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Effect of Bi_2O_3 on phase formation and microstructure evolution of mullite ceramics from mechanochemically activated oxide mixtures

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ABSTRACT

Mullite ceramics with hollow whisker structure have been synthesized firstly through ordinary sintering process. The effects of Bi_2O_3 and processing, on mullitization behavior and morphology development of mullite ceramics, derived from the mechanochemically activated mixture of Al_2O_3 and SiO_2, were investigated in this paper. When the content of Bi_2O_3 was less than 10 mol%, the mullite grains show a short rod-like morphology, without the formation of whisker. As the content of Bi_2O_3 was increased to more than 10 mol%, the formation temperature of mullite was decreased from 1400 °C to 1100 °C. After sintering at 1400 °C, well-developed mullite whiskers with hollow structure were formed. The formation process and growth mechanism of hollow structural whiskers in mullite ceramic doped with high content of Bi_2O_3 were discussed in detail.

1. Introduction

As the only stable crystalline phase in the binary system of Al_2O_3-SiO_2, mullite ($3Al_2O_3{\cdot}2SiO_2$) has important applications in both traditional and advanced ceramics [1,2]. Owing to their excellent thermal stability, high melting point, low thermal expansion coefficient, good thermal shock resistance and high creep resistance, mullite based ceramics have been considered as the key materials in traditional kiln and refractory industries [3]. The low dielectric constant and excellent electrical insulation make mullite ceramics a promising candidate for potential applications in microelectronics industry, such as integrated circuit, electronic packing [2], thermistor [4] and antennas for signal absorption and amplification [5]. In addition, recent studies indicated that mullite could be used for optical applications, due to its small birefringence and excellent infrared transmission capability [6–8].

Mullite crystals are prone to grow with acicular morphologies [9]. Through composition adjustment and process optimization, the length of mullite whisker can reach 20 μm and the aspect ratio can be higher than 60 [10]. Due to the interlocking behavior of mullite whiskers during growth [11], mullite ceramics could have low density or high porosity, while maintaining sufficiently high mechanical strength, which is a very valuable property that is hardly observed in other porous materials. However, the processing of mullite ceramics usually requires very high temperatures (≥ 1700 °C). By using modification techniques, such as the use of additives, the formation temperature of mullite ceramics can be decreased to ≤ 1400 °C. In this case, non-mullite phases are often formed during the sintering process, which severely limited the anisotropic growth of mullite crystals, thus leading to the formation of granular or short rod grains. Among various additives, AlF_3 [10,12,13] and $AlCl_3$ [14] are regarded as the most effective catalysts to synthesize mullite whiskers with high aspect ratios, in which the fluid phase diffusion is considered to be the predominant mechanism for mass transport during the anisotropic growth of mullite crystals [15]. Unfortunately, the high-temperature steams formed due to the presence of these additives at high temperatures are carcinogens [16], which not only causes potential body injuries to the personnel

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during production but also leads to serious environmental pollution.

Our previous studies have shown that high performance mullite ceramics can be obtained by using high-energy ball milling (mechanochemical activation) without the use of any additives [17,18]. Further studies showed that the formation temperature of whisker-structured mullite ceramics can be decreased by doping other oxides coupled with the mechanochemical activation of the raw materials [19,20]. Bi_2O_3 has a low melting temperature (~ 825 °C) and usually used as an additive to reduce the sintering temperature of functional ceramics [21]. In this work, we studied the effects of Bi_2O_3 on phase formation and microstructure evolution of mullite ceramics from mechanochemically activated oxide mixtures. Meanwhile, the formation process and growth mechanism of hollow structural whiskers in mullite ceramic doped with high content of Bi_2O_3 were discussed in detail.

2. Experimental

The mullite ceramic samples were prepared by using traditional high temperature sintering method. Commercial SiO_2 (Laboratory reagent, BDH Chemicals Ltd Poole, England), Al_2O_3 (99+% purity, Aldrich Chemical Company Inc., USA) and Bi_2O_3 (98+% purity, Aldrich Chemical Company Inc., USA) powders were used as the starting materials, with nominal compositions of $(3Al_2O_3{\cdot}2SiO_2)_{1-x}(Bi_2O_3)_x$ (x = 0.05, 0.10 and 0.20). The milling operation was carried out by using a Retsch PM400 type planetary ball milling system in air at room temperature for 10 h. A 250 ml stainless steel vial and 100 stainless steel balls with diameter of 10 mm were used as the milling media. In all cases, powder mixture of about 20 g was milled in dry condition without the use of any liquid, while the milling speed was set to be 200 rpm.

The milled powders were then pressed uniaxially into 10 mm diameter pellets at a pressure of 50 MPa. The green pellets were sintered with a Carbolite RHF 1600 type furnace in air for 4 h at temperatures from 1000 °C to 1400 °C, at both heating and cooling rates of 10 °C/min.

