = \Delta K_{ICPT} + (\Delta K_{ICCD} + \Delta K_{ICGB}) \quad \text{(Eq 1)}$$

where ΔK_{ICPT} is the fracture toughness increment caused by the phase transition toughening of ZrO_2 when combined with Al_2O_3, ΔK_{ICDR} is the fracture toughness increment caused by dispersion toughening of the Al_2O_3, ΔK_{ICCD} is the fracture toughness increment caused by crack deflection, and ΔK_{ICGB} is the fracture toughness increment caused by grain bridging. From Eq 1, the relationship between ΔK_{IC} and the fraction (f) of zirconia may be evaluated if ΔK_{ICPT}, ΔK_{ICCD}, and ΔK_{ICGB} are known.

2.2.1 Phase Transition Toughening. The reinforcing effect attributed to ZrO_2 is caused by the transition between the different crystal structures of zirconia. The main phase transition mechanism is that the tetragonal phase (t-ZrO_2) of transforms to the monoclinic phase (m-ZrO_2) under an applied stress. The phase transition process can absorb fracture energy and can reduce the propagating power of a micro-crack, resulting in an overall increase in ceramic toughness. On the other hand, in the phase transition process, the volume expansion of 3~5% and the tangential strain of 16% in the ZrO_2 may actually prohibit a crack from propagating, and also may increase the K_{IC}. The ΔK_{IC} caused by the phase transition toughening of ZrO_2 may be expressed through the following equation[7]:

$$\Delta K_{IC}{}^{t-m} = kEV\dot{e}_T h^{1/2}(1-\nu)^{-1} \quad \text{(Eq 2)}$$

where $\Delta K_{IC}{}^{t-m}$ is the ΔK_{IC} caused by phase transition toughening of (ZrO_2MPa·$m^{1/2}$), k is a constant related to the crack propagation situation [i.e., when the crack is in the sub-stable situation (k = 0.22) and is approaching a stable situation (k = 0.36)], E is the elastic modulus (in megapascals), V is the volume fraction of tetragonal ZrO_2 that actually transforms into monoclinic ZrO_2, e_T is the expansion strain of phase transition without restrain, h is the height of the phase transition zone (in millimeters), and ν is Poisson's ratio.

It has been shown that a dispersed reinforcing phase such as Al_2O_3 may increase the fraction of tetragonal ZrO_2 in 3Y-PSZ, but the fraction (V) of tetragonal ZrO_2, which can be easily transformed, will decrease.[8] If we assume that all of the physical properties of the material do not change, then,

$$\Delta K_{ICPT} = kE_c V_c e_T h^{1/2}(1-\nu)^{-1} - kE_m V_m e_T h^{1/2}(1-\nu)^{-1} \quad \text{(Eq 3)}$$

where the suffix m and c represent, respectively, the 3Y-PSZ and the composite ceramic ZrO_2/Al_2O_3.

The elastic modulus of the composite ceramic can be attained with the following relationship[9]:

$$E_c = E_m f + E_p(1-f) \quad \text{(Eq 4)}$$

$$V_c = V_m f \quad \text{(Eq 5)}$$

where the subscript p represents the dispersed phase in the composite ceramic, and f is the volumetric fraction of the ZrO_2.

Substituting Eq 4 and 5 into Eq 3 yields

$$\begin{aligned}\Delta K_{ICPT} &= kV_m e_T h^{1/2}(1-\nu)^{-1}(E_c f - E_m)\\ &= kV_m e_T h^{1/2}(1-\nu)^{-1}[(E_m - E_p)f^2 + E_p f - E_m] \end{aligned} \quad \text{(Eq 6)}$$

When the elastic modulus of Al_2O_3 is higher than that of ZrO_2 ($E_{Al2O3} \approx 390$ GPa; $E_{ZrO2} \approx 220$ GPa), then $E_m < E_p$, and $f < 1$. In this case, the ΔK_{ICPT} in Eq 6 is less than zero and verifies the phase transition-toughening effect for 3Y-PSZ with dispersed Al_2O_3, compared with that of 3Y-PSZ for the same fabrication conditions.

