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Alumina ceramics toughened by a piezoelectric secondary phase

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Abstract

A process for ceramic toughening is developed, where a selected piezoelectric or ferroelectric secondary phase is incorporated and dispersed in a ceramic matrix as reinforcement. The toughening effect of $Nd_2Ti_2O_7$ secondary phase on Al_2O_3 ceramic is determined together with the microstructures and phase constitution of the composite ceramics. Dense composite ceramics can be formed with the secondary phase and the Al_2O_3 matrix. Compared to single phase Al_2O_3 ceramics, the $Nd_2Ti_2O_7/Al_2O_3$ composite ceramic shows a significant increase in fracture toughness, and K_{IC} reaches 6.7 MPa $m^{1/2}$ for a composition of 0.03 $Nd_2Ti_2O_7$/0.97Al_2O_3.

Keywords: Al_2O_3; Composites; $Nd_2Ti_2O_7$; Piezoelectric properties; Toughness and toughening

1. Introduction

Low-cost ceramic composite materials that exhibit excellent mechanical properties are attracting considerable interest for advanced engineering applications. For such materials, toughening remains one of the most important issues in enhancing the reliability. Over the past years, considerable advances have been made to improve the fracture toughness of ceramic systems, and a number of toughening methods have been developed. The individual mechanisms include transformation toughing, microcracking, twinning, ductile reinforcement, fiber/whisker reinforcements and grain bridging.[1–14] All these toughening mechanisms are based on the energy dissipation/energy balance approach[1], and the toughness of the composite ceramic can be generally given as

$$K_{IC} = (E^c(J^m + \Delta J))^{1/2} \quad (1)$$

where, K_{IC} is the overall toughness of the composite ceramic, E^c is the Young's modules of the composite ceramic, J^m is the energy change associated with crack extension in the matrix, and the term ΔJ corresponds to the energy consumption due to the presence of the secondary phase which acts as the toughening phase through the different mechanisms, respectively.

Besides the preceding toughening mechanisms, another mechanism called domain switching toughening in ferroelectric–ferroelastic materials has also received much attention. Experimental and theoretical analysis[15–19] indicated that the domain switching plays an important role in the toughness variation of ferroelectrics. Work was done on well-known ferroelectric–ferroelastic materials, such as $BaTiO_3$ and PZT.[18] It was observed that either an applied electrical field or a compressive stress led to domain switching.

Considering the domain switching toughening mechanism and the piezoelectric effect in piezoelectric or ferroelectric materials, the authors[20] proposed and developed a new approach for the toughening of structural ceramics, in which a piezoelectric or ferroelectric secondary phase is incorporated into the matrix ceramic, and the toughening is achieved through energy dissipation due to the piezoelectric effect or/and domain wall motion. That is, a part of the mechanical energy causing crack extension may be transformed into electrical energy or dissipated by domain wall motion in the piezoelectric particles, and this subsequently leads to the enhanced fracture resistance. The fracture toughness of the composite ceramic can be given in an extended form of Eq. (1)

$$K_{IC} = (E^c(J^m + \Delta J + \Delta J^{piezo}))^{1/2} \quad (2)$$

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The term, ΔJ^{piezo}, defines the energy dissipated by the "Piezoelectric energy dissipation mechanism".

To realize the above idea, the densification of such composite ceramics and the stable co-presence of a piezoelectric or ferroelectric secondary phase with the matrix are the primary issues. The previous work has been carried out in the system of $BaTiO_3/Al_2O_3$,[20] where enhanced fracture toughness was achieved with a maximum of 5.1 MPa $m^{1/2}$. However, the undesirable reaction between $BaTiO_3$ and Al_2O_3 was a problem, which limited the toughening effect, and this problem became much more serious for large $BaTiO_3$ concentration. In order to develop a stable system, in which the piezoelectric secondary phase and the matrix are both inert with respect to each other, $Nd_2Ti_2O_7$,[21] a piezoelectric and ferroelectric compound with high Curie temperature, is adopted as the piezoelectric reinforcing phase in Al_2O_3 ceramics. The toughening effect is determined together with the microstructures and phase constitution in the present paper.

