

Effect of microwave sintering temperature on the microstructure and fracture toughness of $\text{La}_2\text{O}_3/\text{Nb}_2\text{O}_5$ doped Al_2O_3 ceramics

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Keywords: Al_2O_3 ceramics; microwave sintering temperature; columnar grains; fracture toughness; $\text{Nb}_2\text{O}_5/\text{La}_2\text{O}_3$

Abstract: Al_2O_3 ceramics doped with 15% Nb_2O_5 and 7.5% La_2O_3 (volume percent) were prepared by microwave sintering at 1450~1550°C for 30min. The effect of sintering temperature on the microstructure, especially Al_2O_3 columnar grains, and the fracture toughness were investigated. The results show that the phases of sintered samples were α - Al_2O_3 , LaNbO_4 and Nb_2O_5 , without La_2O_3 . During the sintering period, the in-situ reacted LaNbO_4 , and the remained Nb_2O_5 after reacting, changed to liquid which promoted the growth of Al_2O_3 columnar grains and sintering densification. With the increase of sintering temperature, more and more columnar Al_2O_3 grains grew instead of equiaxial grains, and the columnar grains length increased from 8 μm to 22 μm . The average ratio of length to diameter of columnar grains increased firstly and reached to the maximum of 5:1 at 1500°C, then keep unchanged. The fracture toughness increased with the increase of sintering temperature, from 4.83 to 6.48 $\text{MPa}\cdot\text{m}^{1/2}$.

Introduction

Alumina (Al_2O_3) ceramic, as a kind of structural ceramics, is widely used in engineering due to its high hardness, high wear resistance and corrosion resistance, excellent high temperature stability, in addition to its low cost. At present the main problems of the application of alumina ceramics are still their high brittleness[1] and high sintering temperature[2]. The studies[3-6] of many types of ceramic materials confirmed that microwave sintering can reduce the sintering temperature and sintering time of alumina ceramic. Moreover, the additives doped to alumina matrix and then changing to liquid in the sintering process, can not only promote the densification of alumina but also make the columnar or sheet alumina grains obtained, which improves the toughness of alumina effectively. In this study, Al_2O_3 ceramics doped with 15% Nb_2O_5 and 7.5% La_2O_3 (volume percent) were prepared by microwave sintering. Furthermore, the effect of sintering temperature on the densification, microstructure and fracture toughness were investigated.

Raw materials and experimental methods

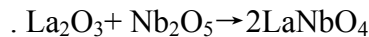
α - Al_2O_3 (99.6 wt%, average particle size of 4 μm) powder are produced by LeiPu Ceramic Material Co. Ltd., Zibo, Shandong, China; Nb_2O_5 (99.99 wt%, average particle size of 8 μm) and La_2O_3 (99.99 wt%, average particle size of 2 μm) powder are both produced by Sinopharm Chemical Reagent Co. Ltd., Shanghai, China. The uniformly mixed powder was obtained after the ball milling of raw material powder followed by drying. Then the composite powder were prepressed into regular bars under a pressure of 100MPa with suitable amount of 5wt% solution of polyvinyl alcohol (PVA) as the binder, and then cold-isostatically pressed under 200MPa. After calcined in conventional furnace at 600°C for 3h to remove organic binder, the green samples were sintered in a 2.45GHz multimode microwave sintering furnace in air atmosphere. Altogether five kinds of samples with the same component of 15% Nb_2O_5 7.5% La_2O_3 77.5% Al_2O_3 were sintered at 1450°C, 1475°C, 1500°C, 1525°C, 1550°C respectively, for the same time of 30min.

The densities of the sintered samples were measured by Archimedes method. The phase

identifications were carried out by X-ray diffractometer (XRD, D8ADVANCE, Bruker-AXS). The microstructure and elemental analysis were performed on fully polished and thermal etched specimens using scanning electron microscope (SEM, Nova Nano SEM450, FEI) and energy dispersed spectroscopy (EDS), respectively. Fracture toughness (K_{IC}) was measured by single edge precracked beam (SEPB) method (National Standard of the People's Republic of China, GB/T 23806-2009).

