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Ceramics International 39 (2013) S433–S436

CERAMICS INTERNATIONAL

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Effect of adding Y_2O_3 on structural and mechanical properties of Al_2O_3–ZrO_2 ceramics

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Abstract

The effects of adding 1–8 wt% Y_2O_3 on phase formation and fracture toughness of Al_2O_3–$x$$ZrO_2$–$Y_2O_3$(AZY) ceramics were studied. Phase formations of the samples were characterized by the X-ray diffraction (XRD) technique. It was found that the major phase was rhombohedral-Al_2O_3, while the minor phase consisted of the monoclinic-ZrO_2, tetragonal-ZrO_2 and monoclinic-Y_2O_3. It was found that Y_2O_3 contents did not clearly influence grain shape of AZY ceramics. The results obtained from the microhardness test could be used to evaluate the fracture toughness. It was found that the smaller grains had high fracture toughness. The maximum fracture toughness of 4.827 MPa $m^{1/2}$ was obtained from 4 wt% Y_2O_3. Refinement of lattice parameters using Rietveld analysis revealed the quantitative phases of AZY ceramics. This shows that under adding Y_2O_3 conditions the proportion of tetragonal-ZrO_2 phase plays an important role for the mechanical properties of AZY ceramics.

Keywords: D. Al_2O_3; D. ZrO_2; Phase formation

1. Introduction

Alumina (Al_2O_3) ceramics have been widely used for structural ceramics application [1]. The high strength and fracture toughness that make these ceramics attractive candidates for many demanding applications are the results of transformation toughening. In spite of the variety of useful physical properties of sintered oxide ceramics based on chemically and thermally stable alpha modification of alumina (α-Al_2O_3) their application as cutting tool inserts working under mechanical loads and thermal shock conditions is limited due to their brittleness and low strength. One of the methods to improve these properties is by making use of transformation strengthening process; through phase transformation some amount of ZrO_2 is introduced into Al_2O_3. The mechanism of this process is based on the polymorphic transformation of $ZrO_2^{(t)}$ tetragonal phase into $ZrO_2^{(m)}$ monoclinic phase during cooling from sintering temperature to room temperature, enabling an increase of the strength and/or fracture toughness of alumina ceramics [2]. The stoichiometry of alumina–zirconia is known to be an important factor for ensuring good mechanical properties. Therefore, the alumina–zirconia system (AZX with 15–50 mol% ZrO_2) is interesting to study [3,4]. It is well known that the tetragonal and/or cubic ZrO_2 can be retained to room temperature by doping small amount of stabilizing oxides (e.g. Y_2O_3, MgO, CaO and some rare-earth oxides) [5]. In addition, the metastable tetragonal zirconia without dopants can also be prepared in the form of fine particles that are smaller than the critical size [6–8].

During the present research powders of Al_2O_3–$x$$ZrO_2$ ($x = 35$ mol%) were produced by coprecipitation of aluminum chloride and zirconium oxychoride. Co-precipitated products were doped to different Y_2O_3 ratios in order to optimize ceramics properties. The characteristics of different ceramic products were evaluated by X-ray diffraction (XRD), scanning electron microscopy (SEM), microhardness and phase analysis.

2. Experimental

The Al_2O_3–ZrO_2 powders with 35 mol% ZrO_2 were synthesized using ($AlCl_3 \cdot 6H_2O$, 99.9% pure), ($ZrOCl_2 \cdot 8H_2O$, 99.9% pure) and ammonia solution (NH_4OH,

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http://dx.doi.org/10.1016/j.ceramint.2012.10.108

