

# Microstructure and mechanical properties of a lithium tantalate-dispersed-alumina ceramic composite

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Received 15 May 2001; received in revised form 6 June 2001; accepted 9 July 2001

## Abstract

Ceramic composites with a ferroelectric/piezoelectric secondary phase were studied. Lithium tantalate ( $\text{LiTaO}_3$ ) was found to be phase compatible with alumina ( $\text{Al}_2\text{O}_3$ ) during sintering.  $\text{LiTaO}_3/\text{Al}_2\text{O}_3$  ceramic composites were fabricated by two processing routes. The microstructure and mechanical properties of the ceramic composites were investigated comparatively. Domain structures were clearly seen in  $\text{LiTaO}_3$ , confirming that  $\text{LiTaO}_3$  remained ferroelectric.  $\sigma_f$  and  $K_{1C}$  reached 438.7 MPa and  $5.4 \text{ MPa}\cdot\text{m}^{1/2}$ , respectively, for 5 vol.%  $\text{LiTaO}_3/\text{Al}_2\text{O}_3$  ceramic composite prepared by cold isostatically pressing and pressureless sintering in air at  $1300^\circ\text{C}$  followed by hot isostatically pressing at  $1300^\circ\text{C}$ , where energy dissipation due to domain switching or/and the piezoelectric effect is suggested as a new toughening mechanism in structural ceramic. The toughening effect is counteracted for higher concentrations of such ferroelectric/piezoelectric secondary phase due to the decreased relative density. © 2002 Elsevier Science Ltd and Techna S.r.l. All rights reserved.

**Keywords:** C. Mechanical properties; C. Toughening; D.  $\text{Al}_2\text{O}_3$ ; Ceramic composite;  $\text{LiTaO}_3$

## 1. Introduction

Recently, the effective strengthening and toughening mechanisms in structural ceramics have been introduced into functional ceramics to improve their mechanical properties significantly [1–3]. Incorporating functional ceramics into structural ceramics has also been reported [4–6]. Though some of their mechanical properties have been also improved, phase compatibility between structural and functional ceramics during sintering is still an urgent problem. In this study, lithium tantalate ( $\text{LiTaO}_3$ ) was found to be phase compatible with alumina ( $\text{Al}_2\text{O}_3$ ) during sintering, and an  $\text{Al}_2\text{O}_3$  matrix ceramic composite dispersed with  $\text{LiTaO}_3$  was successfully fabricated by two processing routes. The microstructure and mechanical properties of the ceramic composites were investigated comparatively.

## 2. Experimental procedure

Commercially available  $\text{Al}_2\text{O}_3$  powder (High Tech Ceramic Institute, Beijing, China) and  $\text{LiTaO}_3$  powder (Dongfang Tantalum Joint-stock Corporation, Ningxia, China) were used as starting powders. The average particle sizes of  $\text{Al}_2\text{O}_3$  and  $\text{LiTaO}_3$  powders were  $\sim 0.65$  and  $3.0 \mu\text{m}$ , respectively. The amount of  $\text{LiTaO}_3$  varied as 5, 10, 15, and 20 vol.%.  $\text{Al}_2\text{O}_3$  and  $\text{LiTaO}_3$  were weighed and then mixed for 24 h with  $\text{Al}_2\text{O}_3$  balls. Ethanol was used as a medium for the ball milling. The slurry was stirred and dried slowly to remove the ethanol. Two processing routes were applied to fabricate the composites: hot pressing (HP) at  $1500^\circ\text{C}$  for 40 min under a pressure of 25 MPa (denoted by No. 1 route), cold isostatically pressing (CIP) under a pressure of 200 MPa and then pressureless sintering in air at  $1300^\circ\text{C}$  for 3 h followed by hot isostatically pressing (HIP) at  $1300^\circ\text{C}$  in argon for 1 h under a pressure of 150 MPa (denoted by No. 2 route). The sintered samples were cut by a diamond-blade saw to sizes of  $3\times 4\times 36 \text{ mm}$  for flexural strength test and  $2\times 4\times 20 \text{ mm}$  for fracture toughness test. The crystalline phase was examined via X-ray diffractometry (XRD) with  $\text{CuK}_\alpha$  radiation as X-