Results and analysis

The XRD spectrum of the sample sintered at 1450°C for 30min were shown in Fig.1. It was found that a new phase (LaNbO_4) was detected, while La_2O_3 phase undetected, indicating that La_2O_3 reacted completely with Nb_2O_5 at $\sim 1000^\circ\text{C}$ [7]:



A little Nb_2O_5 phase remained after this reaction because the moore content of Nb_2O_5 is more than that of La_2O_3 . All the XRD spectrum results of samples sintered at different temperature were similar to Fig.1. Importantly, the LaNbO_4 and Nb_2O_5 may change to liquid phases when sintered at high temperature (above 1400°C) due to their low melting point.

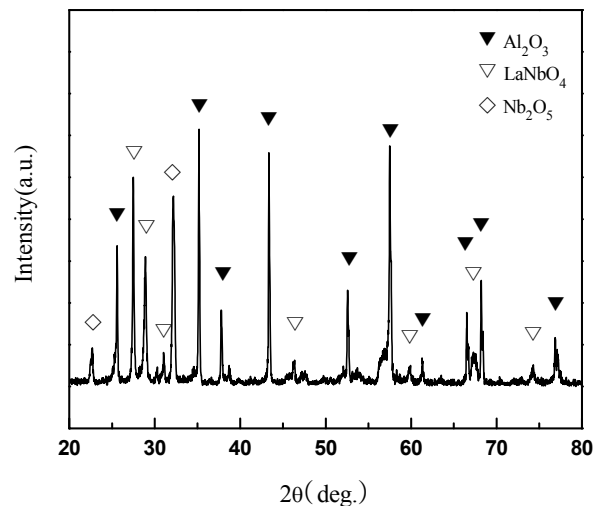


Fig.1. XRD spectrum of 15 Nb_2O_5 /7.5 La_2O_3 /77.5 Al_2O_3 ceramics sintered at 1450°C

Fig.2 presents the surface SEM images of the thermally etched samples. It was inferred according to the EDS analysis that the dark grey, large size phases were Al_2O_3 grains. The light grey phases, with no rules form distribution between the alumina grains, were mainly LaNbO_4 , as shown by arrows in Fig.2. However, the Nb_2O_5 phases were difficult to observe because of their small size and amount. As can be seen from Fig.2(a), Al_2O_3 grains were very small when sintered at 1450°C, and most of them were equiaxial, little were columnar. Simultaneously, some pores were observed, as the result of insufficient sintering driving force, which made the grain growth and pore eduction difficult. With the increase of sintering temperature, the amount and length of columnar grains increased, together with the amount of liquid phases [Fig.2(b)~(e)]. When sintered at 1550°C, the liquid LaNbO_4 phases gathered with large size, and individual Al_2O_3 grain abnormally grew up [Fig.2(e)]. The existence of liquid phases promoted the sintering densification, which caused the relative density increased from 88.8% sintered at 1450°C to 93.2% at 1550°C, as shown in Fig.3.

