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EFFECTS OF Al_2O_3 CONTENT ON THE DENSIFICATION AND MECHANICAL PROPERTIES OF ZrO_2-Al_2O_3 NANOCOMPOSITES

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Abstract- *The influence of Al2O3 addition on densification and mechanical properties of tetragonal 3 mol% Y2O3-ZrO2 ceramics was investigated. The ZrO2-Al2O3 ceramic composites containing 35, 40, 45 and 50 wt% Al2O3 were prepared by means of uniaxial compacting at 210 MPa and sintered at 1550 °C in air for 3 h. The bulk density, porosity, microhardness and average grain size of ZrO2-Al2O3 composites were investigated as a function of Al2O3 contents. The bulk density and porosity of the specimens is significantly decreased, whereas average grain size increased with increasing the Al2O3 concentration. However, the maximum hardness of 14.02 GPa was obtained for higher Al2O3 additions.*

Keywords: Nanocomposite, Scanning electron microscopy, X-ray diffraction, Microhardness

1. Introduction

Zirconia has been one of the most important ceramic materials for well over a century and due to discovery of transformation toughening mechanisms in 1975s resulted in improvement of its mechanical properties [1-2]. Presently the research efforts appeared to be more focused on 3 mol% Y_2O_3 partially stabilized tetragonal zirconia polycrystals (3Y-ZrO_2) exhibited the best performance including high toughness, strength and contained only the tetragonal phase at room temperature [3-5].

However, its poor hardness than other competitive ceramics is ever since main issue. In order to enhance their mechanical properties it may be dispersed as reinforcement in various composite matrices, which intensely depends on the particle size, and specific surface area of the nanopowders, as well as relative distribution of ZrO_2 and Al_2O_3 in the matrix [6]. Therefore, ZrO_2-Al_2O_3 ceramic systems were thus developed to further improve the mechanical properties of zirconia. This material combined the hardness and wear resistance of alumina with the fracture toughness and bending strength of zirconia [7, 8]. Moreover, the addition of alumina helped to suppress the propagation of phase transformation into the bulk of the material and increased the hydrothermal stability of the tetragonal phase [7, 9].

In the present work attention is concentrated on 3 mol% Y_2O_3 partially stabilized tetragonal zirconia polycrystals because they exhibited very good results on different properties like, toughness, strength and contained only the tetragonal phase at room temperature. The influence of the Al_2O_3 contents of ZrO_2-Al_2O_3 nanopowders on the porosity, relative density, average grain size, and Vicker's microhardness of the ZrO_2-Al_2O_3 ceramic composites were investigated.

2. Experimental procedure

2.1 Processing

As starting powders in this study of Al_2O_3 (99.9% purity, average particle size ~150 nm) and 3 mol% Y_2O_3 stabilized ZrO_2 (designed YSZ, purity 99.9%, average particle size 30-60 nm) were used for experiments. Four compositions were prepared varying the Al_2O_3 content in the ZrO_2 matrix from 35, 40, 45 and 50 wt%. The composite powders were prepared by wet ball-milling for 24 hours in ethanol (C_2H_5OH), using high purity zirconia balls (10 mm diameter), as medium to obtain a homogeneous mixture. After milling, the powder mixtures were dried at 90 °C for 21 h to remove the ethanol. The powder was crushed gently with the help of mortar and pestle to get proper mixing by adding a few drops 5 wt% polyvinyl alcohol (PVA) solution as a binder and dried 110 °C for 6 h to eliminate the water content of the binder solution. The sieved powders were then uniaxially pressed at 210 MPa pressure and green compacts sintered at 1550 °C for 3 h in air. The heating rate varied according to the following program: 8 °C/min up to 600 °C for 3 h; 5 °C/min up to 1400 °C; and 3 °C/min until the final temperature for 3 h. The cooling rate was 5 °C/min down to until the inertia of the furnace prevailed.