28%) as raw materials. Aluminum chloride and zirconium oxychoride were mixed and dissolved in distilled water. The solution was thoroughly stirred at room temperature, and then precipitated by adding ammonia to get $Zr(OH)_4$ gel. The precipitate was washed repeatedly using distilled water to remove all the chloride ions (as tested by $AgNO_3$ solution), and then dried in air at 100 °C. Precursor powders were calcined at 1100 °C. Al_2O_3–ZrO_2 powders were added to Y_2O_3 (99.9% pure) with 1–8 wt% and this mixture was ball milled for 24 h. Sintering was done at 1600 °C for 2 h. The bulk densities of sintered sample were calculated using Archimedes's method. The phase compositions of Al_2O_3–ZrO_2–Y_2O_3 ceramics were measured by X-ray diffraction (XRD) using Cukα radiation (Philips PW 1729 diffractometer, Netherlands). Phase analysis was calculated using the Rietveld method. Microstructural evolution of the powders and ceramics were observed using the scanning electron microscopy: SEM (JEOL, JSM 840A, Japan). Microhardness of bulk ceramics was measured using a microscan Vickers and Knoop (FM-700etype D, Future Tech., Japan).

3. Results and discussion

The XRD patterns of Al_2O_3–35 mol% ZrO_2 ceramics with added 2–8 wt% Y_2O_3 after sintering at 1600 °C for 2 h are shown in Fig. 1, which presents the difference of content phases.

Apart from α-Al_2O_3 and Y_2O_3, both t- and m-ZrO_2 are detected. The main reflections in the pattern formed around $2\theta = 25°$, 30°, 35°, 43°, 50°, 57° and 59° match well with characteristic reflections of Al_2O_3–ZrO_2 [4,9]. The tetragonal phases of ZrO_2 are detected by the presence of high intensity peak at $2\theta = 30°$ and splitting of peaks at around $2\theta = 35°$ and 50°. It was found that at $2\theta = 50°$ it is clearly indicated that the t-ZrO_2 phase transformed

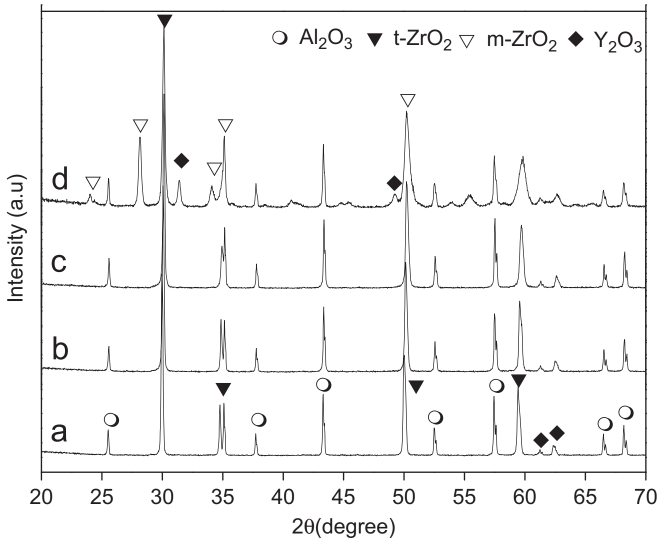

Fig.1. XRD patterns of Al_2O_3–35 mol% ZrO_2 ceramics with added 2–8 wt% Y_2O_3: (a) 2 wt%, (b) 4 wt%, (c) 6 wt% and (d) 8 wt%.

Table 1
Densities shrinkage and average grain size of Al_2O_3–35 mol% ZrO_2 ceramics with different Y_2O_3 contents.

