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In situ TEM observation of crack propagation in $LiTaO_3$ particle toughened Al_2O_3 ceramics

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Abstract

Direct observation of crack propagation in $LiTaO_3/Al_2O_3$ composite ceramics was carried out using *in situ* transmission electron microscopy (TEM). Domain switching induced by crack propagation, crack deflection and branching at domain boundaries and ripples similar to the contrasts of 180° domains at the microcrack tip inside $LiTaO_3$ grains were detected evidently. Domain switching, crack deflection, branching and energy dissipation resulting from the formation of contrasts similar to the 180° domains at the microcrack tip, were proposed as the toughening mechanisms in $LiTaO_3/Al_2O_3$ ceramics.

Keywords: $LiTaO_3/Al_2O_3$ composite ceramics; Crack propagation; Domain switching; Toughening mechanism; Transmission electron microscopy (TEM)

1. Introduction

For advanced ceramics, a series of toughening approaches has been proposed and developed [1–13]. Recently, a new approach for the toughening of ceramics, in which a piezoelectric secondary phase was incorporated into the matrix ceramic, was proposed [14–23]. For example, $BaTiO_3/Al_2O_3$ [14], $Nd_2Ti_2O_7/Al_2O_3$ [15], $Sr_2Nb_2O_7$/3Y-TZP [16], $LiTaO_3/Al_2O_3$ [17–19], $Nd_2Ti_2O_7$/8Y-FSZ [20], PZT/glass [21] and $BaTiO_3$/MgO [22,23] have been found to exhibit improved mechanical properties. It was suggested that the toughening was achieved through energy dissipation due to the piezoelectric effect or/and domain switching [14–21]. However, toughening mechanisms in ceramics with the piezoelectric second phase have not been experimentally determined overall.

The Vickers indentation technique is commonly used to study the fracture toughness resistance of materials. But, since the domain size of piezoelectric ceramics ranges from nanometers to micrometers [24], and the fracture process zone is limited to a few micrometers for most ceramics, transmission electron microscopy (TEM) would be an ideal technique for observation purposes. *In situ* TEM observation on crack propagation paths is often adopted to investigate the fracture resistance of metal materials, but has seldom been used for that of ceramics due to its brittleness. On the basis of our previous investigations [17–19], in this paper, *in situ* TEM observation was employed to examine crack propagation in Al_2O_3 ceramics with the $LiTaO_3$ piezoelectric second phase. Toughening mechanisms of the composite have been illustrated experimentally.

2. Experimental

Commercially available alumina, Al_2O_3, powder (High Tech Ceramic Institute, Beijing, China) and lithium tantalate, $LiTaO_3$, powder (Dongfang Tantalum Joint-stock Corporation, Ningxia, China) were used as the starting powders. The average particle sizes of Al_2O_3 and $LiTaO_3$ powders were ~0.3 μm and 3.0 μm, respectively. The amount of $LiTaO_3$ was 15 vol%. Al_2O_3 and $LiTaO_3$ were weighed accurately and then ball milled for 24 h with Al_2O_3 balls. Ethanol was used as a medium for the ball milling. The slurry was stirred and dried slowly to remove the ethanol. The sintering was performed by hot pressing at 1300 °C with a graphite die for 90 min under a pressure of 25 MPa in a vacuum atmosphere.

In situ TEM observation was carried out to study the crack propagation paths in the composite. The sintered specimens

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were sliced to 0.5 mm in thickness and polished down to less than 30 μm using 600# SiC abrasives. The thin foil specimens were then cut to 3 mm in width and 5 mm in length and prepared by ion beam thinning at 6 kV, and then examined using TEM (TECNAI 20) at an accelerated voltage of 120 kV and under a slowly increased tensile load [25,26].