Previous research has revealed that there is no phase transition-toughening effect in 6 mol% Y_2O_3-stabilized ZrO_2 (6Y-ZrO_2), as the stabilizer (Y_2O_3) fraction is great, and all the ZrO_2 is stabilized and cannot transform.[10] The fracture toughness of 3Y-PSZ is 2 MPa√m higher than that of 6Y-ZrO_2 for the same fabrication conditions. This result is attributed to the phase transition toughening from the tetragonal ZrO_2. For 3Y-PSZ, $\Delta K_{IC}{}^{t-m}$ = 2 MPa√m, that is,

$$kE_m V_m e_T h^{1/2}(1-\nu)^{-1} = 2 \quad \text{(Eq 7)}$$

Substituting Eq 7 into Eq 6 leads to the following result,

$$\Delta K_{ICPT} = 2\left[\left(1 - \frac{E_p}{E_m}\right)f^2 + \frac{E_p}{E_m}f - 1\right] \quad \text{(Eq 8)}$$

2.2.2 Crack Deflection Toughening. Toughening mechanisms that are the result of a dispersed second phase can include some or all of the following factors: residual stress; micro-crack and crack deflection; crack bending and branching; crack bridging; and sticking toughening.[11] Faber and Evans[5] determined that the toughening mechanism caused by second-phase particles in a ceramic matrix composite was mainly due to crack deflection, although crack bridging, crack bowing, micro-cracking, or crack branching are also possibilities.

In accordance with the research results by Taya et al.,[12] the ΔK_{IC} due to crack deflection includes an increment from the residual stress resulting from the thermal expansion coefficient mismatch and an increment due to a geometry effect, resulting from the interaction of the crack with the dispersed phase. Thus, the fracture toughness increment due to the crack deflection may be expressed in the following way:

$$\Delta K_{ICCD} = \Delta K_{ICRS} + \Delta K_{ICGE} \quad \text{(Eq 9)}$$

where ΔK_{ICRS} is the fracture toughness increment caused by residual stress toughening from the Al2O3, while ΔK_{ICGE} is the fracture toughness increment caused by the geometry effect.

The residual stress in the composite ceramic is caused by the mismatch of the thermal expansion coefficient for the matrix and dispersed material when the composite cools down from the fabricating temperature to room temperature (RT). It is difficult to measure or calculate the residual stress due to its complicated distribution. Generally, it is evaluated using the model proposed by Taya et al.[12] The residual stress distribution in the composite ZrO_2/Al_2O_3 is shown in Fig.1, in which the thermal expansion coefficient of 3Y-PSZ is greater than that of the Al_2O_3. The residual strain in the grain may be expressed as follows:

$$\alpha^* = \int_{T_P}^{T_R} (\alpha_p - \alpha_m)\delta \, dT \quad \text{(Eq 10)}$$

where α^* is the residual strain in the grain, T_P is the fabrication temperature of the ZrO_2/Al_2O_3 composite, T_R is the RT, α_m and

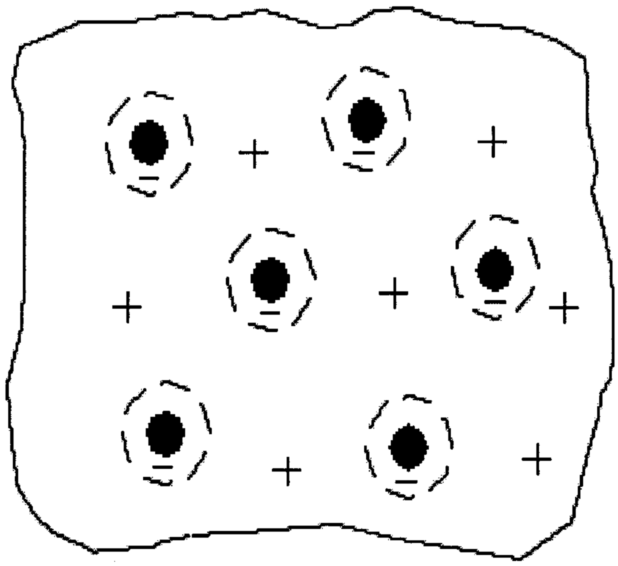

Fig. 1 The residual stress distribution in the ZrO_2/Al_2O_3 composite. The solid black point denotes the Al_2O_3 dispersed grain; the dashed circle denotes the stress field of the grain; + denotes the tensile stress; and − denotes the compressive stress.

