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Microstructure development and phase evolution of alumina–mullite nanocomposite

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Abstract

In this work, the alumina–mullite composite was prepared using sol–gel derived alumina composite nanopowders. Results revealed the intragranular mullite was embedded in the alumina grain and the intergranular mullite was embedded on the grain boundary. Accordingly, the intragranular mullites (average grain size, 0.3 μm) were smaller than the intergranular mullites (average grain size, 0.5 μm). Moreover, the alumina grains (average grain size, 1.0 μm) are larger than the mullites. Meanwhile, the mullites showed positive results in the prevention of the alumina grains growth, known as the Zener law behavior, and the retardation of densification. The relative density of alumina–mullite that was sintered at 1650 °C for 2 h was obtained as 98.7%. After sintering at 1700 °C for 2 h, the mullite was decomposed.

Keywords: A. Sintering; B. Microstructure-final; D. Al_2O_3; [illegible]

1. Introduction

Alumina–mullite ceramics may have a high potential for armour and wear resistance applications [1,2]. The important applications of alumina–mullite composite are components and structures for gas turbine engine, burner tubes and heat shields for re-entry space vehicles [3]. Mullite in the alumina matrix reduces the Young's modulus and thermal expansion coefficient of the composite, leading to a better thermal shock resistance [4–6]. Meanwhile, mullite has low toughness and hardness [7]. Small mullite additions (5–15 vol%) allow desirable values of hardness and toughness of alumina to be maintained while reducing the Young's modulus below that of alumina, so that it is expected that the thermal shock behaviour will be improved [8].

A number of recent works have involved the addition of impurities in order to achieve better densification behaviour and, as a consequence, higher densities and better microstructures and mechanical properties [9,10]. Schehl et al. [11] presented a modified processing route which consists in the doping of a commercial high-purity alumina powder so that its microstructure is modified with such nanoparticles as zirconia and mullite, formed at the sintering stage. As a result, the grain boundaries of the high-purity alumina powder are modified by segregation of the secondary phases or by the formation of well-distributed zirconia and mullite nanoparticles. Thus it should be possible to tailor microstructures by means of secondary phases by referring to the corresponding phase equilibrium diagrams.

Very high homogeneous multicomponent ceramics and composite ceramics can be obtained via sol–gel method, since the synthesis temperature of this method is low [12].

In this work, alumina–mullite composites (5–15 vol%) were prepared using sol–gel derived alumina composite nanopowders, with the ultimate aim to investigate the positioning of mullite and its effect on the microstructure of the alumina–mullite composite. Meanwhile, the phase evolution of this composite was studied.

2. Experimental

The flowchart of the procedure is shown in Fig. 1. Homogeneous distribution of mullite in the matrix of alumina can be

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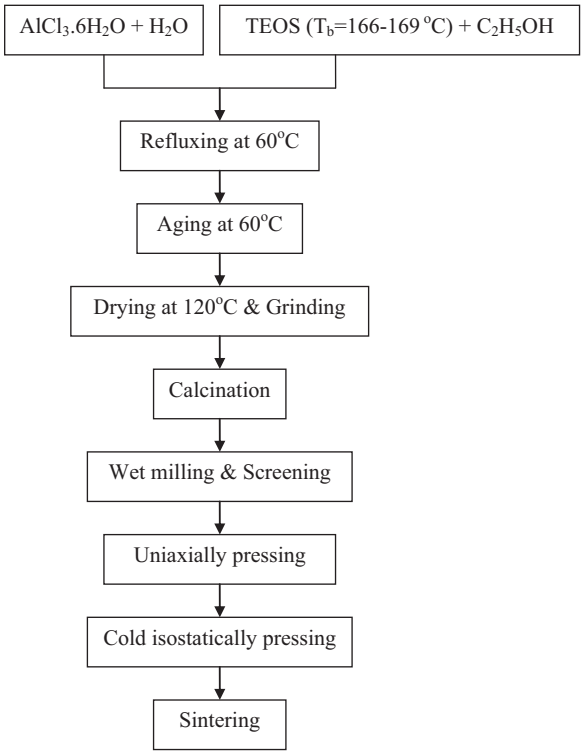

Fig. 1. The flow chart of the processing of alumina–mullite nanocomposite.

Table 1
Properties of the alumina—15 vol% mullite precursor calcined at 900 °C for 2 h.