3. Results and discussion

In our previous study [18], co-existent $LiTaO_3/Al_2O_3$ (denoted as LTA) ceramic composites with high density of more than 99.5% were prepared by hot pressing at 1300 °C. It was found that the fracture toughness of the composites containing 15 vol% $LiTaO_3$ (denoted as 15LTA) particles was improved over that of monolithic Al_2O_3. That is to say, weak $LiTaO_3$ particles have toughening effects on Al_2O_3 ceramics. An observation on the indentation crack propagation paths showed that the crack penetrated into the $LiTaO_3$ particles and particle debonding or pullout was negligible. The crack did not propagate in an undisturbed manner but produced large deflection through the $LiTaO_3$ particles. SEM observation showed that many steps, strips and layers or river patterns, which were all in the size of the domains, were observed on the fracture surface of the $LiTaO_3$ particles in the Al_2O_3 matrix [27]. It is ascertained that the intrinsic microstructure of $LiTaO_3$ particles should be helpful to counteract the crack growing.

A visible phenomenon was detected during *in situ* TEM observation on the crack propagation in $LiTaO_3$ grains. It was found that several 90° domains similar to daggers were produced on both sides of the crack growing path, as shown in Fig. 1. The crack growing direction is indicated by a white arrow in the figure. These domains were less than 0.1 μm in length. It is well known that domains in ferroelectric ceramics can be switched either by an electric field or by a mechanical stress [28–30]. When the crack propagates, high stress concentration will be caused at the crack tip. When the total stress concentration reaches the threshold for the domain to switch, domain switching will occur. In fact, domain switching is a process of nucleating, growing and amalgamating of new domains. According to the location and the shape of the domains in Fig. 1, they seemed to be domain nuclei of domain switching caused by crack propagation. It was found that these domain nuclei did not grow into bigger domains. For pure ferroelectric ceramics, domain switching induced by an electric field or mechanical stress has been widely investigated both theoretically and experimentally. But, direct observation on domain switching by crack propagation in ceramic composites with a ferroelectric second phase has seldom been reported to date. This study provides direct experimental evidence for 90° domain switching caused by crack growing in Al_2O_3 ceramics with a $LiTaO_3$ second phase.

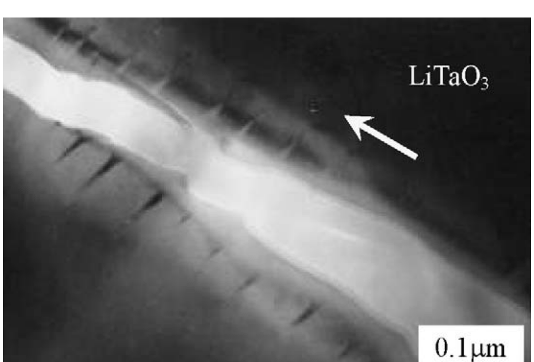

Fig. 1. TEM micrographs of domain nuclei located along a crack due to domain switching caused by crack propagation in $LiTaO_3$ grains.

It was confirmed by *in situ* TEM observation [27] that deflections and branching both occurred at the boundaries of the 90° domains. That is to say, the domain boundary was in favor of crack propagation because of its high energy. In this study, various crack deflections at the domains in the composite were detected, as shown in Fig. 2. Crack deflections of about 90° in Fig. 2(a) produced at the other domain boundary occurred after cutting across the domain. This is different in Fig. 2(b) and (c), where the cracks produced deflections of less than 90° at the former domain boundary and then grew into the domain and continued to extend. When the cracks reached the other domain boundary, they produced another deflection of less than 90° and then developed along the other domain boundary or another domain boundary entirely. Crack branching was also observed in Fig. 2(b). A crack deflection with a large angle of more than 90° occurred in Fig. 2(d). The propagating crack cut across the domain and reached the interface between the $LiTaO_3$ and Al_2O_3 grains. But the crack did not continue to propagate along the interface between them. Instead, it produced a deflection at an angle of more than 90° and then developed along another domain boundary entirely. This result indicated that the domain boundary assisted crack propagation more than the interface between the $LiTaO_3$ and Al_2O_3 grains. It was concluded that the domain structures in the $LiTaO_3$ grains greatly affected the crack development.

Besides crack deflection and branching, several ripples were produced at or near the crack tips [18]. It can be seen from Fig. 3 that mass ripples were induced right before or on one side of the microcrack tip and extended for a long distance, as indicated by the white arrows. It seemed that these ripples had regular intervals and were different from stacking dislocations often observed in the study on metal ductile materials. The stacking dislocations have smaller intervals near the crack tip and larger intervals far from the crack tip. *In situ* TEM observation showed that it was common for the propagating crack in the $LiTaO_3$ grains in the as-hot-pressed LTA ceramic composite to cause ripples ahead of it. From the observation, these ripples were similar to contrasts of the 180° domains. According to the previous studies [31], mechanical stress can only produce 90° domain switching. 180° domain switching is irrelevant of stress and activated only by an electric field. It remains to be seen whether these ripples are also contrasts of other domain structures.