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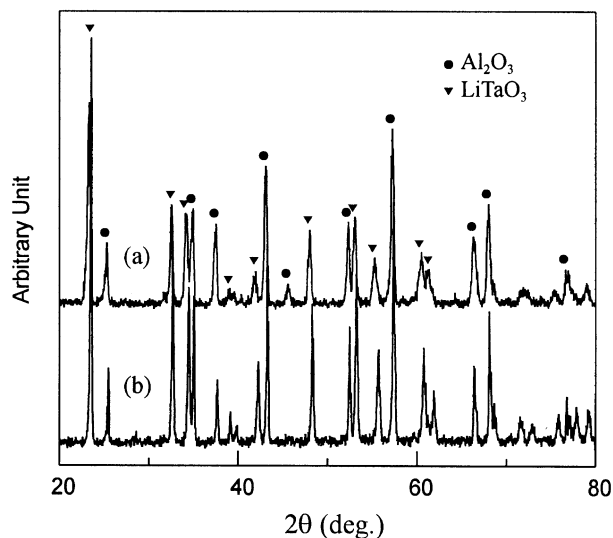


Fig. 1. XRD profiles of 20LTA ceramic composites fabricated by (a) No. 1 route and (b) No. 2 route.

ray source. The microstructure was investigated using scanning electron microscopy (SEM). Domain structures were studied using transmission electron microscopy (TEM). The flexural strength was measured via three-point-bending technique with a span of 30 mm and a crosshead speed of 0.5 mm/min. The fracture toughness was determined via single-edge-notched-beam (SENB) test with a span of 16 mm and a crosshead speed of 0.05 mm/min. Three to five specimens were tested to determine the flexural strength and fracture toughness for each condition.

### 3. Results and discussion

Fig. 1 shows the XRD profiles of 20 vol.%LiTaO<sub>3</sub>/Al<sub>2</sub>O<sub>3</sub> (denoted by 20LTA) ceramic composites fabricated by different routes. All the peaks of 20LTA ceramic composites were assigned to Al<sub>2</sub>O<sub>3</sub> or LiTaO<sub>3</sub>, and no reaction phase between Al<sub>2</sub>O<sub>3</sub> and LiTaO<sub>3</sub> was detected.