2.2 Characterization

The densities of the sintered samples were measured by Archimedes' method using distilled water

as the immersion medium in accordance with ASTM: C 20-00 [10]. The relative density was calculated using a theoretical density of 3.99 gm/cm^3 for Al_2O_3, 6.09 gm/cm^3 for 3Y-ZrO_2, respectively. The crystalline phases of the composite specimens was conducted via a D8 ADVANCE X-ray diffractometer (Bruker AXS, Karlsruhe, Germany) with CuKα radiation in a continuous mode from 2θ range 20-90° with a scanning rate of 2 °/min. The voltage and current were set at 40 kV and 40 mA, respectively. The average grain size was obtained by analyzing scanning electron microscopy (SEM) images of randomly selected areas of specimens using the linear intercept method. Microhardness of the composites was evaluated at ambient temperature by a Vickers microhardness indenter (HMV-2, Japan) with an indent load of 19.614 N with 6 sec dwell time using ten indents for each composite in accordance with ASTM C1327-99 [11]. The size of the impression was measured with the aid of a calibrated microscope. After the diagonal length measurement, the values of the Vickers hardness (in GPa) were calculated by using the following equation:

$$H_V = 0.0018544\left(\frac{p}{d^2}\right) \tag{1}$$

where, p being the applied indentation load and d is the arithmetic mean of the two diagonal length.

3. Result and discussion

The Bragg diffraction condition for X-rays is expressed by Bragg diffraction law, denoted by $2d\sin\theta = n\lambda$. The X-ray diffraction patterns of the ZrO_2-Al_2O_3 samples containing different percentage of Al_2O_3 contents sintered at 1550 °C for 3 h are shown in Fig. 1. X-ray diffraction analysis of the samples indicated no m-ZrO_2 phase is present as characterized by diffraction peak at 2θ = 28° and 31°, only t-ZrO_2 phase has been observed, showing that the m-ZrO_2 phase content in the starting powder has been completely transformed, and indicating the complete stabilization of the tetragonal phase during cooling. The peak intensities of the Al_2O_3 increased due to increasing amounts in the ZrO_2 matrix. The result of bulk density and apparent porosity of the composite samples are shown in Fig. 2 and Fig. 3, respectively. It is noted that the composites demonstrated reduced porosity level with increasing Al_2O_3 contents, thus Al_2O_3 particles influence densification. Fig. 4 presents the results of relative density as a function of Al_2O_3 contents. The result indicates that the relative density increases by increasing the Al_2O_3 content. Earlier, similar results were obtained in ZrO_2/Al_2O_3 composites an increasing in relative density when alumina content increases [12].

All the sintered composite materials are characterized by a very high density ranging from 97.86% to 98.58% of theoretical and for the composition with the addition of 50 wt% Al_2O_3 content the relative density (> 98%) improving the mechanical properties and increasing the reliability and therefore leading to materials with improved properties for structural applications.
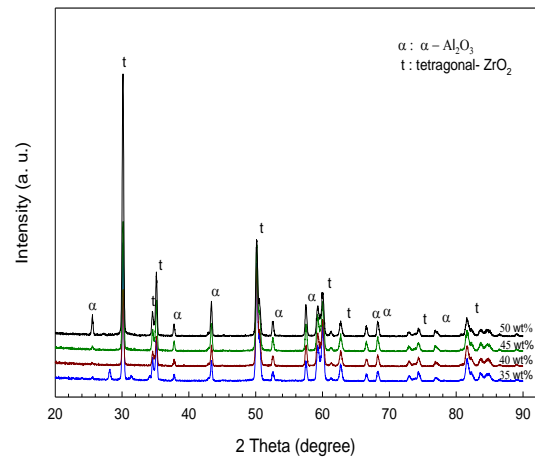

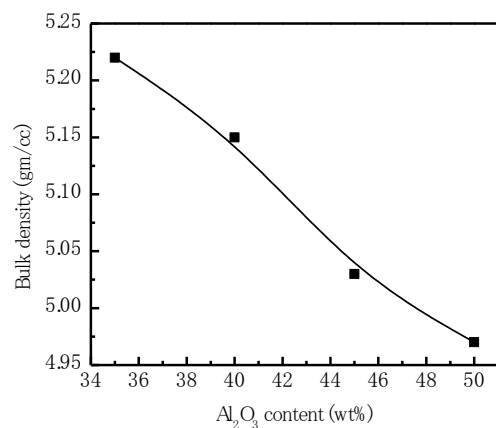Fig. 1 X-ray diffraction patterns of the ZrO_2-Al_2O_3 composites, sintered at 1550 °C for 3 h.
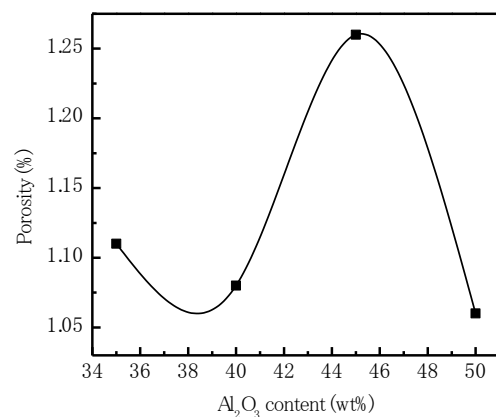

