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Effect of rod-like grain on properties and toughening mechanism of 3Y-TZP/Al_2O_3 ceramics

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Abstract: In-situ rod-like Al_2O_3 grain was prepared by adding CAS multiphase additives under the sintering condition of 30 MPa, 1 550 ℃ and 1 h. The sintering behaviors, microstructure, toughening mechanism and access of Al_2O_3 ceramics were investigated by SEM, EDS and WDW omnipotent electronic mechanical testing machine, etc, and the crack propagating model of cylindrical crystal/3Y-TZP composite toughening Al_2O_3 ceramics was established. The results show that the composite additives prompt the anisotropic growth of Al_2O_3 grain, which strengthens toughening effect of 3Y-TZP in 3Y-TZP/Al_2O_3 composite ceramics. Moreover, the experimental material density is near to theoretical density, bending strength is 556.35 MPa, and fracture toughness is 6.73 MPa·m$^{1/2}$. The mechanical properties of the materials are obviously improved.

Key words: rod-like grain; multitoughening; additives; Al_2O_3; microstructure; mechanical properties

1 Introduction

Alumina ceramic is one of the most widely used structural ceramics based on its abundance resource and excellent mechanical performance. However, lower fracture toughness limits its wide application in engineering[1−3]. So many scientists present a series of ameliorating methods. Phase transformation strengthening is a traditional and effective method, which utilizes the transformation of metastable tetragonal zirconia from tetragonal phase to monoclinic one by inducing stress to improve alumina ceramics fracture toughness due to the formation of unelastic distortion areas around cracking tips[4−7]. Recently, the mechanical properties of alumina ceramics synthesized are improved by adding nanoparticle[1,8−9], but the costs of the material and the preparation are increased [10−11]. In-situ toughening is a kind of new effective technology for advancing fracture toughness[3,12−13], which can change alumina ceramics grains from equiaxed to flake, platelets or rod-like shape through introducing additives or seeds[14−18] to toughen the matrix, and in this case, the toughening effect is similar to that of crystal whiskers by providing the bridging of the crack, the deflection of the crack or the pulling out of the grain[19−21].

In the present work, a new type of CaO-Al_2O_3-SiO_2(CAS) composite additives was added into the aluminum oxide ceramics containing 3Y-TZP, TiO_2 and MgO to obtain in-situ rod-like grains based on the phase transformation toughening. The toughening effect of the obtained in-situ resultant is similar to that of the whiskers and cooperates with the phase transformation strengthening toughness to further enhance significantly the mechanical properties.

2 Experimental

2.1 CAS preparation

According to the Al_2O_3-CaO-SiO_2 ternary phase diagram, the powder mixtures of industrial Al_2O_3, CaO and SiO_2 were mixed corresponding to a certain mass fraction, and were ground to micrometer level for 24 h. Subsequently, the mixtures were placed into the sintering furnace to sinter for 3 h at 1 500 ℃ after being sifted

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through 147 μm sieve. And then, the sintered samples were quenched, pestled and wet ground for 48 h in the ceramic mortar. The XRD result showed that the uncrystallized glass powders were obtained after drying through 63 μm sifter.

2.2 Material preparation

Industrial Al_2O_3, self-made CAS glass powder and other materials were ground to micrometer level (average micro size 2 μm) by ultra-fine ball milling. According to the designed components, the mixed powders were wet ground together with ZrO_2 grinding balls and CH_3CH_2OH for 36 h, and the volume ratio is 1:2.5:3. Subsequently, the mixtures were sifted to get particles after being dried at 80 ℃, and was hot-pressing sintered to form sintered body for 1 h under 1 550 ℃ and 30 MPa.

In order to improve the sintered body density and decrease the sintering temperature, 0.4%TiO_2 and 0.1%MgO were added into the materials. The compositions of the sample are listed in Table 1.