α_p are the thermal expansion coefficients of 3Y-PSZ and Al_2O_3, respectively (i.e., $\alpha_m \approx 10.4 \times 10^{-6}$ [spaceband] K^{-1} and $\alpha_p \approx 7.9 \times 10^{-6}$ [spaceband] K^{-1}), and δ is the equiaxed tensor.

For simplification, α^* is approximately expressed as follows[13]:

$$\alpha^* = (\alpha_p - \alpha_m)(T_P - T_R) = \Delta\,\alpha\,\Delta T \quad \text{(Eq 11)}$$

Therefore, the mean residual stress in the composite can be calculated as[12]

$$q = \langle\sigma_m\rangle = \frac{2(1-f)\beta\,\alpha^* E_m}{A} \quad \text{(Eq 12)}$$

where $q = \langle\sigma_m\rangle$ is the mean residual stress in the composite. In this equation

$$\beta = \left(\frac{1+\nu_m}{1-2\nu_p}\right)\left(\frac{E_p}{E_m}\right) \quad \text{(Eq 13)}$$

$$A = f(\beta+2)(1+\nu_m) + 3\beta(1-f)(1-\nu_m) \quad \text{(Eq 14)}$$

The stress intensity factor for dispersed grain toughening composite can be calculated with the following equation[5]:

$$K_{IC} = K_{I0} + 2q\sqrt{\frac{2D}{\pi}} \quad \text{(Eq 15)}$$

where K_{I0} is the stress intensity factor of the matrix, and D is the crack propagation length.

Assuming that the crack propagates from one stress concentration zone to another, $D = (\lambda - d)$.[12] So the fracture toughness increment due to residual stress is,

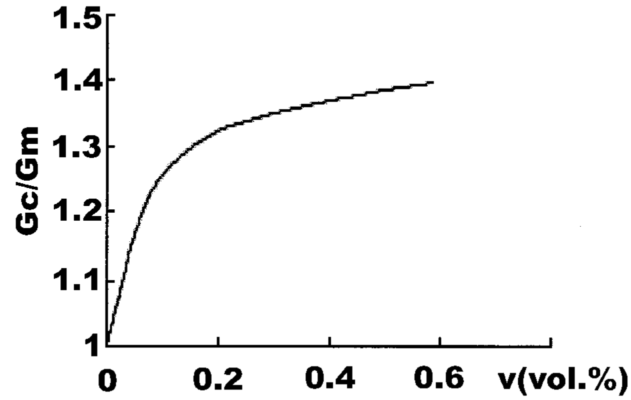

Fig. 2 The ratio of the critical strain energy release rate to the volume fraction of the dispersed phase

$$\Delta K_{IC} = K_{IC} - K_{I0} = 2q\sqrt{\frac{2(\lambda - d)}{\pi}} \quad \text{(Eq 16)}$$

where λ is the mean distance between two adjacent grains, and d is the mean diameter of a typical grain.

Equation 16 can be applied to the case in which the thermal expansion coefficient of the matrix is greater than that of the dispersed phase,[12] and, thus, the fracture toughness increment due to any residual stress for the ZrO_2/Al_2O_3 composite is,

$$\Delta K_{ICRS} = 2q\sqrt{\frac{2(\lambda_p - d_p)}{\pi}} \quad \text{(Eq 17)}$$

where λ_p is the mean distance between two adjacent Al_2O_3 grains (in meters), and d_p is the mean diameter of Al_2O_3 grain (in meters).

As λ_p is related only to the volume fraction of the dispersed phase, and the mean diameter of the dispersed grain, the mean distance between two adjacent Al_2O_3 grains, is[13]

$$\lambda_p = 1.085\, d_p\,(1-f)^{-1/2} \quad \text{(Eq 18)}$$

The ΔK_{ICGE} of Al_2O_3 can be evaluated in accordance with the relationship between the ratio of the critical strain energy release rate of the ZrO_2/Al_2O_3 composite to that of the 3Y-PSZ matrix and the volume fraction of Al_2O_3. This is shown in Fig. 2.[5] In Fig. 2, G_c is the critical strain energy release rate of the ZrO_2/Al_2O_3 composite, while G_m is the critical strain energy release rate of the 3Y-PSZ matrix. The (1-f) term is the volume fraction of the dispersed Al_2O_3.