Fig. 2 Bulk density of the ZrO_2-Al_2O_3 composites

Fig. 3 Porosity of the ZrO_2-Al_2O_3 composites

Figure 5 represents the SEM photographs of fracture surface of the ZrO_2-Al_2O_3 composites containing different percentage of Al_2O_3 contents sintered at 1550 °C for 3 h. The presence of two distinct phases Al_2O_3 (dark phase) and ZrO_2 (white phase) can be clearly seen, an increasing amount of Al_2O_3 grains and agglomerate with increasing Al_2O_3 contents in the composite materials.
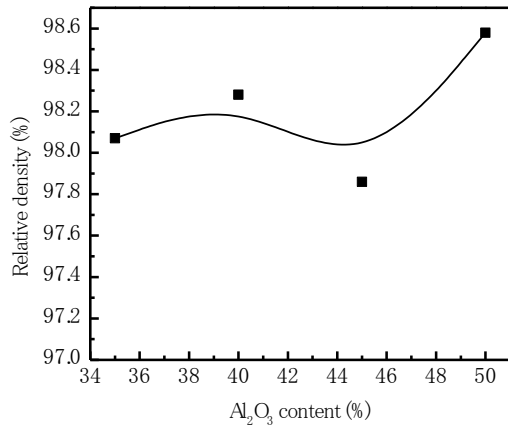
Fig. 4 Relative density of the ZrO$_2$-Al$_2$O$_3$ composites

The hardness of ceramic materials is generally affected by the intrinsic deformability of the ceramic and micro- structural parameters such as multiphases, grain size and orientation, porosity as well as boundary constitution [13]. Suzuki et al. [14] reported that Vickers hardness is correlated with porosity in the former sintering stage while in the latter sintering stage is correlated with grain size. From Fig. 6 it is expected that the composites (55wt% SZ-3Y + 45wt% Al$_2$O$_3$) would have the lowest hardness as a consequence of its lowest sintered density. Lin and Duh [15] reported similar appreciations in Ce-Y-TZP, where hardness results have been related with porosity according to the equations described by Mccolom [16] for hardness. Hong et al. [17] and Lee et al. [18] reported generally the Vickers hardness of the YSZ/Al$_2$O$_3$ ceramics is 12-14 GPa, which is in good agreement with the result obtained a hardness value of 14.02 GPa for (50wt% SZ-3Y + 50wt% Al$_2$O$_3$) composites in our present work.
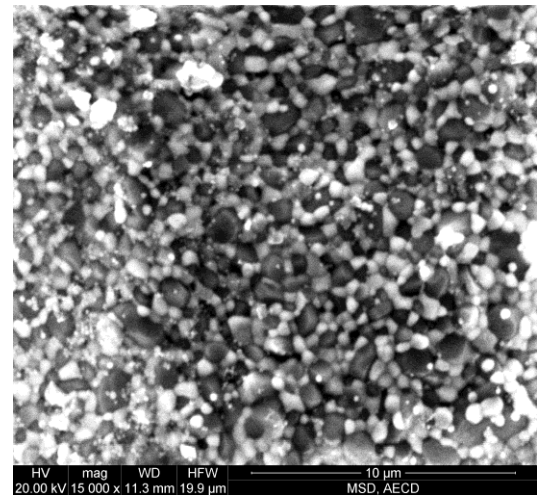

(b)

(c)