Phase compositions of the original raw materials, the milled mixtures, and the sintered samples were characterized by using a Rigaku ultima+ type diffractometer (XRD) with Cu/Kα radiation. Surface morphology of the milled mixture and the sintered samples, without surface chemical treatment, was examined by using a JEOL JSM-6340F type field emission scanning electronic microscope (FESEM). Particle sizes of the milled powders were measured by using a SALD-2300 type laser diffraction particle size analyzer (Shimadzu, Japan).

3. Results and discussion

Fig. 1 shows XRD patterns of the Al_2O_3-SiO_2 mixtures doped with different contents of Bi_2O_3 before and after milling. A broad peak at about 26° suggests that the main phase of the milled powders is amorphous structure (Fig. 1a). Some weak diffraction peaks can also be observed in the samples, which is attributed to Al_2O_3. Fig. 1b shows XRD patterns of the raw materials used for the mixtures. It can be seen that all the original raw materials are in typical crystalline state. The strong contrast in XRD patterns of the samples before and after milling indicates that the high-energy milling resulted in amorphization of SiO_2 and Bi_2O_3. In addition, the diffraction peaks of Al_2O_3 are broadened obviously after milling, implying refinement of the powders. Fig. 2 shows SEM image and the corresponding particle size distribution curve of the representative milled mixture. Average particle size of the milled mixture is about 65 nm. All these results reveal that Bi_2O_3 and SiO_2 were amorphorized in the milled mixtures and the powders were refined to nanosized scale. The nanosized powders are undoubtedly reactive in terms of the formation and growth of mullite crystals.

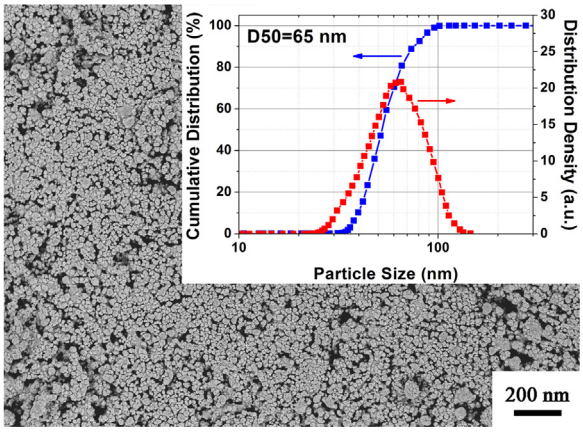

Fig. 2. SEM image of the milled mixture containing 5 mol% Bi_2O_3, with the inner to be the corresponding particle size distribution curve.

Fig. 3 shows XRD patterns of ceramic samples with 5 mol% Bi_2O_3 sintered at different temperatures for 4 h, while Fig. 4 shows XRD patterns of the samples with different contents of Bi_2O_3 sintered at 1100 °C. When the sintering temperatures are in the range of 1000–1100 °C, the main crystal phases are SiO_2 (cristobalite) and Al_2O_3, no characteristic diffraction peaks of Bi_2O_3 or mullite phase are observed in the sample containing 5 mol% Bi_2O_3. Theoretically, the formation temperature of mullite from the mixture of Al_2O_3 and SiO_2 is higher than 1400 °C [22,23], so these sintering temperatures might not be high enough to overcome the activation energy of mullite phase formation. The absence of Bi_2O_3 phase in the XRD patterns mainly

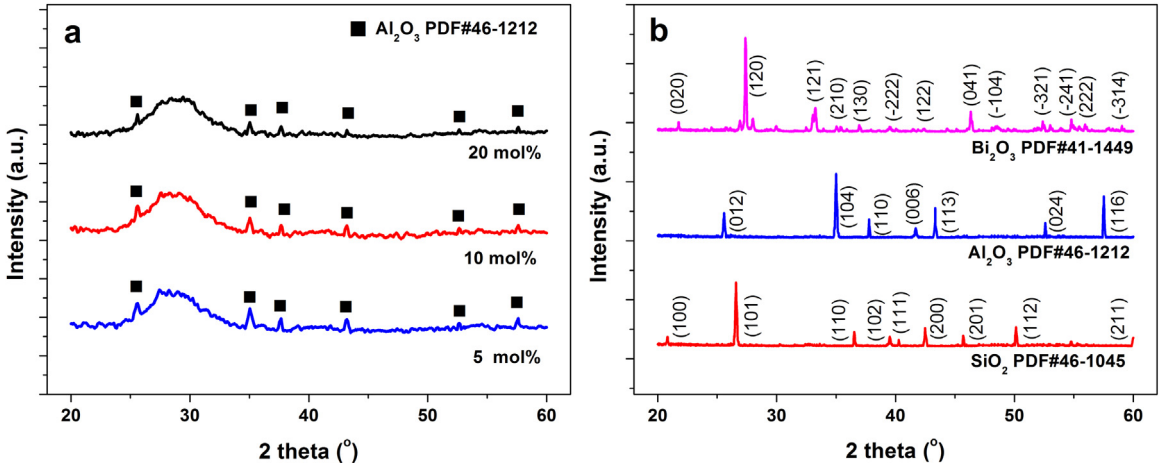

Fig. 1. XRD patterns of the milled mixtures doped with different contents of Bi_2O_3 (a) and the raw materials used in the mixtures (b).