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Effect of sintering temperature on properties of Al_2O_3 whisker reinforced 3 mol% Y_2O_3 stabilized tetragonal ZrO_2 (TZ-3Y) nanocomposites

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ABSTRACT

In present study, effect of sintering temperature on density and hardness of 3 mol% yttria-stabilized tetragonal zirconia (referred to as TZ-3Y) composite reinforced with alumina whiskers (5, 10, 15 and 20 wt.%) has been studied. Initially, Ammonium Aluminum Carbonate Hydroxide (AACH) whiskers were added in TZ-3Y composite and transformed into alumina during sintering performed at different temperatures i.e. 1400, 1500 and 1650 °C. Results revealed that for all sintering temperatures, with increase in whisker concentration, sintered density decreased and hardness increased conversely. Maximum hardness of 14.47 GPa was achieved with 10 wt.% whiskers addition when sintered at 1500 °C. However, with addition of CTAB (1 wt.%) as deflocculating agent the hardness was further improved to 15.11 GPa. While sintering at 1650 °C a decrease in hardness was observed. It was mainly due to high temperature morphological change of whiskers i.e. transformation of whiskers into alumina rich grains.

1. Introduction

For use as bio-ceramics the 3 mol% Y_2O_3 stabilized tetragonal ZrO_2 (TZ-3Y) is unique because of its excellent strength and toughness at room temperature [1,2]. However, to increase the phase stability [3] and hardness of tetragonal zirconia [4], a composite of zirconia–alumina is recommended [5,6].

Other factors which may affect the mechanical properties of the composite are the shape or morphology of the reinforcement. The use of ceramic whiskers as reinforcement in dental resin has proved a useful approach for improving the mechanical properties of the composites [7]. The whiskers are single crystals and posses high degree of structural perfection and superior mechanical properties. By using ceramic whiskers as reinforcement, the mechanical properties of the resulting composite improves due to the crack bridging, crack deflection and crack bowing [8–11] effects. Because of the positive effect of whiskers on the mechanical properties of the composite many researchers [12–17] have reported on the synthesis and characterization of SiC whisker reinforced zirconia composite. However the non-oxide ceramic whiskers may oxidize or react with the matrix at high temperature during processing or operational life. Therefore, the whiskers made of oxide ceramics like alumina are ideal for use as reinforcement in the Zirconia based ceramics. In spite of the advantages of using alumina whisker as reinforcement in the Zirconia ceramics, very limited work [18,19] has been reported on alumina whisker reinforced TZ-3Y composite. The main hindrance in the use of alumina whiskers as composite reinforcement is its high production cost. However, recently, Li et al. [20] have reported a low cost hydrothermal synthesis route for the production of alumina whiskers. By using this synthesis technique the AACH whiskers were produced and used for the formation of the composite, these whiskers were then transformed in situ to Alumina whiskers during sintering.

For better mechanical properties of the composite the parameters such as density, grain size, and porosity should be controlled. Optimum sintering temperature is of great importance because it affects density and grain size; both of which directly influence the mechanical properties of the composite.

In the present work, the effect of different sintering temperatures on hardness and density of the in situ transformed alumina whisker reinforced tetragonal Zirconia ceramics composite has been studied for the first time. The change in hardness and density of the composite was correlated with whisker morphology. Moreover the beneficial use of CTAB on the dispersion of alumina reinforcement in these composites has also been investigated. Finally, on the basis of the results obtained, an optimum sintering temperature for this type of composite has been proposed.

2. Experimental procedures

For the present study, alumina whiskers were transformed in situ from Ammonium Aluminum Carbonate Hydroxide (AACH) whiskers, which were synthesized through hydrothermal technique as reported by Li et al. [20] by using high pressure reactor

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(Limbo-350). The precursor materials were aluminum nitrate and urea and the process parameters were 120 °C for 24 h.

The hydrothermally synthesized AACH whiskers, 3 mol% Y_2O_3 stabilized ZrO_2 (hereafter abbreviated as TZ-3Y; Make: Terio, China; purity >99.99%; Average particle size ~0.2 μm) and cetyl trimethyl ammonium bromide (CTAB) were used as starting materials. The TZ-3Y and AACH whiskers were dispersed separately in analytical grade ethanol (with or without 1 wt.% CTAB) and sonicated for 30 min for breakage of agglomerates. Both the suspensions were mixed together and kept at 60 °C while stirring until nearly all the ethanol was evaporated. Samples with varying amounts of AACH corresponding to 5, 10, 15 and 20 wt.% Al_2O_3 were prepared. Composites with higher concentration of whisker (i.e. greater than 20 wt.%) were not included in this study because the higher concentration produces agglomeration which deteriorates the composite properties [18].