Domain switching under an electric field or/and a mechanical field in ferroelectric ceramics has been widely studied for ferroelectric materials. It is reported that polarization switching occurs only when a switching criterion is satisfied [31]. That is to say, there exists a threshold energy for

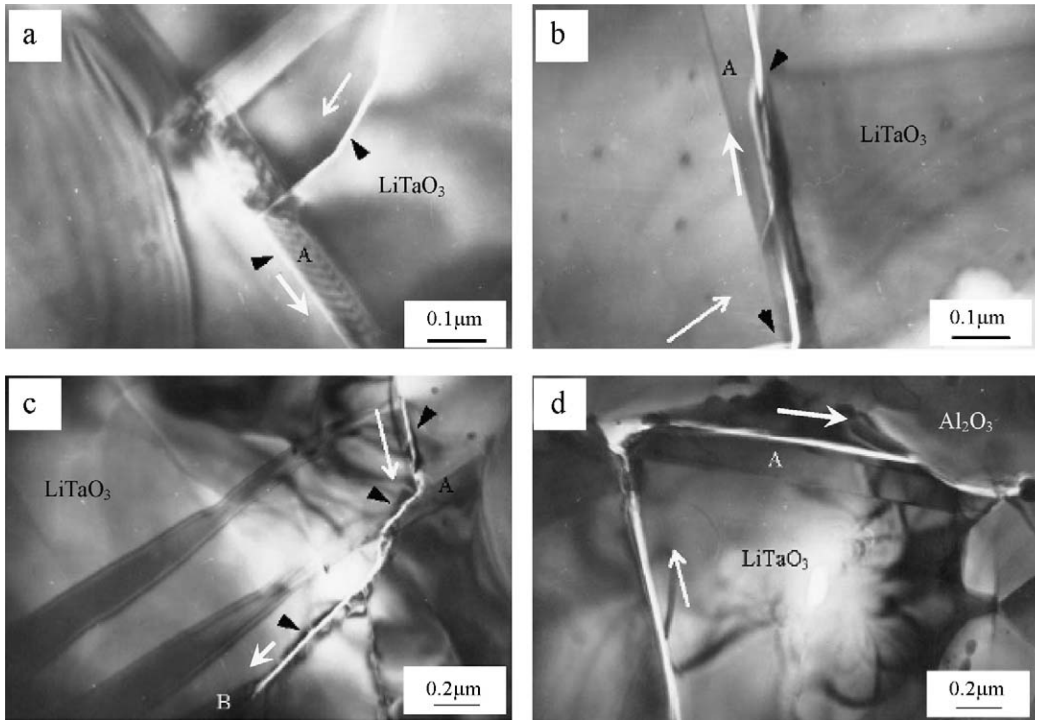

Fig. 2. TEM micrographs of crack deflections and branching in $LiTaO_3$ grains in the as-hot-pressed 15LTA ceramic composite. (a) Crack deflection of about 90°, (b) crack deflection less than 90° and crack branching, (c) crack deflection less than 90°, (d) crack deflection more than 90°.

polarization switching. In the as-hot-pressed LTA ceramic composite, contrasts similar to 180° domains could be caused at or near the crack tip. When the contrasts reached a certain quantity, high stress concentration would be induced. When the total stress concentration reached the threshold for the domain to switch, then 90° domain switching occurred. Domain switching contributes to a toughening of the materials in two ways. On one hand, domain switching will dissipate some of the energy for crack propagation [32]. On the other hand, domain switching right before the crack tip can also change the stress field from tensile to compressive [33]. The above two factors are both helpful to impede the crack propagation and contribute to the improvement of the LTA ceramic composite. This toughening mechanism has relations with the distribution and contents of ferroelectric particles in the matrix, the density of the composite and the domain configuration such as the size and quantity of domains in the piezoelectric second phase. For the as-hot-pressed LTA ceramic composite, only when the composite had a high relative density and enough stable $LiTaO_3$ particles were distributed in the Al_2O_3 matrix and remained ferroelectric at the same time, could the domain switching toughening mechanism function effectively.