2.3 Detecting technique

After being wet ground and polished, specimens were etched with NaOH solution, and were detected by JSM-6360 SEM after being sprayed with Pt. The bending strength was evaluated by three point bending with a specimen size of 3 mm×4 mm×36 mm, span of 30 mm, and loading speed of 0.5 mm/min. Fracture toughness was examined by the single edge notched beam(SENB) method on notched specimens of 2.5 mm×5 mm×30 mm with the notched depth of 1 mm, notched width of 0.2 mm, the span of 20 mm and loading speed of 0.05 mm/min. The crack propagating path was studied by Vicker indentation method. The crack propagating path and the fracture morphology of the specimen were investigated by SEM. The actual densities of the composites were determined using Archimedes method, and relative density was calculated.

3 Results and discussion

3.1 Material properties

3.1.1 Density

Fig.1 shows the relative density change of multiphase alumina ceramics with different mass fractions of CAS powder on the condition of unchanging other additives. It can be seen that with increasing CAS mass fraction, the relative density of the materials increases first, then decreases. When the CAS mass fraction is 0.5%, the relative density of alumina ceramics is approximate to theoretical value, and reaches 98.4%, which is in relation to the shape and size of Al_2O_3 grain, as shown in Fig.2. When the mass fraction of the sinter doping is higher, a great deal of glass phases are formed, and this prompts the growth of Al_2O_3 grain and increases relative density of the material.

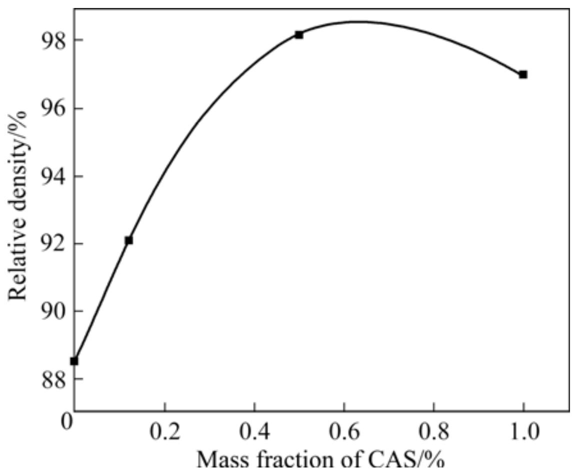

Fig.1 Effect of mass fraction of CAS on relative density

But too much glass phases can lead to the formation of the big size grains, which hampers the flowing of liquid phase. With the grains growing and macrocrystals increasing, the interleaving and bridging of the grains increase, which results in more micro-holes and further reduces the density. Therefore, the content of the additive is not too high.

3.1.2 Mechanical properties

Fig.3 shows the bending strength and fracture toughness of the materials with different mass fractions of CAS powder. It can be observed that the bending strength and fracture toughness are obviously improved

Table 1 Composition of Al_2O_3 ceramics composite

Sample No.	$w(Al_2O_3)$/%	w(3Y-TZP)/%	$w(TiO_2)$/%	w(MgO)/%)	w(CAS)/%
1	89.5	10	0.4	0.1	
2	89.4	10	0.4	0.1	0.1
3	89.2	10	0.4	0.1	0.3
4	89.0	10	0.4	0.1	0.5
5	88.8	10	0.4	0.1	0.7
6	88.6	10	0.4	0.1	0.9
7	88.4	10	0.4	0.1	1.1

with increasing mass fraction of CAS. In this case, with the increase of the CAS content, the creases of interface energy difference between the base facet of Al_2O_3 grain and other ones[12] leads to the change of the growth speed of the grain on various facets. This promotes the anisotropic growth of the grains, and the quantity of the grains increases gradually, as shown in Figs.2(a)−(c). Thus, the density of sintered body is improved (Fig.1).