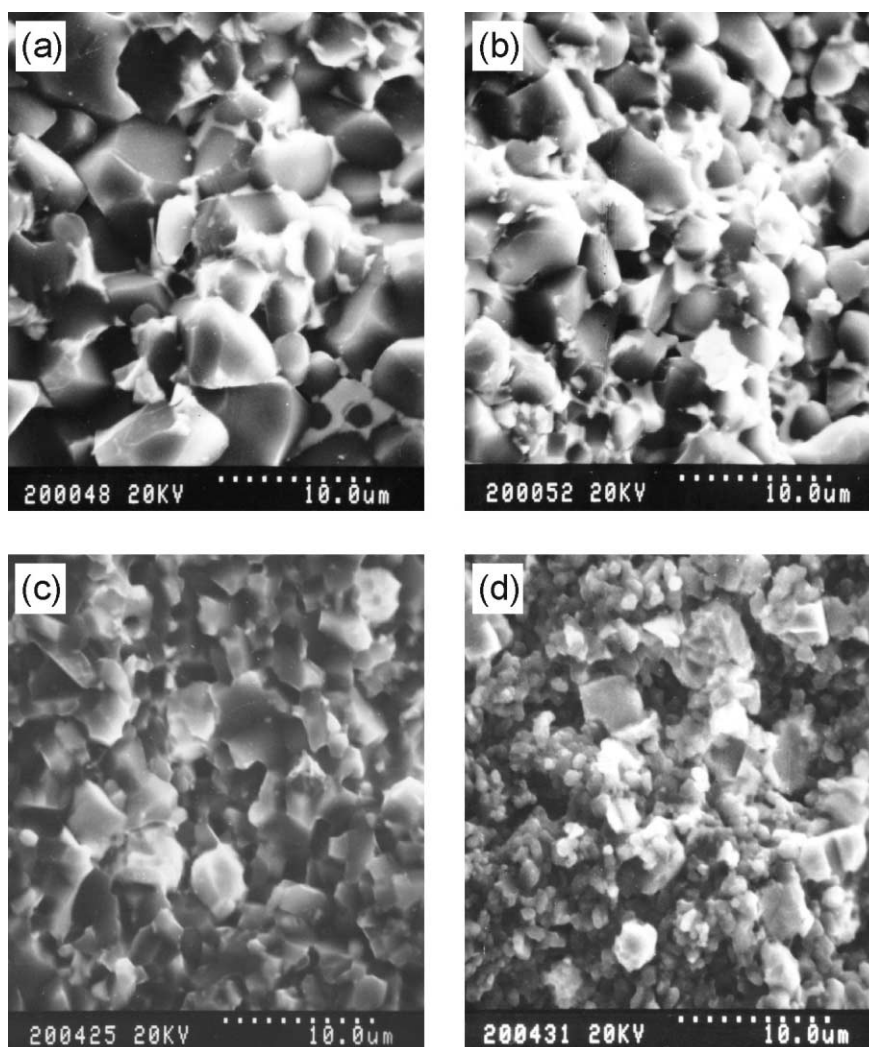


Fig. 2. SEM fractographs of (a) 5LTA, (b) 20LTA fabricated by No. 1 route and (c) 5LTA, (d) 20LTA fabricated by No. 2 route.

Thus,  $\text{Al}_2\text{O}_3$  was found to be phase compatible with piezoelectric  $\text{LiTaO}_3$  during sintering.

Typical fractographs of LTA ceramic composites fabricated by different routes are shown in Fig. 2. When No. 1 processing route was used,  $\text{LiTaO}_3$  melted and was located at the grain boundaries of  $\text{Al}_2\text{O}_3$  matrix after cooling. At the same time, the grain size of  $\text{Al}_2\text{O}_3$  matrix increased significantly mainly due to the melting of  $\text{LiTaO}_3$ . While  $\text{LiTaO}_3$  particles were homogeneously distributed in  $\text{Al}_2\text{O}_3$  matrix when No. 2 processing route was adopted. With the increase of  $\text{LiTaO}_3$  content, more  $\text{LiTaO}_3$  particles agglomerated and the relative density of the composites decreased sharply. Fig. 3 shows the TEM micrograph of 10 vol.% $\text{LiTaO}_3/\text{Al}_2\text{O}_3$  (denoted by 10LTA) ceramic composite fabricated by No. 2 route. Domain structures were clearly seen in  $\text{LiTaO}_3$  grain, confirming that  $\text{LiTaO}_3$  remained hexagonal ferroelectric phase.

The mechanical properties of LTA ceramic composites fabricated by different routes are shown in Fig. 4. When No.1 processing route was adopted, the addition of piezoelectric  $\text{LiTaO}_3$  decreased the flexural strength and fracture toughness of LTA ceramic composites due to the coarsening of  $\text{Al}_2\text{O}_3$  matrix grains and melted  $\text{LiTaO}_3$  with lower strength along the grain boundaries of  $\text{Al}_2\text{O}_3$  matrix. While the LTA ceramic composite with modest amount of  $\text{LiTaO}_3$  by No. 2 processing route showed a significant increase in mechanical properties.  $\sigma_f$  and  $K_{IC}$  reached 438.7 MPa and  $5.4 \text{ MPa}\cdot\text{m}^{1/2}$ , respectively, for 5 vol.% $\text{LiTaO}_3/\text{Al}_2\text{O}_3$  (denoted by 5LTA) ceramic composite. With further increase of  $\text{LiTaO}_3$  content, the mechanical properties of the composites decreased due to the decreased relative density. It indicated that a toughening effect resulted when a compatible ferroelectric/piezoelectric secondary phase was introduced into alumina ceramic by suitable processing route. The toughening effect was limited for higher