2. Experimental procedure

Reagent-grade (99.9% purity) Nd_2O_3 and TiO_2 in 1:2 mole ratio were mixed for 24 h by ball milling in ethanol using zirconia media. The slurry was dried and then calcined at 1425°C for 6 h to create $Nd_2Ti_2O_7$. Then, $xNd_2Ti_2O_7/(1\text{-}x)Al_2O_3$ composite powders ($x=0.02$, 0.03, 0.05, 0.1, 0.15) were mixed by ball milling with zirconia media in ethanol for 24 h. Such mixed powders were pressed into disc compacts of 12 mm in diameter and 2 to 5 mm in height, and these compacts were sintered at 1400 to 1500°C in air for 3 h.

Microstructural development was examined via scanning electron microscopy (SEM). The phase content of the composite ceramics was characterized by X-ray powder diffraction (XRD) analysis using CuK_α radiation. The fracture toughness was evaluated by the modified indention method[22,23] at room temperature using a diamond Vikers indenter with a loading time of 15 s at a constant load of 60 N. The results were averaged over six indentations per specimen, and the following formula was used in the calculations:

$$(3K_{IC}/Ha^{1/2})(H/3E)^{2/5} = 0.129(c/a)^{-3/2} \quad (3)$$

where K_{IC} is the toughness of the composite ceramic, H is the Vickers hardness, E is the elastic modulus, c is the radius of the crack, and a is the half diagonal length of an indentation.

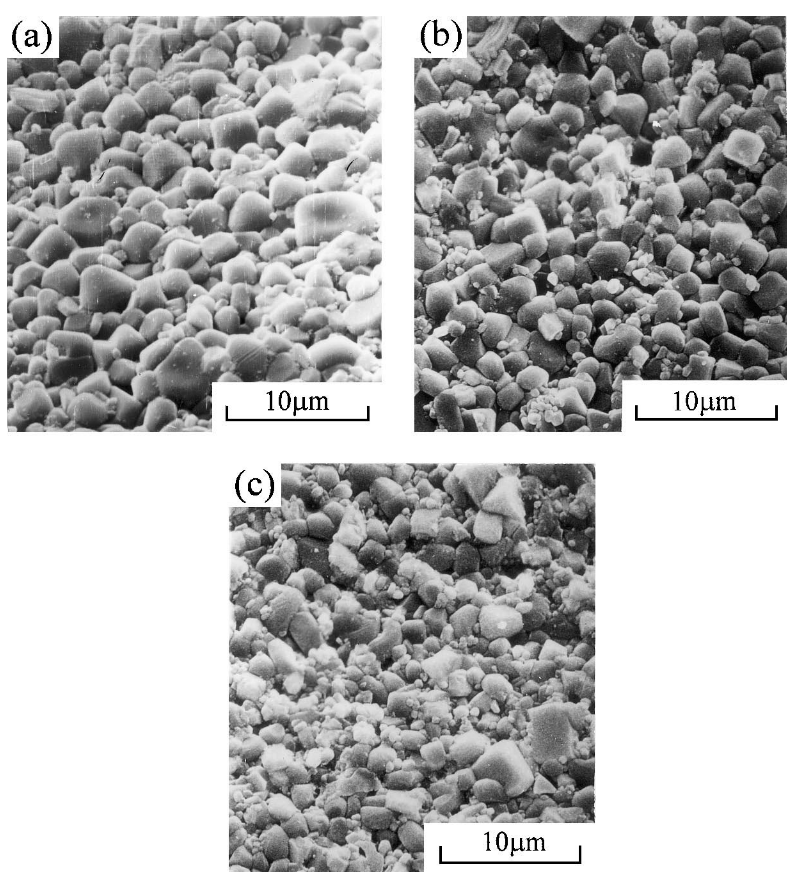

Fig. 1. SEM photomicrographs of $xNd_2Ti_2O_7/(1\text{-}x)Al_2O_3$ ceramics sintered at 1450°C in air for 3h: (a) $x=3$ mol%, (b) $x=5$ mol%, (c) $x=10$ mol%.