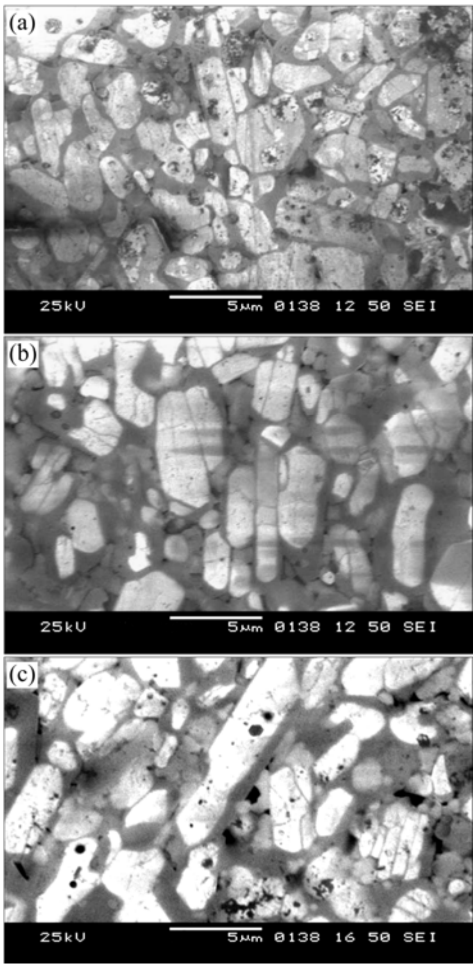

Fig.2 Effect of CAS content on anisotropic grain: (a) 0.1% CAS; (b) 0.5% CAS; (c) 1.0% CAS

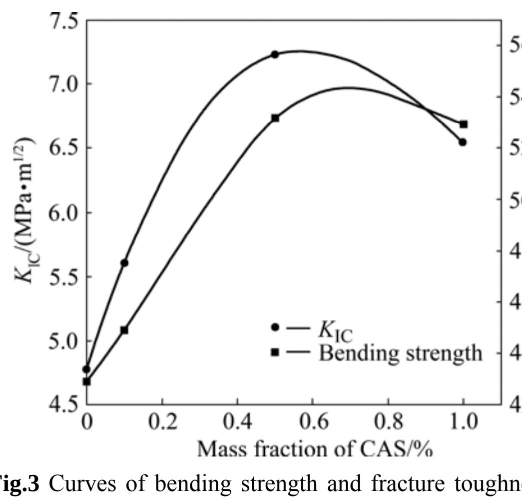

Fig.3 Curves of bending strength and fracture toughness with mass fraction of CAS

When CAS content is low, the solute dragging action of MgO restrains the diffusion speed of Al_2O_3 grain boundary, which leads to incomplete anisotropic grain growth and the increasing of the pore and other defects, and descends the bending strength of the sintered body (Fig.2(a)). With further increasing CAS content, the solute dragging action of partial MgO is restrained, and the cylindrical grain is obtained (Fig.2(b)). Furthermore, the mechanical properties are also enhanced significantly. However, more CAS induces the abnormal growth of the grain, and the big grain usually contains some pores that are difficult to eliminate (Fig.2(c)). This not only reduces the relative density but also weakens the matrix restriction to tetragonal crystal ZrO_2[14−15], leading to more monoclinic phase and relieving the effect of stress induced phase transformation, and further deteriorating the material properties.

In the present work, the optimal effect is obtained when mass fraction of CAS is 0.5%. The bending strength of the material reaches the highest of 556.35 MPa, and fracture toughness achieves 6.73 MPa·m$^{1/2}$.

3.2 Crack propagating path and toughening mechanism

According to the Griffith equation[22]:

$$\sigma_{\mathrm{f}} = \frac{1}{Y}\sqrt{\frac{2E\gamma}{C}} = \frac{1}{Y}\frac{K_{\mathrm{IC}}}{\sqrt{C}} \tag{1}$$

where Y and E are shape factor and elastic modulus; γ, C, K_{IC} are fracture energy, fracture size and fracture toughness.