BET surface area [m^2/g]	5.4 ± 4
Apparent density [g/cm^3]	3.646 ± 0.007
Mean particle size [nm]	–
Surface area of pores [m^2/g]	1
Total pore volume [cm^3/g]	0.50
Average pore diameter (4V/A by BET) [nm]	19.0

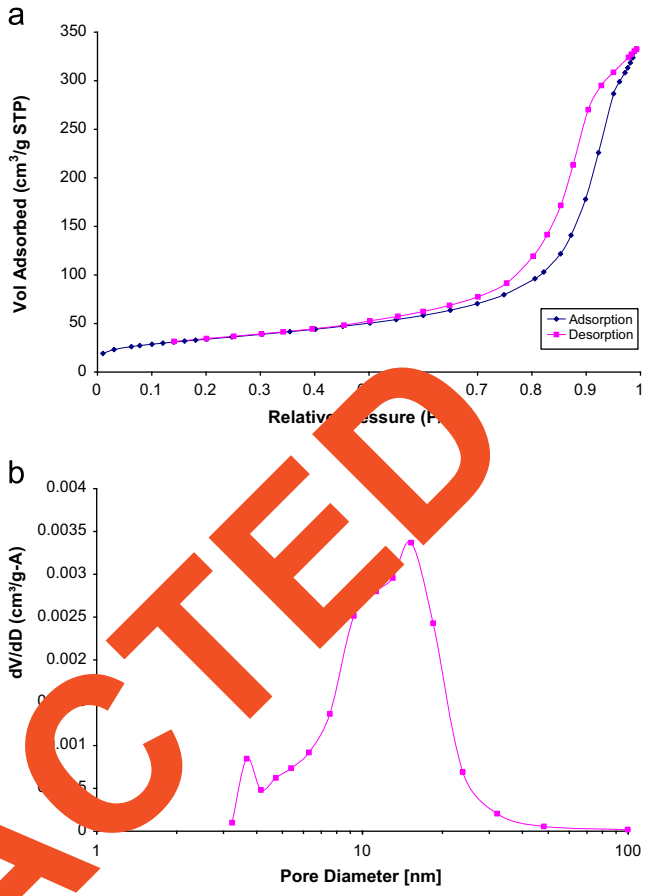

Fig. 2. (a) Nitrogen adsorption–desorption isotherm and (b) pore distribution of the alumina–mullite 15 vol% precursor calcined at 900 °C for 2 h.

obtained through sol–gel method. Some alumina composite nanopowders (Table 1) were synthesized by sol–gel method. Aluminum chloride hexahydrate (Merck 1027[illegible]4) was dissolved in distilled water and tetraethyl orthosilicate (Sigma-Aldrich 131903) was dissolved in absolute ethanol. Based on the stoichiometric ratio ($3Al_2O_3$ $2SiO_2$) and the desired volume percentage (0, 5, 10, 15 vol%) of mullite, the aqueous solution of salt with the required amount of the alcoholic solution of tetraethyl orthosilicate (TEOS) was refluxed at 60 °C for 24 h. After condensation, the gel was dried at 120 °C for 24 h and ground in an agate mortar. The precursors of the alumina and the alumina–mullite composites were calcined at 900 °C for 2 h and subsequently attrition-milled with high purity alumina balls and absolute ethanol for 1 h. After drying, the powders were sieved using an 80 μm mesh.

Fig. 2 presents isotherm patterns of nitrogen adsorption–desorption (a) and pore distribution of alumina-15 vol% mullite precursor calcined at 900 °C for 2 h (b). It can be seen that the powder exhibits the Type IIb group with the presence of mesopores. Shape of the curve and the hysteresis loop (H3-type) can justify the presence of aggregates containing platy particles (alumina shows this typical behavior) with the presence of non-rigid slit-shaped pores which were formed by the aggregates. These platy particles can be observed in the TEM micrograph (Fig. 3(a) and (b)) presents the smaller particles.

The powders were uniaxially pressed under 38 MPa. Then they were cold isostatically pressed (CIP) at 380 MPa for achieving greater uniformity of compaction, and finally sintered in air at 1650 °C for 2 h. Moreover, the alumina-15 vol% mullite specimens were sintered at 1300, 1500, 1650 and 1750 °C for 2 h to investigate their microstructure development.

X-ray diffraction (XRD) was carried out for phase characterization of the alumina and the alumina–mullite composites (5, 10 and 15 vol%) sintered at 1650 °C for 2 h. The phase evolution of the alumina-15 vol% mullite, thermally treated at different temperatures (400–1750 °C), was studied by XRD. The XRD patterns were recorded in the range of $10 < 2\theta < 80$ using Philips X-pert model with Cu $K\alpha$.

The microstructure of the sintered bodies was studied by a XL 30, Field Emission Environmental Scanning Electron Microscope (FEI-Philips) equipped with a Link Energy Dispersive X-ray system. The specimens for SEM were polished to 1 μm surface finish using diamond spray. Thereafter, they were thermally etched for 1 h at 100 °C below the sintering temperature. The average grain size was defined as the average apparent