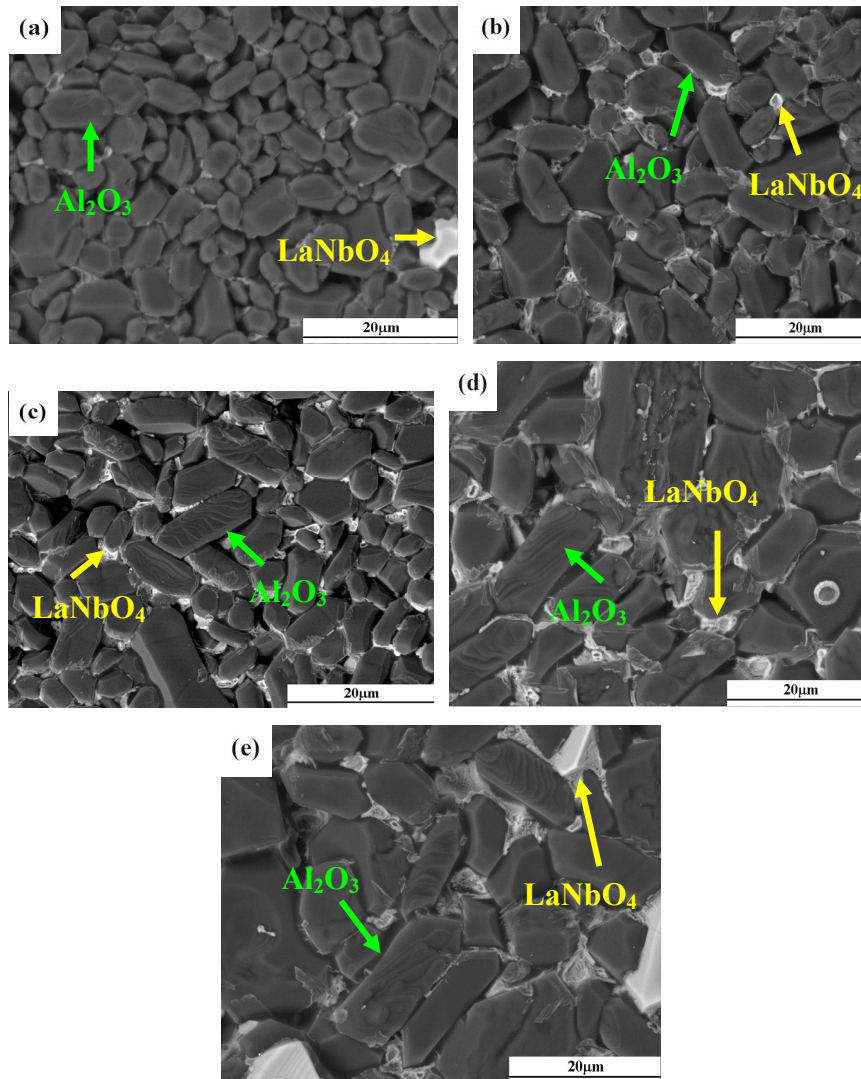


Fig.2. SEM images of 15Nb₂O₅7.5La₂O₃77.5Al₂O₃ ceramics sintered at (a)1450°C; (b)1475°C; (c)1500°C; (d)1525°C; (e)1550°C

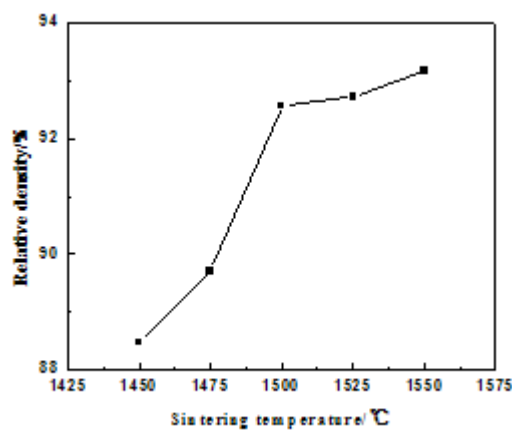


Fig.3. Relative densities of 15Nb₂O₅7.5La₂O₃77.5Al₂O₃ ceramics sintered at different temperature

The average length of Al₂O₃ columnar grains increased with the increase of sintering temperature as well, from 8µm at 1450 °C to 22µm at 1550°C (Fig.4). This phenomenon could be mainly attributed to the increasing amount of liquid phases, including LaNbO₄ and Nb₂O₅. The liquids

accelerated the diffusion speed and reduce the interface energy, which induced the anisotropic growth of the Al_2O_3 crystal. Moreover, the grains were easier to grow along the vertical direction of base plane, because the base plane was easier to be wetted by liquid phase[8]. Thus columnar Al_2O_3 grains gradually formed owing to the higher growth rate along the vertical direction of base plane.

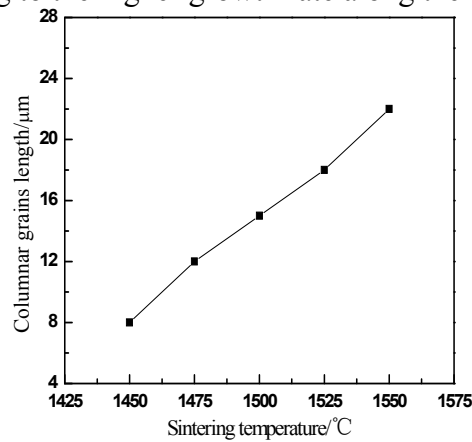


Fig.4. Columnar grains length of $15\text{Nb}_2\text{O}_57.5\text{La}_2\text{O}_377.5\text{Al}_2\text{O}_3$ ceramics sintered at different temperature