(a)
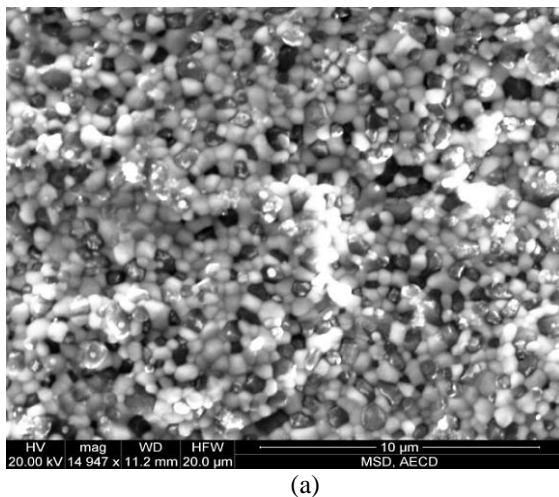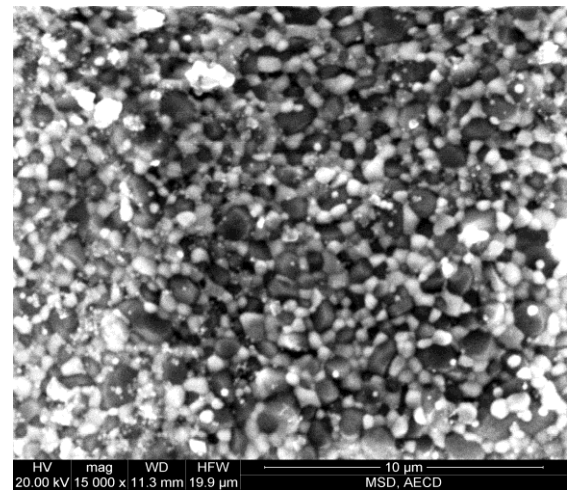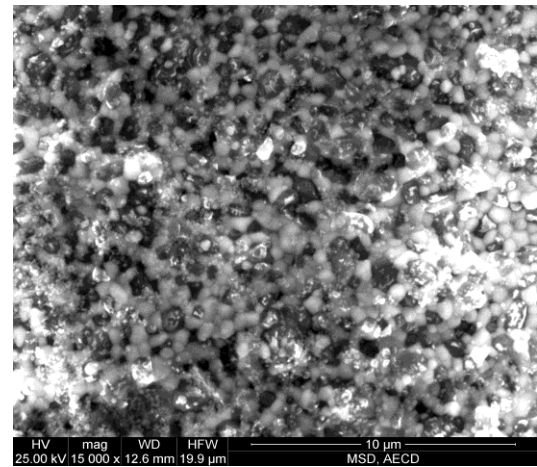
(d)

Fig. 5 SEM images of the ZrO$_2$-Al$_2$O$_3$ composites; (a) 35 wt% Al$_2$O$_3$ (b) 40 wt% Al$_2$O$_3$ (c) 45 wt% Al$_2$O$_3$ and (d) 50 wt% Al$_2$O$_3$ content.
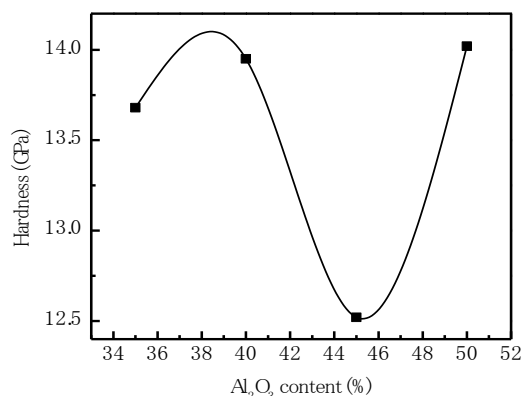
Fig. 6 Hardness of the ZrO_2-Al_2O_3 composites

4. Conclusion

ZrO_2-Al_2O_3 nanocomposites were prepared by varying the Al_2O_3 content in the ZrO_2 matrix from 35wt% in the step of 5 up to 50wt%. The prepared powder was uniaxially pressed at a pressure 210 MPa and green compacts are sintered at 1550 °C for 3 h in air.

In all sintered materials only the tetragonal ZrO_2 phase has been observed, indicating the complete stabilization of the tetragonal phase during cooling. Al_2O_3 had no influence on the phase transformation but influenced the peak intensities and grain growth. Increasing the Al_2O_3 content results in a decrease in porosity and an increase in relative density of the sintered ceramic composites. The Vicker's microhardness increases from 12.52 GPa to 14.02 GPa for the addition of 50wt% Al_2O_3 content due to lower porosity and higher density.

5. Acknowledgment

6. Reference

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7. Nomenclature

Symbol	Meaning	Unit
H_V	Vickers microhardness	GPa
SEM	Scanning Electron Microscopy	No unit
YSZ	Yttria-Stablized-Zirconia	No unit