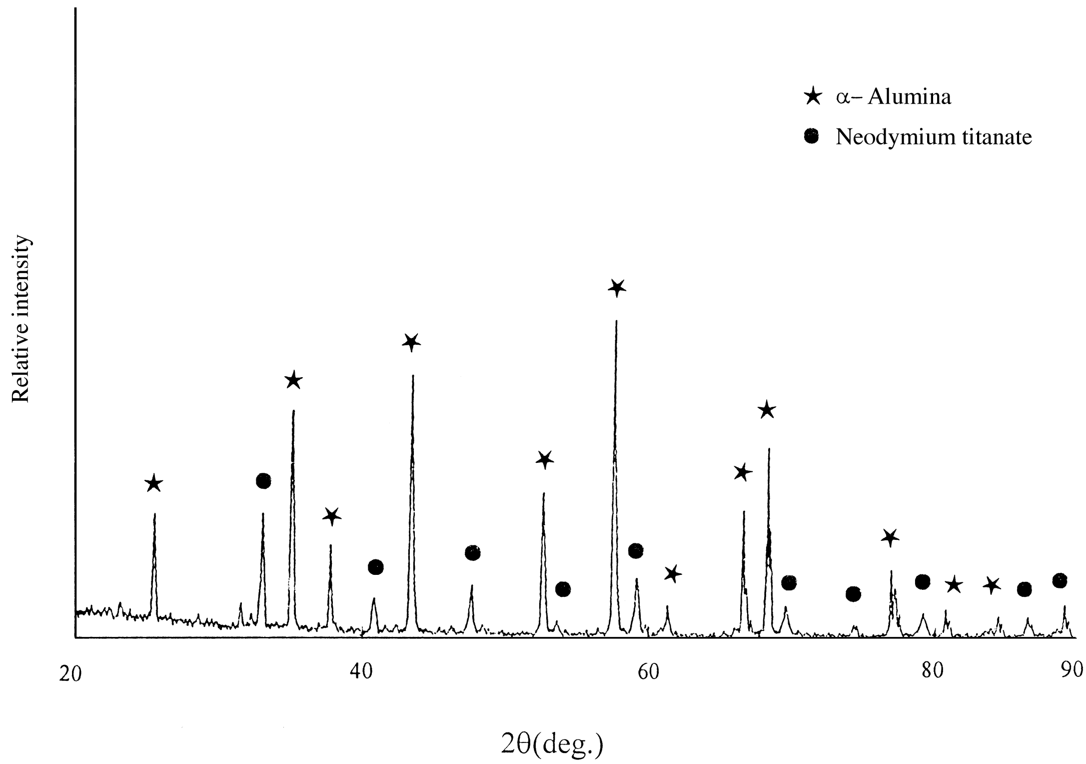

Fig. 2. XRD pattern of $0.03Nd_2Ti_2O_7/0.97Al_2O_3$ ceramics sintered at 1450°C in air for 3 h.

3. Results and discussion

Fig. 1 gives the typical SEM micrographs where homogeneous microstructures with little porosity are observed. In other words, the dense composite ceramics can be created with ease by conventional low-cost pressureless sintering. And there is no significant microstructural difference observed between the composite ceramics with different concentrations of $Nd_2Ti_2O_7$.

To obtain the optimum toughening effect, that is, to maximise the term $\Delta J^{\mathrm{piezo}}$ in Eq. (2), the co-existence of the piezoelectric secondary phase and the matrix is a very important issue in the present approach. As shown in Fig. 2, we can see that the co-presence of neodymium titanate with the alumina major phase is confirmed for $0.03Nd_2Ti_2O_7/0.97Al_2O_3$ ceramic sintered at 1450°C in air for 3 h, and the situations for other compositions in $x$$Nd_2Ti_2O_7/(1\text{-}x)Al_2O_3$ ceramics are similar.

Fracture toughness as a function of composition is given in Fig. 3 for $Nd_2Ti_2O_7/Al_2O_3$ ceramics sintered at 1450°C in air for 3 h. It is obvious that the fracture toughness K_{IC} of Al_2O_3 ceramics is significantly improved by incorporating a modest amount of $Nd_2Ti_2O_7$ piezoelectric secondary phase; the maximum fracture toughness of 6.7 MPa $m^{1/2}$ is achieved for the composition of $0.03Nd_2Ti_2O_7/0.97Al_2O_3$, being almost twice as tough as that of the Al_2O_3 matrix. For the situation of large $Nd_2Ti_2O_7$ concentration, the toughening effect is limited due to the lower Young's modules of the composite ceramics (see Table 1).