Contents of Y_2O_3 (wt%)	Density (g/cm^3)	Shrinkage (%)	Average grain size (μm)
2	4.90	24.54	1.19
4	4.95	24.62	1.65
6	4.89	24.48	1.95
8	4.90	24.54	1.19

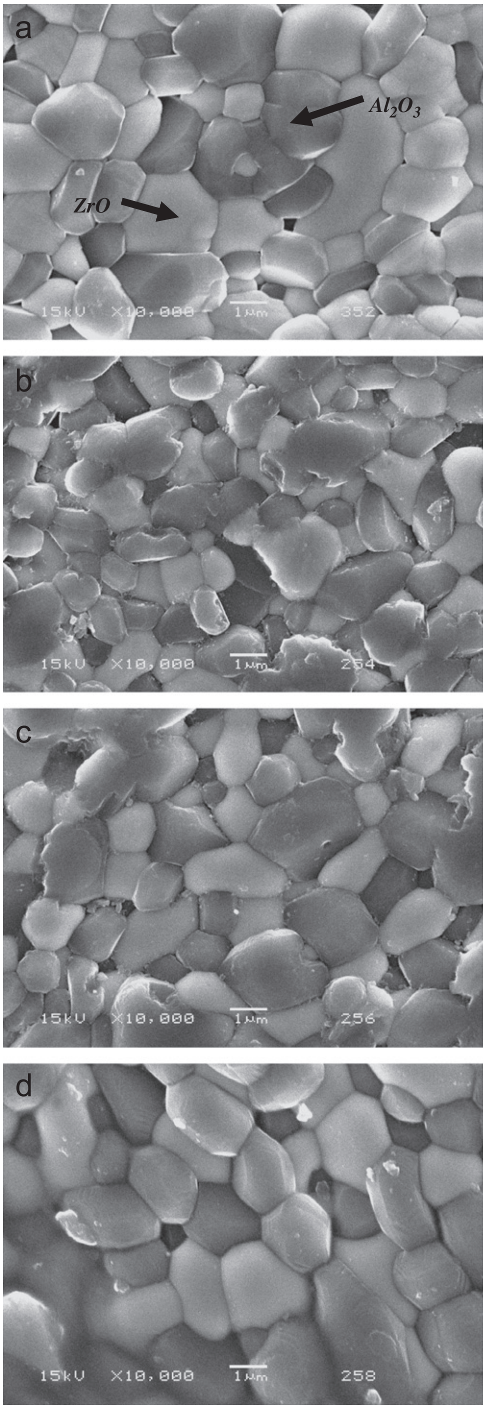

Fig. 2. SEM micrographs of the Al_2O_3–35 mol% ZrO_2 ceramics with different Y_2O_3: (a) 2 wt%, (b) 4 wt%, (c) 6 wt% and (d) 8 wt%.

completely to m-ZrO_2 phase with increasing Y_2O_3. Many studies [1–4] showed that alumina could stabilize the cubic zirconia and the tetragonal zirconia if the Al_2O_3–ZrO_2- powders were prepared by the alkoxide route where the mixture of zirconium isopropoxide and aluminum isopropoxide was used, or by the aqueous coprecipitation route, starting with mixture of zirconium oxychloride and aluminum chloride.

Densities of the sintered samples were determined by using the Archimedes principle. Densities of between 4.87 g/cm^3 and 4.95 g/cm^3 were obtained. Table 1 contains the data on the densities and shrinkage of the Al_2O_3–35 mol% ZrO_2 ceramics with different Y_2O_3 contents. The maximum density was obtained in the samples of Al_2O_3–35 mol% ZrO_2 ceramics with 6 wt% Y_2O_3 added. Densities tend to increase with increasing concentrations of Y_2O_3. Moreover, linear shrinkage was shown to increase with Y_2O_3 contents, corresponding to densities. Fig. 2(a)–(d) shows the SEM micrograph of as-received Al_2O_3–35 mol% ZrO_2 ceramics with different Y_2O_3 contents, indicating typical microstructures. The microstructure of the sintered Al_2O_3–35 mol% ZrO_2 with Y_2O_3 added is presented in Fig. 2, where the white and gray phases are ZrO_2 and Al_2O_3, respectively (as arrow). Microstructural characteristics were observed, i.e., uniformly sized grains with well-packed and continuous grain structure. Almost no abnormal grain growth appeared. Supporting other work, the addition of ZrO_2 prevented the abnormal grain growth in alumina ceramics [10]. By applying the linear intercept method [11] to these SEM images, grain sizes were estimated for these samples as given in Table 1. It can be seen that Al_2O_3–35 mol% ZrO_2 ceramics with 2–4 wt% Y_2O_3 added exhibited average grain sizes range of 1.19–1.30 μm, while Al_2O_3–35 mol% ZrO_2 ceramics with 6–8 wt% Y_2O_3 added exhibited average grain sizes range of 1.65–1.95 μm. Comparing with the grain sizes of Al_2O_3–ZrO_2 ceramics [12] and grain Al_2O_3–35 mol% ZrO_2 ceramics with addition Y_2O_3, it is found that the grain of Al_2O_3–35 mol% ZrO_2 ceramics with addition of Y_2O_3 is smaller in size than grain sizes of Al_2O_3–ZrO_2 ceramics. Thus, the optimal content of Y_2O_3 is an important parameter for development of ceramic microstructures.

