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Fabrication and properties of mullite-bonded porous SiC membrane supports using bauxite as aluminum source

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Abstract

Mullite-bonded porous SiC membrane supports were fabricated at 1350–1500 °C in air from bauxite and graphite. The phase composition, open porosity, microstructure and mechanical strength as well as pore size distribution of membrane supports were systematically investigated. Due to the enhancement of necks at the contacting points growth by the addition of bauxite (30 wt%), a high three-point flexural strength of 85.2 MPa was achieved at an open porosity of 31.4%. Moreover, when 20 wt% C was added to the above composition, the strength of 36.6 MPa was achieved at an open porosity of 47.8%.

Keywords: D. Mullite; Bauxite; Membrane support; Reaction bonding; Porous SiC ceramic

1. Introduction

Widely used in high-temperature filters, catalysts and catalyst carriers, membranes and membrane supports, and refractory materials, etc. [1–6], porous SiC ceramics have attracted more and more attention in recent years due to their great mechanical and chemical stability, large internal surface area, low thermal expansion coefficient, and high thermal conductivity, etc. [7–9]. It is, however, difficult to fabricate SiC ceramics due to the strong covalent nature of the Si–C bond [10,11]. Although self-bonded SiC ceramics demonstrate excellent properties, they would be oxidized to SiO_2 during long-term operation at high temperature [12]. Therefore, the change of heat treatment environment or the addition of sintering aids has been proposed [8,13] so as to produce porous SiC-based ceramics at a lower processing temperature. She et al. [14] firstly developed an in-situ bonding technique which involves heating the SiC powder compact in air instead of inert atmosphere. As a result of oxidation, the SiC particles were bonded by SiO_2 or/and mullite, which exhibited good oxidation resistance.

As silica–alumina refractory and substrate materials, mullite ($3Al_2O_3 \cdot 2SiO_2$) has attracted much attention owing to its outstanding high-temperature properties [6,15]. It has a close thermal expansion match and good chemical compatibility with SiC. Ding et al [16,17] fabricated porous mullite-bonded SiC ceramics, which demonstrate superior high-temperature mechanical properties and thermal shock resistance [15–18]. Alumina is considered as, in general, aluminum source for the fabrication of mullite-bonded porous SiC ceramics [16–20]. Bauxite is the major source of starting materials for Al_2O_3-based products [21–23]. Bauxite, consisting mainly of Al_2O_3, is the appropriate raw material as the aluminum source to form mullite [23–30].

Recently, the method of using bauxite to fabricate mullite has been studied from different viewpoints. By using bauxite and fly ash as the raw materials, Dong et al. [26] examined the influence of processing temperature on relative density, fracture strength, and microstructure of cost-effective mullite ceramics. In their

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study, the samples exhibited an average fracture strength of 186.19 MPa with a relative density of 93.94% after sintering at 1600 °C for 4 h. And the porous mullite membrane supports were successfully fabricated from 1100 °C to 1550 °C via nature bauxite and fly ash by Dong et al. [27] and via mineral coal gangue and bauxite by Lü et al. [28]. High strength mullite ceramics were successfully fabricated from industrial waste fly ash, bauxite, and V_2O_5 at relatively low temperature (1500 °C) by Li et al. [29].

In this paper, mullite-bonded porous SiC membrane supports (MPSMS) were synthesized from SiC, bauxite, and graphite in air at 1300–1450 °C. The oxidation-bonding characteristics, phase composition, microstructure, pore size distribution, open porosity, and flexural strength of the porous SiC membrane supports were investigated.

2. Experimental procedures

2.1. Starting materials and sample preparation

The average particle size of commercially available SiC powder (22.8 μm), calcined bauxite (4.2 μm) and graphite (18.7 μm) used in the experiment was measured by a particle size analyzer (PSA, Master Sizer 2000, Malvern Instruments, Worcestershire, U.K.) using distilled water as dispersing medium. Fig. 1 shows the particle morphology and the particle size distribution of the raw materials. As can be seen in Fig. 1, the particle morphology of SiC (Fig. 1a) is relatively regular, but SEM micrograph of bauxite (Fig. 1b) and C (Fig. 1c) is irregular. The PSA results as depicted in Fig. 1(A–C) show good agreement with the SEM results.