The above results indicate that a significant toughening effect results when a suitable piezoelectric secondary

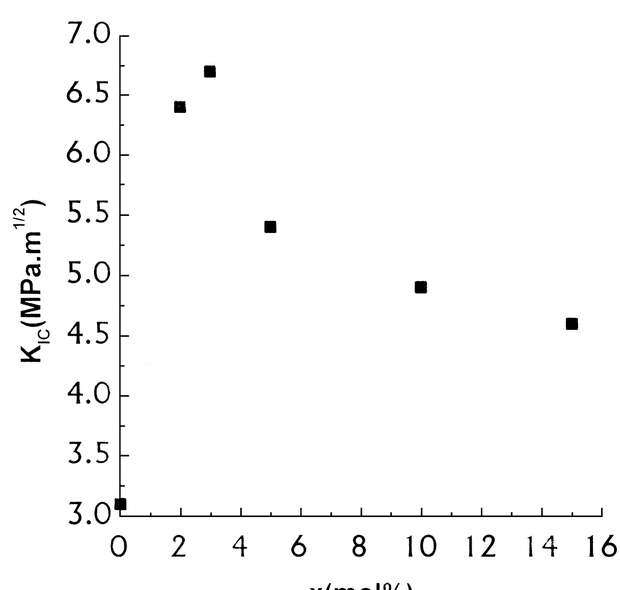

Fig. 3. Fracture toughness of $x$$Nd_2Ti_2O_7/(1\text{-}x)Al_2O_3$ ceramics sintered at 1450°C vs. concentration of $Nd_2Ti_2O_7$.

Table 1
Calculated Young's modulus of $x\mathrm{Nd_2Ti_2O_7}/(1\text{-}x)\mathrm{Al_2O_3}$ composite ceramics

x	0.02	0.03	0.05	0.10	0.15
E_U[a] (GPa)	350.7	346.4	338.2	320.3	305.3
E_L (GPa)	344.6	337.9	325.8	302.3	285.0

[a] $E_U = E_1V_1 + E_2V_2$, $1/E_L = V_1/E_1 + V_2/E_2$, where, E_U is the upper estimation and E_L is the lower estimation of the Young's Modulus for the composite ceramics, E_1 and E_2 the Young's modulus of Al_2O_3 and $Nd_2Ti_2O_7$, respectively ($E_1 = 360$ GPa, $E_2 = 208$ GPa), V_1 and V_2 the fractions of Al_2O_3 and $Nd_2Ti_2O_7$, respectively.

phase is introduced into alumina ceramics, where a piezoelectric energy dissipation mechanism is suggested as providing the toughening.

According to the previous work by Wahi et al.,[24] the fracture toughness for brittle particle toughened ceramics can be expressed by the following equation:

$$K_{IC}^2 \approx 2\{(\Delta E \cdot \gamma_m + \Delta\gamma \cdot E_m)f_p + E_m\gamma_m\} \quad (4)$$

where, K_{IC} is the fracture toughness, ΔE and $\Delta\gamma$ are the difference of Young's modulus and fracture surface energy between particle and the matrix, E_m and γ_m is the Young's modulus and fracture surface energy of the matrix. The necessary condition for brittle particle toughening is $\Delta E > 0$ and $\Delta\gamma > 0$. In the present system, however, this condition is unsatisfied, confirming that the special character of the piezoelectric secondary phase toughening.

4. Conclusions

A piezoelectric, ferroelectric secondary phase has been introduced to a ceramic matrix as the reinforcement, and the dissipation of the mechanical energy causing crack extension by way of the piezoelectric effect is suggested as a new toughening mechanism. High fracture toughness has been achieved in Al_2O_3 ceramics by incorporating $Nd_2Ti_2O_7$ piezoelectric secondary phase, where the co-presence of secondary phase and matrix can be maintained. The highest fracture toughness of 6.7 MPa $m^{1/2}$ is obtained for the composition of $0.03Nd_2Ti_2O_7/0.97Al_2O_3$, and the toughening effect is limited for higher concentrations of such secondary phase because of the lower Young's modulus.

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