Corresponding EDX analysis and chemical compositions for some of these Al_2O_3–35 mol% ZrO_2 ceramics with different Y_2O_3 contents are shown in Fig. 3 and Table 2. It is seen that the Y concentration increases with increasing Y_2O_3 contents. Compositions and mechanical property relationships of Al_2O_3–35 mol% ZrO_2 ceramics with different Y_2O_3 contents were investigated.

Table 3 shows the relationship between the crystallographic data and mechanical properties of Al_2O_3–35 mol% ZrO_2 ceramics with differentY_2O_3 contents.

Table 2
Chemical compositions of Al_2O_3–35 mol% ZrO_2 ceramics with different Y_2O_3 contents from EDX.

Contents of Y_2O_3 (wt%)	Composition (at%)			
	Al (K)	Zr (K)	Y (K)	O (K)
2	36.02	17.12	1.62	50.69
4	30.58	15.41	2.58	52.86
6	29.15	17.54	4.15	46.50
8	31.81	17.12	1.62	50.69

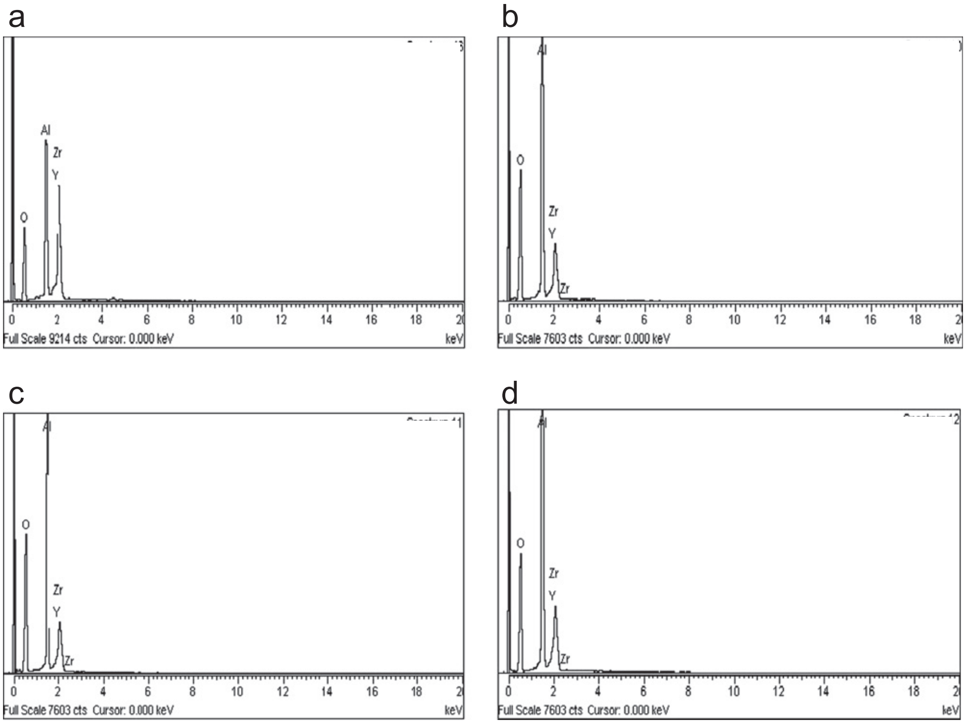

Fig. 3. EDX analyses of Al_2O_3–35 mol% ZrO_2 ceramics with various Y_2O_3 contents:(a) 2 wt%, (b) 4 wt%, (c) 6 wt% and (d) 8 wt%.

Table 3
Parameters obtained from Rietveld analysis and mechanical properties of Al_2O_3–35 mol% ZrO_2 ceramics as a function of different Y_2O_3 concentrations.

Contents of Y_2O_3 (wt%)	Phase present	Lattice parameter				Phase content (%)	Hardness HV (MPa)	Hardness Knoop (MPa)	Fracture toughness (MPa $m^{1/2}$)
		a (nm)	*b* (nm)	*c* (nm)	*β* (deg.)				
2	α-Al_2O_3	0.4058	0.4058	0.7091	98.10	68.4	11.30	10.83	2.53
	m-ZrO	0.5120	0.5040	0.5260		23.1			
	t-ZrO_2	0.3413	0.3413	0.3500		5.27			
4	α-Al_2O_3	0.3759	0.3759	0.7503	96.10	64.4	12.51	16.28	4.87
	m-ZrO_2	0.5672	0.5218	0.5438		16.3			
	t-ZrO_2	0.4038	0.4038	0.7401		15.4			
6	α-Al_2O_3	0.4754	0.4754	0.7892	98.88	66.6	11.45	15.86	3.46
	m-ZrO_2	0.5900	0.5742	0.5841		13.3			
	t-ZrO_2	0.5040	0.5040	0.5910		14.1			