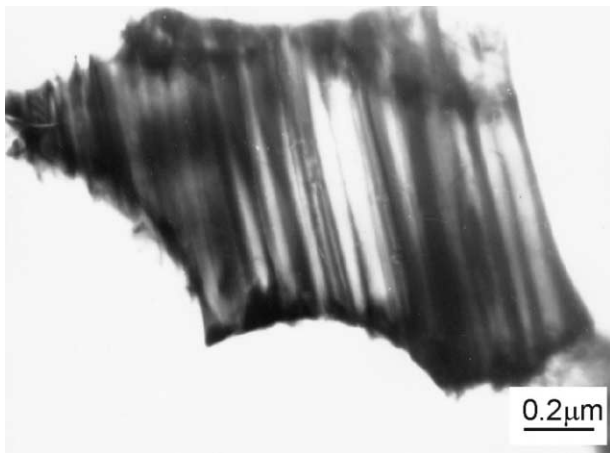


Fig. 3. TEM micrograph of domain structures in  $\text{LiTaO}_3$  grain of 10LTA ceramic composite fabricated by No. 2 route.

concentrations of such ferroelectric/piezoelectric secondary phase due to the decreased relative density.

According to the previous work by Wahi et al. [7,8], the necessary condition for brittle particle toughening is:

$$\Delta E > 0, \quad \Delta \gamma > 0 \quad (1)$$

where,  $\Delta E$  and  $\Delta \gamma$  are the differences of Young's modulus and fracture surface energy between the brittle particle and the matrix, respectively. In the present system, however, the condition is unsatisfied. At the same time, brittle particles can toughen ceramics through different mechanisms, such as crack deflection and microcrack, respectively. As shown in Fig. 5, the crack did not deflect at  $\text{LiTaO}_3$  particles and cut across  $\text{LiTaO}_3$  instead. Neither did microcrack occur at  $\text{LiTaO}_3$  particles. It indicated that some special toughening mechanism took into effect. For ferroelectric/piezoelectric ceramics under mechanical loading, a part of mechanical energy, causing

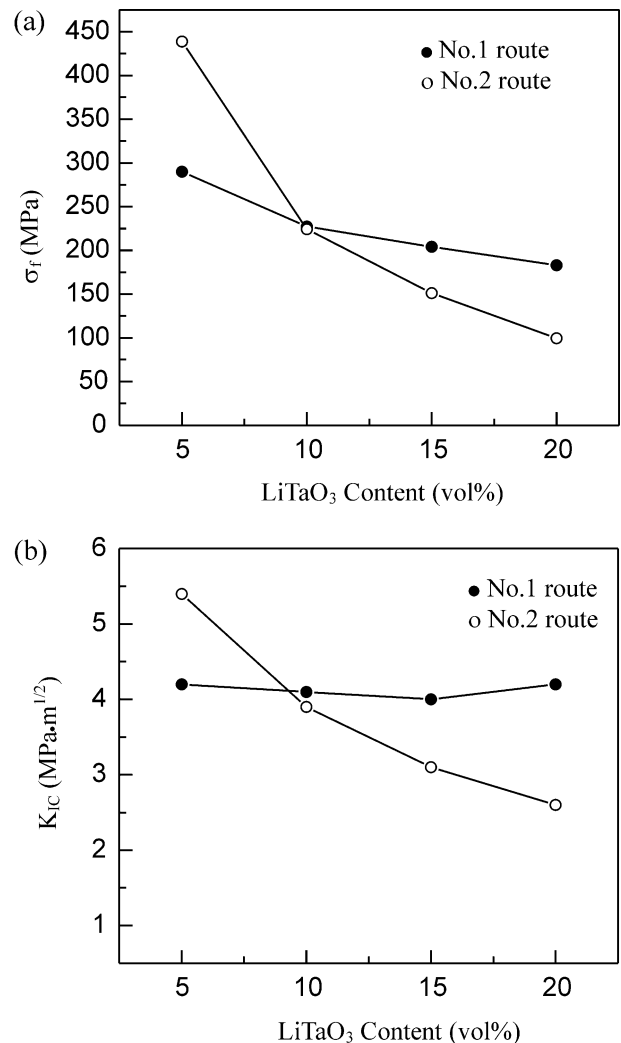


Fig. 4. Three-point-bending strength (a) and fracture toughness (b) of LTA ceramic composites fabricated by different routes vs.  $\text{LiTaO}_3$  content.