Assuming that $G_c/G_m = F$, the coefficient F can be confirmed from Fig. 2 according to the volume fraction of Al_2O_3. According to fracture mechanics theory,[14] the relationship between the strain energy release rate and fracture toughness is

$$G_c = \frac{K_{IC}^{c2}}{E_c}, \quad G_m = \frac{K_{IC}^{m2}}{E_m} \quad \text{(Eq 19)}$$

where $K_{IC}{}^c$ and $K_{IC}{}^m$ represent, respectively, the fracture toughness of the ZrO_2/Al_2O_3 composite and the 3Y-PSZ matrix.

Substituting Eq 19 into $G_c/G_m = F$,

$$K_{IC}^{c} = K_{IC}^{m}\sqrt{\frac{E_c}{E_m}\cdot F} \quad \text{(Eq 20)}$$

Subsequently, the ΔK_{ICGE} is,

$$\Delta K_{ICGE} = K_{IC}^{c} - K_{IC}^{m} = K_{IC}^{m}\left(\sqrt{\frac{E_c}{m}\cdot F} - 1\right) \quad \text{(Eq 21)}$$

2.2.3 Grain Bridging Toughening. Grain bridging toughening is greatly affected by the grain size of the dispersed phase. The fracture toughness increment due to grain bridging toughening ΔK_{ICGB} was determined to be[6]:

$$\Delta K_{ICGB} = 2.5\,(1-f)\,E_P\,(\alpha_p - \alpha_m)\Delta T\left(\frac{d_p}{2}\right)^{1/2} \quad \text{(Eq 22)}$$

2.2.4 Relationship Between Fracture Toughness and Volume Fraction of ZrO_2. The relationship between the fracture toughness of the ZrO_2/Al_2O_3 composite and the fraction of ZrO_2 is

$$K_{IC} = K_{IC}^{m} + \Delta K_{IC} \quad \text{(Eq 23)}$$

where $K_{IC}{}^{m}$ is the fracture toughness of the 3Y-PSZ matrix and may be measured by experiment.

$$\Delta K_{IC} = \Delta K_{ICPT} + \Delta K_{ICCD} + \Delta K_{ICGB}$$

$$\Delta K_{ICPT} = 2\left[\left(1 - \frac{E_p}{E_m}\right)f^2 + \frac{E_p}{E_m}f - 1\right]$$

$$\Delta K_{ICCD} = 2q\sqrt{\frac{2(\lambda_d - d_p)}{\pi}} + K_{IC}^{m}\left(\sqrt{\frac{E_c}{E_m}\cdot F} - 1\right)\Delta K_{ICGB}$$
$$= 2.5\,(1-f)\,E_P\,(\alpha_p - \alpha_m)\Delta T\left(\frac{d_p}{2}\right)^{1/2}$$

Thus, the volume fraction of 3Y-PSZ can be expressed as the function of the fracture toughness of the ZrO_2 matrix (50% < f < 100%) composite:

$$f = g(K_{IC}) \quad \text{(Eq 24)}$$

The volumetric fraction of 3Y-PSZ and Al_2O_3 now can be simulated in accordance with the requirement that the K_{IC} of the composite ceramic tool conform to the relationship in Eq (24).

3. Simulation Results

The simulation model was programmed using the C language. The composition of the composite ceramic tool material was simulated in accordance with the requirement for K_{IC}. The physical properties and the fabricating technological parameters are as follows: E_m = 220 GPa; E_p = 390 GPa; α_m = 10.4×10^{-6} [spaceband] K^{-1}, $\alpha_p = 7.9 \times 10^{-6}$ [spaceband] K^{-1}, $\nu_m = 0.31$, $\nu_p = 0.27$, ΔT = 1475 K (as the fabricating and RT are 1500 °C and 20 °C, respectively), d = 0.8 μm, $K_{IC}{}^{m}$ = 5 MPa [spaceband] $m^{1/2}$ (by experiment). The partial simulated results are shown in Table 1.