8	α-Al_2O_3	0.3756	0.3756	0.6092	98.00	64.6	10.46	16.08	3.52
	m-ZrO_2	0.4010	0.4260	0.4020		18.1			
	t-ZrO_2	0.4670	0.4670	0.5300		9.3			

The sample with 4 wt% Y_2O_3 added had high fraction of tetragonal phase; it was found that hardness increases with increasing Y_2O_3 concentration up to 4 wt% and it decreased with high contents of Y_2O_3 (6 and 8 wt%). Moreover, these results indicate that increase in hardness resulted from small grain size of Al_2O_3–35 mol% ZrO_2 ceramics with different Y_2O_3 contents. The optimal Y_2O_3 addition (4 wt%) inhibits grain growth of ceramics giving rise to homogeneous and dense ceramics. The fracture toughness of Al_2O_3–35 mol% ZrO_2 ceramics as a function of different Y_2O_3 concentrations is shown in Table 3. The toughness of all the compositions is higher than that of Al_2O_3–ZrO_2 ceramics. The highest value of fracture toughness is from Al_2O_3–35 mol% ZrO_2 ceramics with 4 wt% Y_2O_3 added, which corresponds to a high ratio of tetragonal phase as shown in Fig. 1 by XRD and an optimal microstructure.

4. Conclusions

The key parameter that controlled the phase formation, microstructure, and mechanical properties here would be the addition of Y_2O_3. Al_2O_3–35 mol% ZrO_2 ceramics with 4 wt% Y_2O_3 added have the average grain size of 1.19 μm which showed the highest microhardness and fracture toughness. The samples had phase compositions of α-alumina combined with tetragonal zirconia phase and Y_2O_3 phase.

Acknowledgments

This work was supported by the Thailand Research Fund (TRF), the Commission on Higher Education (CHE) and Faculty of Science, Mahasarakham University (MSU), Thailand. The authors are grateful for financial support by Faculty of Science (MSU) to AMEC-8.

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