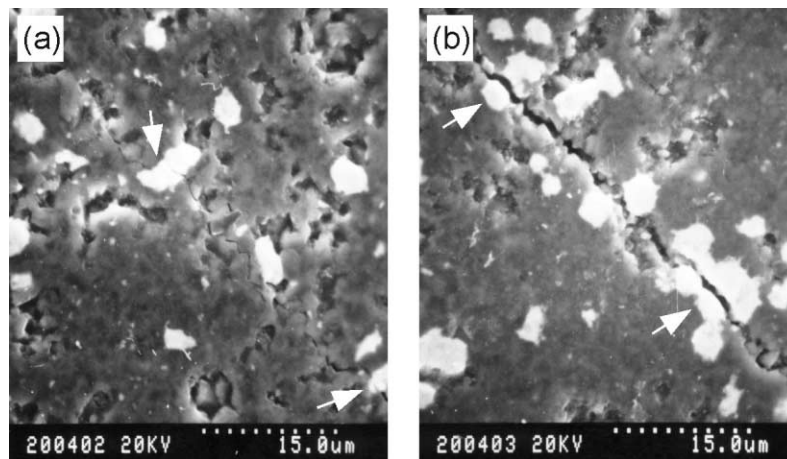


Fig. 5. Crack propagation paths in (a) 5LTA and (b) 10LTA ceramic composites fabricated by No. 2 route.

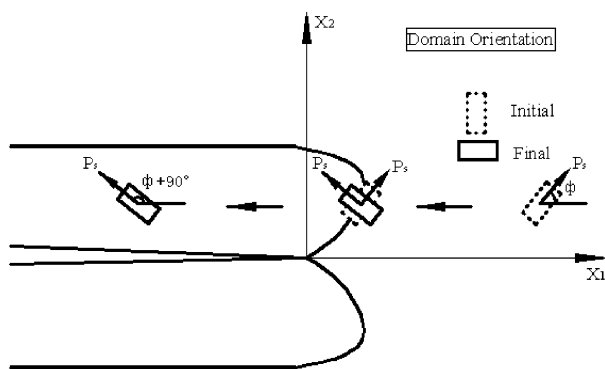


Fig. 6. Schematics of 90° domain switching in ferroelectric materials activated by the stress concentration near a growing crack tip [10].

a crack extension, may be transformed into electrical energy due to piezoelectric effect or dissipated by stress-induced ferroelastic phase transformation and domain switching [4,9]. As the crack in ferroelectric materials advances, stress concentration near the crack tip produces a confined domain switching zone. Thus, a growing crack would result in domain switching strips as shown in Fig. 6 [10]. Therefore, the strengthening and toughening of LTA ceramic composite may be attributed to the domain switching in LiTaO<sub>3</sub> particles. According to the TEM observation, the clearly visible domain structures in LiTaO<sub>3</sub> grain of LTA ceramic composite provided further evidence for domain switching toughening mechanism. Thus, when a compatible ferroelectric/piezoelectric secondary phase is introduced into alumina ceramic by suitable processing route, the energy dissipation due to domain switching or/and the piezoelectric effect is suggested as a new toughening mechanism.

#### 4. Conclusions

In this study, LiTaO<sub>3</sub> was added into Al<sub>2</sub>O<sub>3</sub> matrix to explore the influence of ferroelectric/piezoelectric phase

on the mechanical properties of structural ceramics. When hot pressed at 1500 °C, the flexural strength and fracture toughness of the LTA ceramic composites decreased greatly due to the coarsening of Al<sub>2</sub>O<sub>3</sub> grains and weaker LiTaO<sub>3</sub> along the grain boundaries of Al<sub>2</sub>O<sub>3</sub> matrix. LiTaO<sub>3</sub> particles were homogeneously distributed in Al<sub>2</sub>O<sub>3</sub> matrix after CIP and pressureless sintering in air at 1300 °C followed by HIP at 1300 °C. The LTA ceramic composite with modest amount of LiTaO<sub>3</sub> showed a significant increase in mechanical properties, where energy dissipation due to domain switching or/and the piezoelectric effect is suggested as a new toughening mechanism in structural ceramic. The toughening effect is counteracted for higher concentrations of such ferroelectric/piezoelectric secondary phase due to the decreased relative density.

#### Acknowledgements

We would like to thank the support from the National Nature Science Foundation of China.

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