So the fracture toughness of ceramics material is

$$K_{\mathrm{IC}} = \sqrt{2E\gamma} \tag{2}$$

The higher γ and E, the higher the toughness of the ceramics. But due to limited improvement of E, the main approach of achieving high toughness is enhancing K_{IC}. Therefore, the essential of the toughening lies on the introduction of the mechanism of consuming fracture energy, which can hamper crack propagating and weaken the driving power of crack tip.

Fig.4 shows the typical expanding morphology of pressing marks of the experiment material with CAS mass fraction of 0.5%. When the crack is enlarged to meet with cylindrical crystals, it can deflect and diverge (Fig.4(a)), which changes expanding fracture direction and absorbs fracture energy. The diverging fractures distract the expanding fracture energy, and reduce the stress concentration of the cracking tips and the stress strengthening factor, which relaxes the crack expanding,

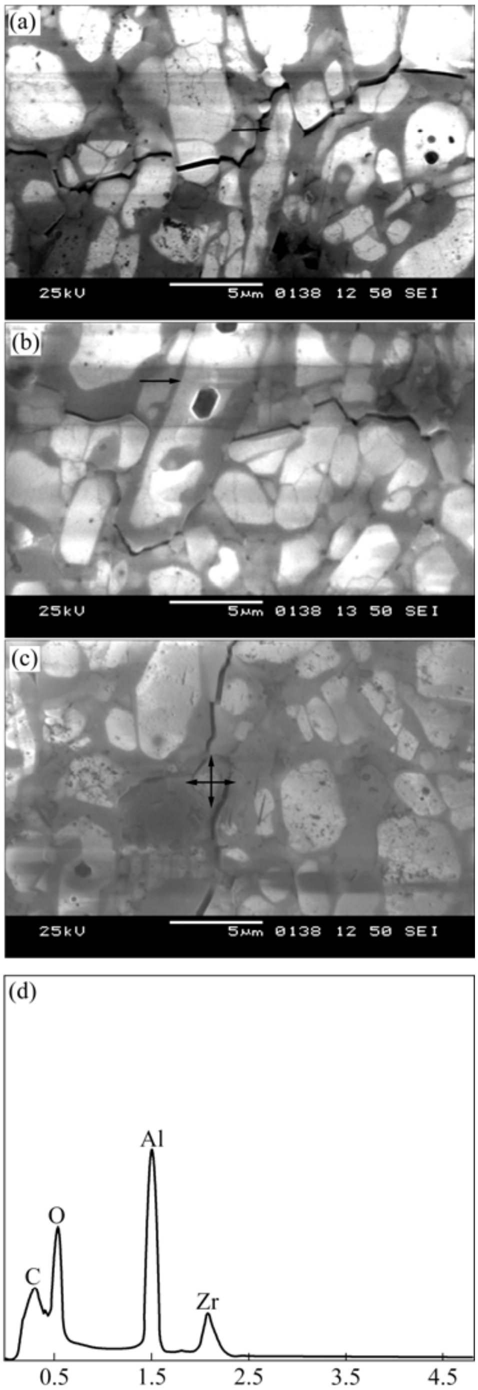

Fig.4 Expanding fracture feature of experimental materials: (a) Crack diverging and deflecting; (b) Rod-like grain pulling out; (c) Rod-like grain friction and grubbing; (d) EDS of Fig.4(c)

and indicates that it is necessary to provide more energy for further enlargement of the main crack. In the process of rod-like grains being pulled, the energy is improved by way of pulling power (Fig.4(b)), which increases the crack expanding resistance and raises the fracture toughness. In addition to the bridging rod-like grains generate pulling effect, and also probably fracture (Fig.4(c)), but the fractured grains do not lose the toughening capability. With the opening of crack, the fractured cylindrical grain segments remaining inside the crack facet go on generating friction with matrix and consuming energy. On the other hand, during the process of bridging and slipping friction of cylindrical grains, a part of matrix near the crack sides is probably shattered because of rod-like grain grubbing, forming the restricting mechanism of rod-like grain bridging and slipping friction, which consumes more energy.