To remove the volatile products of AACH and CTAB the samples without CTAB were calcined at 400 °C while the samples containing CTAB were calcined at 650 °C. The pellets of diameter 10 mm were prepared by using uniaxial hydraulic press at a load of 5×10^3 kg for 2 min. The green pellets were sintered at three different temperatures, i.e. 1400, 1500 and 1650 °C in a muffle type (Carbolite HTF-18/8) furnace at heating and cooling rates of 5 °C/min.

The density of sintered pellets was measured in absolutely pure ethanol through Archimedes principle by using high accuracy weighing balance. The morphology of the whisker and the surface of the sintered/fractured pellets was observed by using scanning electron microscope (JEOL JSM-6490LA) at an accelerating voltage of 2–20 kV. The average grain size was measured through linear intercept method. The chemical composition and phase purity were determined through Energy Dispersive X-ray Spectroscopy (EDS or EDX) installed with the SEM.

The Vicker hardness of sintered pellets was measured with the help of Microhardness tester (Series 401MVD WOLPERT Group). For each composition four samples were used while approximately 10–15 indents were made for each measurement and the average hardness was calculated. The separation between the neighboring indents was kept more than four diagonal lengths of indented impression as per ASTM C 1327-99 standard [21]. For Vicker micro-hardness test, a load of 1 kg was applied for 15 s and dimension of the formed indent was observed by using installed optical microscope.

3. Results and discussion

3.1. Microstructure analysis

Fig. 1a–c shows the SEM micrographs of the hydrothermally synthesized AACH whiskers. These whiskers appear to be agglomerated in the form of clusters of diameter 500 nm and length 5–6 μm (Fig. 1b). However on close observation, it was revealed that the diameter of the individual whisker was approximately 100–200 nm (Fig. 1c). Probably, the whiskers formed during synthesis were joined sidewise in the form of thick clusters due to Van der Waal's and/or hydrogen bonding [22]. The average diameter of these nano-whiskers was calculated as 150 nm.

Fig. 2a–c shows the SEM micrographs of the surface of composite samples sintered at three different temperatures (1400, 1500, 1650 °C). From these micrographs the average grain size (measured through Lineal Intercept method) is given in Table 1. As expected with increase in the sintering temperature the grain size increases.

Unlike the samples sintered at 1400 °C the composite samples sintered at 1500 °C and 1650 °C showed equiaxed grains with well developed grain boundaries and minimum residual porosity. The reason for grain growth was the high sintering temperature, which provided the activation energy required for grain boundary migration (which minimizes the grain boundary area and Gibbs free energy) through enhanced high temperature diffusion phenomenon [23].

3.2. Effect of sintering temperature and whisker concentration on density

Fig. 3a and b shows the effect of alumina whiskers concentration (5, 10, 15 and 20 wt.%) and sintering temperature on the relative density of the composite. Three main observations can be inferred from these graphs (Fig. 3a and b). First, with increase of whisker content the sintered density decreases. The reason is, at higher whiskers concentration, the whiskers form hard agglomerates which resist complete shrinkage of the composite during sintering and produce porosity which result in decreasing the sintered density.

The second observation that can be drawn from comparison of density vs alumina concentration graphs for samples with/without CTAB addition (Fig. 3a and b) is, with the addition of deflocculating agent i.e. 1 wt.% CTAB the density improves for all the compositions of the composite due to better dispersion of alumina whiskers in the matrix material. If the whiskers are not dispersed and remain in the form of agglomerate, then during sintering due to difference in shrinkage rate (of agglomerate and matrix), comparatively large voids will produce at the interface and the composite density will decrease [24].

The third observation that can be made from Fig. 3a and b is that for same whisker concentration the density increases with increasing the sintering temperature. It can also be correlated with the micrographs of sintered surface of the composite as shown in Fig. 2a–c, where comparatively large pores can be observed on the samples sintered at 1400 °C (as encircled on the micrograph). However, for the samples sintered at 1500 °C and 1650 °C very small pores at the triple and quadra-junctions of the grains are present (indicated with arrows). Moreover, the numbers of triple junction pores is more in the samples sintered at higher temperature (i.e. 1650 °C). The reason is, the quadra-junction pores is comparatively unstable configuration [25] and with the increase

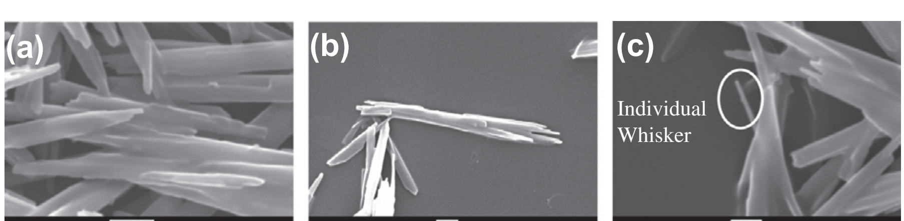

Fig. 1. SEM micrographs of AACH whiskers prepared through hydrothermal synthesis (a) cluster of multiple whiskers (b) longitudinal image of a whisker cluster, (c) an individual whisker marked.