The ratio of length to diameter of Al_2O_3 columnar grains firstly increased with the increase of sintering temperature, from 3:1 at 1450°C to 5:1 at 1500°C (shown in Fig.5). However, the ratio increase was not always linear when sintered at temperature above 1500°C . At higher temperature more liquid phases formed around Al_2O_3 grains, causing the grain growth rate almost same along all the directions. Furthermore, the diffusion at higher temperature tended to be more uniform, so the average ratio of length to diameter remained unchanged after reaching the maximum of 5:1.

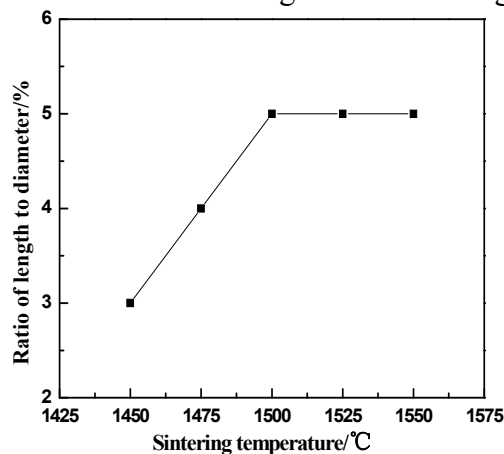


Fig.5. Ratio of length to diameter of Al_2O_3 columnar grains of $15\text{Nb}_2\text{O}_57.5\text{La}_2\text{O}_377.5\text{Al}_2\text{O}_3$ ceramics

The test results of fracture toughness were presented in Fig.6. The fracture toughness increased with the increase of sintering temperature, from 4.83 to $6.48 \text{ MPa}\cdot\text{m}^{1/2}$. The toughening reasons can be summarized as follows except the increase of density:

Firstly, the amount and length of Al_2O_3 columnar grains increased with the increase of sintering temperature. The toughening mechanisms of columnar grains had be concluded by crack deflection, crack branching and columnar grains pullout[9], which increased the crack propagation path and the fracture energy.

Secondly, the in-situ grown LaNbO_4 , which has multiple domain structure[10,11], can realize the domain switching in order to reduce the stress concentration at crack tip by absorbing part of the crack propagation energy[12].

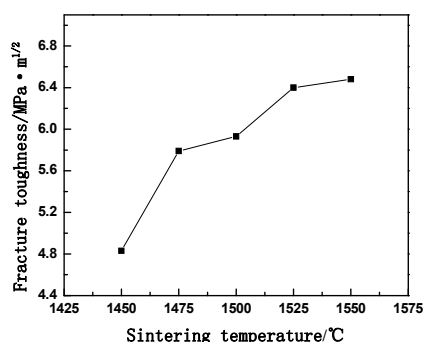


Fig.6. Fracture toughness (K I C) of 15Nb₂O₅7.5La₂O₃77.5Al₂O₃ sintered at different temperature

Conclusions

15%Nb₂O₅7.5%La₂O₃77.5%Al₂O₃ composite ceramics were prepared by microwave sintering at the temperature range of 1450~1550 °C for 30min. The phases of sintered samples were α -Al₂O₃, LaNbO₄ and Nb₂O₅, without La₂O₃. During the sintering period, the in-situ reacted LaNbO₄, and the remained Nb₂O₅ after reacting, changed to liquid which helped to the growth of Al₂O₃ columnar grains and sintering densification. With the increase of sintering temperature, more and more columnar Al₂O₃ grains grew instead of equiaxial grains, and the columnar grains length increased from 8 μ m to 22 μ m. The average ratio of length to diameter of columnar grains increased firstly and reached to the maximum of 5:1 at 1500 °C, then keep unchanged. The fracture toughness increased with the increase of sintering temperature as well, from 4.83 to 6.48 MPa·m^{1/2}.

Acknowledgements

This work was financially supported by National Natural Science Foundation of China (51064022).

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