Fig.5 shows the crack expanding model through the multitoughening of rod-like grain and 3Y-TZP, where *A* and *B* are Al_2O_3 rod-like grain and ZrO_2; *C* is alumina ceramic matrix; *D* is the microcrack stress affected region; *E* is the grubbing position; *F* is the direction of the main crack; *G* is the pulling and sliding friction direction of rod-like grain; and *h*, *i*, *j* are pulling directions of rod-like grain, bridging position and crack propagating direction respectively.

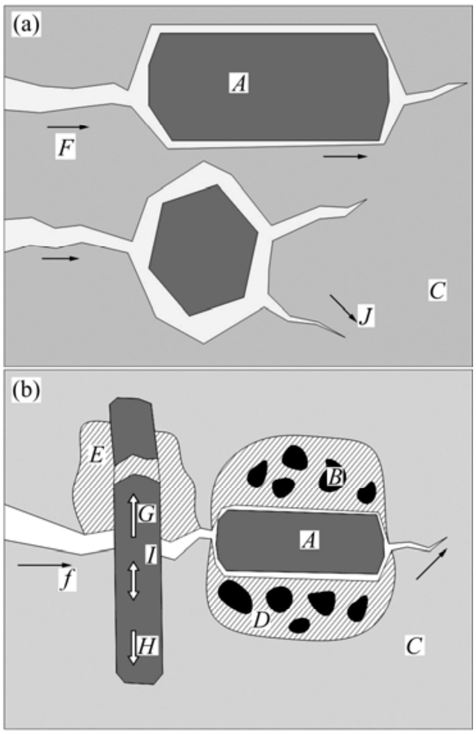

Fig.5 Expanding fracture model of experimental materials

The multitoughening effect is not simple summation of rod-like grain and 3Y-TZP action. In the toughening process through crack deflecting and diverging, rod-like grains extend the crack path and produce stress areas, which prompts the increasing of martensite and strengthens phase transformation toughening effect.

From the analysis of cracking expanding process, it can be concluded that many factors including stress induced phase transformation, crack bridging, crack divergence and crack deflection interact and cooperate to toughen the material, which is superior to the summation of single toughening action.

4 Conclusions

1) The content of CAS additive has remarkable effect on 3Y-TZP/Al_2O_3 composite ceramic materials. The relative density of the materials first increases and then decreases with increasing CAS mass fraction. The excess CAS leads to coarse grain and low density, and the optimal content of CAS is 0.5%.

2) The rod-like grain obviously improves Al_2O_3 ceramics mechanical properties. The bending strength and fracture toughness reach 556.35 MPa and 6.73 MPa·$m^{1/2}$, respectively.

3) The in-situ rod-like grains produced by the inducing of composite additive play an role in toughening by the crack deflecting and diverging; meanwhile, it extends the phase transformation toughening path and prompts the increasing of martensite quantity. And its cooperated toughening effect with 3Y-TZP is superior to the summation of single toughening action.

References

[1] CHOU I A, CHAN H M, HARMER M P. Machining-induced surface residual stress behavior in Al_2O_3/SiC nano-composites [J]. Am Ceram Soc, 1996, 79(12): 2403−2415.

[2] HUANG Yong, ZHANG Li-ming, WANG Chang-an, YANG Jin-long, XIE Zhi-peng. Advanced structure ceramics research and review [J]. Silicate Transaction, 2005(5): 97−107. (in Chinese)

[3] XIE Zhi-peng, GAO Li-chun, LI Wen-chao, et al. Seed inducing rod-like grain growth and higher toughness alumina [J]. Science in China (Series E), 2003(1): 13−20. (in Chinese)

[4] HUANG S G., LI L, VAN DER BIEST, O VLEUGELS J. Microwave sintering of CeO_2 and Y_2O_3 co-stabilised ZrO_2 from stabiliser-coated nanopowders [J]. Journal of the European Ceramic Society, 2007, 27: 689−693.