The weight ratio of bauxite to SiC + bauxite was fixed as 0.3, and the mass ratio of C in SiC + bauxite was defined as x. SiC, bauxite, and graphite were mixed in the ball milling with ethanol for 20 h to improve the homogeneity. The slurry was dried in an air oven at 90 °C for more than 4 h, and then screened through a 300-μm sieve. The powders were uniaxially pressed at 30 MPa pressure in a stainless steel die. The green bodies (circular disks of radius of 10 mm and thickness 4 mm and rectangular bars of 3 mm × 4 mm × 36 mm) were dried in an air oven for 24 h at 110 °C, then the specimens were put in a furnace (HTC 03/16, Nabertherm, Germany) to the set temperature for 3 h. The heating rate from room temperature to burn out C before 900 °C for 2 h was 2.5 °C/min, from 900 °C to 1350–1500 °C was 5 °C/min, and finally cooled naturally. In addition, using pure SiC and bauxite as raw materials, SiO_2-bonded porous SiC specimens and bauxite specimens were also fabricated, respectively.

2.2. Assessment methods

The open porosity of the sintered specimens was measured by using the Archimedes method. Pore size distribution was tested by Hg-intrusion porosimeter (Autopore IV 9500, Micromeritics, USA). Flexural strengths were characterized via three-point bending test (AGS-X, Shimadzu, Japan) with a cross-head speed of 0.5 mm/min and a support distance of 30 mm. Five rectangular specimens were measured to determine the average flexural strength and the corresponding standard deviation.

Thermal decomposition behavior and phase transformation characteristics of bauxite were evaluated by thermal gravimetric analysis and differential scanning calorimetry analysis (TGA–DSC, STA449C, Netzsch Co. Ltd. Germany) with a heating rate of 10 °C/min up to 1500 °C in air. Phase identifications of the raw materials and the specimens were determined by using an X-ray diffraction (XRD; D8 advance, Bruker, Germany). In the analysis, radiation CuKα, 40 kV, 40 mA and a step width 0.02° were used. The microstructure and morphology of the raw materials and porous specimens were observed by a field emission-scanning electron microscopy (FE-SEM; S-4800, Hitachi, Japan), with platinum covered. The well-developed neck element composition of the porous specimens was characterized by energy dispersive X-ray spectroscopy (EDS; XFlash Detector 5030, Bruker, Germany), which is attached in the SEM instrument.

3. Results and discussion

3.1. Characterization of starting materials

3.1.1. Phase composition

Fig. 2 shows the TGA–DSC curves of the calcined bauxite; the DSC curve shows two endothermic peaks at 965 °C and 1273 °C. Lü et al. [28] found that there is a significant sintering shrinkage with temperature increasing between 969 °C and 1276 °C. Simultaneously, the TGA curve shows no obvious weight change. Since C oxidation starts at 570–850 °C [19], the green bodies were heated from room temperature to 900 °C at a low heating rate to ensure that the network structure is stabilized.

Fig. 3 presents XRD spectra of the calcined bauxite (a) and SiC (b) sintered at various temperatures for 3 h. As can be seen in Fig. 3a, mullite ($3Al_2O_3 \cdot 2SiO_2$), corundum (Al_2O_3), and aluminum titanate ($Al_2O_3 \cdot TiO_2$) exist at all the six sintering temperatures and in the raw materials. Dong et al.[26] also showed that the temperature of the aluminum titanate formed by the reaction between rutile and alumina is above 1400 °C. Simultaneously, it can be inferred that both primary mullite crystals (formed in the kaolinite from metakaolinite decomposition) and secondary mullite (formed via the reaction between corundum and cristobalite) can be observed. The peak ratio of mullite increased with the increase of the processing temperature from 1400 °C to 1500 °C. The diffraction peak at 35° is the most intense because of the overlapping of the peaks of the (1 0 4) plane of corundum and the (1 1 1) plane of mullite. Previous studies [26–29] support the data obtained in this study.

Fig. 3b displays the XRD patterns of the pure SiC powder compact heated at indicated temperatures for 3 h. A small number of cristobalite peaks were observed at 900 °C. In addition, our previous work [19] showed that when SiC particles are heated up to 750 °C, the surface of the particles was oxidized to SiO_2. The DSC curve shows an exothermic peak at ~883 °C, as the oxidation-derived amorphous SiO_2 begins to be transformed into cristobalite. The peak intensity of cristobalite peaks was found to enhance continuously as the heat treatment temperature increased from 900 to 1400 °C.