Table 1 Model Simulation Results for $ZrO_2Al_2O_3$

K_{IC}, MPa · $m^{1/2}$	Fraction of Al_2O_3, vol.%	Fraction of 3Y-PSZ, vol.%
7.6	20	80
7.8	26	74
8.0	28	72
8.2	29	71
8.4	31	69

It was found from Table 1 that the fraction of Al_2O_3 in the composite should be about 28 vol.% (or 22 wt.%), if the required fracture toughness is to be 8.0 MPa$\sqrt{m}$. This simulation result also was verified with practical experiments. It was seen from the experiments that a mean ± SD experimental fracture toughness of 8.0 ± 0.7 MPa$\sqrt{m}$ can be obtained if the weight fraction of Al_2O_3 is about 20%. It was shown that the simulation model result was in good agreement with the experimental one, and the established simulation model was to some extent valid in determining the composition of the ZrO_2 matrix composite with dispersed Al_2O_3.

4. Conclusions

The relationship between the ΔK_{IC} and the volume fraction of ZrO_2 was established through model simulation by adjusting the composition of the ceramic composite in accordance with the requirement for desired fracture toughness. It was shown that the simulation model result was in good agreement with the experimental one, and that the model was, to some extent, valid in establishing the composition level in the ZrO_2 matrix composite with Al_2O_3 additions.

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References

1. L. Yunwen and L. Hengde: "Development and Design of New Materials," *Mater. Rev.*, 1993, (3), pp. 1-3 (in Chinese).
2. W. Jiangwei, H. Huaan, and H. Huahui: "Study on Expert System of CAD on Magnetic-Optic Film Material," *Mater. Rev.*, 1996, (5), pp. 11-14 (in Chinese).
3. H. Tsubakino and R. Nozrod: "Effect of Alumina Addition on the Tetragonal-to-Monoclicic Phase Transformation in Zirconia-3 mol% Y_2O_3," *J. Am. Ceram. Soc.*, 1991, *74*(2), pp. 440-43.
4. M. Fukuhara: "Properties of [Y]ZrO_2-Al_2O_3 and [Y]ZrO_2-Al_2O_3-[Ti or Si]C Composites," *J. Am. Ceram. Soc.*, 1989, *72*(2), pp. 236-42.
5. K.T. Faber and A.G. Evans: (a) "Crack Deflection Processes – I. Theory," *Acta Metall.*, 1983, *31*(4), pp. 565-76; and (b) "Crack De-

flection Processes – II. Experiment," *Acta Metall.*, 1983, *31*(4), pp. 577-84.
6. P.F.Becher: "Microstructural Design of Toughened Ceramics," *J. Am. Ceram. Soc.*, 1991, *74*(2), pp. 187-206.
7. K. Yanmei and J. Xinhai: "Reinforcing Mechanisms of Composite Ceramics and Their Effects," *J. Ceram.*, 1998, *19*(4), pp. 23-26 (in Chinese).
8. Z. Zhenjun and G. Changbin: "Study on the Mechanical Properties of ZrO_2-Al_2O_3 Ceramic Roll Materials," *Mech. Eng. Mater.*, 1997, *21*(3), pp. 56-60 (in Chinese).
9. Z. Yu and L. Tingquan: *Ceramic Material Science*, Harbin University of Technology Press, Harbin, 1995 (in Chinese).
10. G. Qilu, G. Zengsen, L. Tingquan, and J. Weiguang: "Study on the Organization of Al_2O_3-ZrO_2 (6mol% Y_2O_3) Composite," *Silicate Rev.*, 1995, *14*(2), p. 8 (in Chinese).
11. Z. Hong and J. Zongzhe: "Study on the Residual Stress and Reinforcing Mechanism of Particle Dispersed Composite Ceramics," *J. Silicate*, 1996, *24*(5), pp. 491-96 (in Chinese).
12. M. Taya, S. Hayashi, A.S. Kobayashi, and H.S. Yoon: "Toughening of a Particulate-Reinforced Ceramic-Matrix Composite by Thermal Residual Stress," *J. Am. Ceram. Soc.*, 1990, *73*(5), pp. 1382-91.
13. J-F. Li and R. Watanabe: "Fracture Toughness of Al_2O_3-Particle-Dispered Y_2O_3-Partially Stabilized Zirconia," *J. Am. Ceram. Soc.*, 1995, *78*(4), pp. 1079-85.
14. *Mechanical Property of Ceramics*, Bao Wang, ed., Shanghai Science and Technology Literature Press, Shanghai, 1981 (in Chinese).