Crack deflection and branching toughening caused by the 90° domains in the $LiTaO_3$ particles should be one of the main toughening mechanisms in Al_2O_3 ceramics with a $LiTaO_3$ second phase. The schematic of crack deflection and the branching toughening mechanism in Al_2O_3 ceramics with a $LiTaO_3$ second phase is illustrated in Fig. 4. When the crack propagated into $LiTaO_3$ grains with a 90° domain, crack deflection and the branching occurred at the domain and grew along the domain boundaries. The mechanism is different from the conventional crack deflection and branching toughening mechanism. The latter is produced due to the change of the stress field caused by the existence of a second phase. The

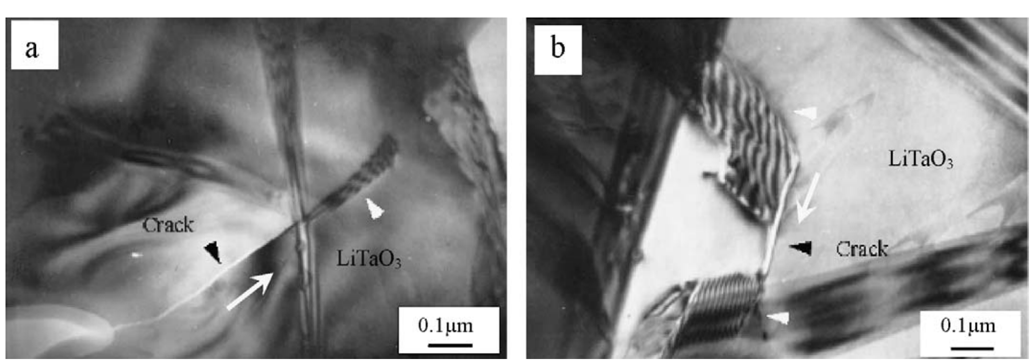

Fig. 3. TEM micrographs of contrasts similar to 180° domains right before a microcrack tip (a) and on one side of a microcrack tip (b) in $LiTaO_3$ grains.

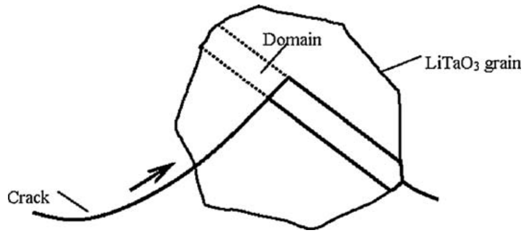

Fig. 4. Schematic illustration of crack deflection and branching at the domain boundaries in a $LiTaO_3$ grain.

former was caused by the domain configuration in the $LiTaO_3$ second phase. Crack deflection and branching extend the crack propagation path, increase the fracture energy and dissipate some of the energy required for cracks to grow. As a result, the toughness of the composite was enhanced.

Formation of ripples similar to the contrasts of 180° domains at or near the propagating crack tip in $LiTaO_3$ grains would also dissipate some of the energy required for the crack to grow. As a result, the propagating of the crack was counteracted. This effect could also contribute to the improvement of the mechanical properties of LTA ceramic composites. This toughening mechanism is named as energy dissipation toughening resulting from the formation of contrasts similar to 180° domains at the microcrack tip.

4. Conclusions

In situ TEM observation was adopted to investigate the crack propagation paths in Al_2O_3 ceramics reinforced by a $LiTaO_3$ secondary phase. Domain switching induced by crack propagating, crack deflection and branching at domain boundaries and ripples similar to contrasts of the 180° domains at the microcrack tip inside the $LiTaO_3$ grains were detected evidently. The improvement of fracture toughness of the Al_2O_3 ceramics with the $LiTaO_3$ secondary phase is the complex result of several toughening mechanisms, including domain switching toughening, crack deflection and branching toughening due to the presence of domains, and energy dissipation toughening resulting from the formation of contrasts similar to the 180° domains at the microcrack tip. These mechanisms were all related directly with the domain configurations in the $LiTaO_3$ second phase.

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