[5] CESARI F, ESPOSITO L, FURGIUELE F M. Fracture toughness of alumina-zirconia composites [J]. Ceramics International, 2006, 32(3): 249−255.

[6] TEKELI S. Fracture toughness, hardness, sintering and grain growth behaviour of 8YSCZ/Al_2O_3 composites produced by colloidal processing [J]. Journal of Alloys & Compounds, 2005, 391: 217−224.

[7] SZUTKOWSKA M. Fracture resistance behavior of alumina-zirconia composites [J]. Journal of Materials Processing Technology, 2004, 153/154: 868−874.

[8] ZHANG Guo-jun, YANG Jan-feng, ANDO M. Reactive hot pressing of aluminum-silicon carbide nanocomposites [J]. Am Ceram Soc, 2004, 87(2): 299−301.

[9] RUHLE M, EVANS A G, McMEEKING R M, CHARALAM BIDES P G, HUTCHINSON J W. Microcrack toughening in alumina/ zrconia [J]. Acta Metall, 1987, 35(11): 2701−2710.

[10] ACCHAR W, FONSECA J L. Sintering behavior of alumina reinforced with (Ti, W) carbides [J]. Materials Science & Engineering A, 2004, 371: 382−387.

[11] LI Jian-gong, YE Yin-ping. Densification and grain growth of Al_2O_3 nanoceramics during pressureless sintering [J]. Am Ceram Soc, 2006, 89(1): 139−143.

[12] FABER K T, EVANS A G. Crack deflection processes (I): Theory [J]. Acta Metall, 1983, 31(4): 565−576.

[13] DONG Wei-min, JAIN H, HARMER M P. Liquid phase sintering of alumina(I): Microstructure evolution and densification [J]. Am Ceram Soc, 2005, 88(7): 1702−1707.

[14] OLEVSKY E A, KUSHNAREV B, MAXIMENKO A, TIKARE V. Modelling of anisotropic sintering in crystalline ceramics [J]. Philosophical Magazine, 2005, 85(19): 2123−2146.

[15] SHUI A, SAITO H, UCHIDA N, UEMATSU K. Development of anisotropic microstructure in uniaxially pressed alumina compacts [J]. Journal of the European Ceramic Society, 2002, 22(8): 1217−1223.

[16] KWON O S, HONG S H, LEE J H, CHUNG U J, KIM D Y, HWANG N M. Microstructural evolution during sintering of TiO_2/SiO_2-doped alumina: mechanism of anisotropic abnormal grain growth [J]. Acta Materialia, 2002, 50(19): 4865−4872.

[17] HSU Yung-fu. Influence of Nb_2O_5 additive on the densification and microstructural evolution of fine alumina powders [J]. Materials Science & Engineering, 2005, 399: 232−237.

[18] XU Li-hua, XIE Zhi-peng. Synthesis, evaluation and characterization of alumina ceramics with elongated grains [J]. Ceramics International, 2005, 31(7): 953−958.

[19] TARTAI J, MESSING G L. Anisotropic grain growth in α-Fe_2O_3-doped alumina [J]. Journal of European Ceramic Society, 1997, 17: 719−725.

[20] HORN D S, MESSING G L. Anisotropic grain growth in TiO_2-doped alumina [J]. Materials Science and Engineering A , 1995(195): 169−178.

[21] BAE I, BAIK S. Abnormal grain growth of alumina [J]. Am Ceram Soc, 1997, 80(5): 1149−1156.

[22] GREEN D J. Ceramics material mehanics [M]. GONG Jiang-hong, transl. Beijing: Tsinghua University Press, 2003: 194−203. (in Chinese)

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