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Toughening macroporous alumina membrane supports with YSZ powders

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Abstract

Macroporous alumina is an important support in membrane fields because of its stabilities to withstand exposure to high temperature, harsh chemical environment and high mechanical strength. However, the essence of brittleness can greatly shorten the life span and restrict the application fields. In this paper, YSZ (ZrO_2 stabilized by 3 mol% Y_2O_3) powders were added into alumina powders to improve the fracture toughness of macroporous Al_2O_3 supports sintered at 1400 °C and 1600 °C. The results show that the fracture toughness and the corresponding bending strength of supports are simultaneously greatly influenced by various YSZ contents. When YSZ content is 6 wt%, the maximum value of the fracture toughness is 3.0 MPa·m$^{1/2}$, and the bending strength is up to 90 MPa. By SEM and XRD analysis, the phase transformation of the uniform distribution t-ZrO_2 into m-ZrO_2 is the main cause which improves the fracture toughness of macroporous Al_2O_3 supports. Lowering of the sintering temperature by adding YSZ additives is also discovered here. The fracture toughness of the supports sintered at 1400 °C by adding YSZ powder is higher than that of the supports sintered at 1600 °C without adding any additives.

Keywords: Ceramic membrane; Support; Fracture toughness; Bending strength; YSZ

1. Introduction

Macroporous ceramic membranes have received much attention in recent years due to their stabilities to withstand exposure to high temperatures, harsh chemical environment and high mechanical strength [1,2], and are widely applied in fields of foodstuff, chemical industry, environmental protection, etc. [3–6]. Such ceramic membrane usually has an asymmetric structure, consisting of a top membrane layer with separation performance and a support which can provide mechanical strength for a top layer to withstand the stress induced by the different pressures applied over the entire membrane, and must simultaneously have a low resistance to filtrate flow [7].

Alumina ceramic with macroporous structure is one of the most important supports because of its advantages of durability, high temperature stability and chemical resistance [8]. However, its fracture toughness shows a low value due to the brittleness essence as well as that of dense ceramic [9], which limits its applications and/or shortens the life span during such processes as the sealing of ceramic membrane, the assembly processing step and the back flush. Therefore, the fracture toughness of macroporous ceramic supports requires improvement for a greater effectiveness.

At present, there are few literatures reporting on toughening methods and mechanism for porous ceramic. Researchers mainly aim at the novel preparing methods [10,11], the potential advantage properties and more widely application fields [12]. Naturally, high apparent porosity and high mechanical strength are simultaneously required under severe operating condition. When the apparent porosity increased, the corresponding mechanical properties will severely decrease [13]. The improvement of fracture toughness is usually at the expense of high porosity or bending strength. Therefore, it is worthy to study the toughening method and mechanism of macroporous Al_2O_3 support which usually has above 30% porosity to guarantee the fluent flow.

For a dense Al_2O_3 ceramic body, adding ZrO_2 has been considered as one of the most effective approaches [14,15] for giving damage-tolerance ability to brittleness. Some successful approaches were put into practice in many application fields [16,17], such as making ceramic knives, wear-resisting

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materials and other reactor vessels. In porous ceramic fields, it is also expected to improve the fracture toughness by adding suitable content of ZrO_2 powders. This paper mainly aimed at the improvement of fracture toughness for Al_2O_3 macroporous supports by adding YSZ (Zirconia stabilized by 3 mol% Y_2O_3) powders. The variation of apparent porosity, bending strength and fracture toughness with various feed YSZ powders and sintering temperatures were investigated. The toughening mechanism was also discussed.

2. Experimental procedure

2.1. Preparations of supports

The alumina powders purchased are prepared by hydrothermal method and the average particle size (D_{50}) is 24 μm (MasterSizer2000, Malvern Instrument, Co., UK). Fig. 1 shows the particle size distribution of Al_2O_3 powders (China Great Wall Aluminium Corporation, Zhengzhou, China). The average particle size (D_{50}) of YSZ powders (Material self-prepared) as additives is 0.7 μm. Wet-ball-milling was adopted to get the Al_2O_3/YSZ uniform mixture powders. According to the given composition of supports, mixture powders were put into the nylon jars. De-ionized water as dispersion and high hardness corundum balls were added into the mixture which pH was 6, milling for 24 h. The mixture described above was heated under stirring. After drying in a drying-oven at 110 °C for 48 h and grinding, the uniform Al_2O_3/YSZ mixture powders that passed through an 80 mesh screen were obtained. PVA (0.15 wt%) and paraffin (3 wt%) were added simultaneously. By dry pressing method, green-body rectangular bars of 6 mm × 6 mm × 50 mm and pellets of Ø30 mm × 2 mm were prepared under a pressure of 8 MPa. After dried at 110 °C for 24 h, the green-body samples were sintered in air for 2 h in an electric-furnace at 1400 °C and 1600 °C, respectively, with a heating rate of 3 °C/min and cooling naturally.

2.2. Characterization

Apparent porosity was measured by the Archimedes method with an immersion medium of water. The calculated formula was given as follows:

$$P = \frac{m_3 - m_1}{m_3 - m_2} \times 100\% \tag{1}$$

where P represents the apparent porosity of the support, m_1 is the weight of the dry support, m_2 is the weight of the support saturated of water suspended in water, m_3 is the weight of the support saturated water. The bending strength was tested by three point bending method. The fracture toughness values (K_{IC}) were determined using the Single Edge Notched Beam (SENB) technique [18]. Each sample was notched precrack of ~2 mm long before testing. The fracture toughness was given by [19]:

$$K_{IC} = Y\frac{3PL}{2bw^2}\sqrt{a} \tag{2}$$

where P is the fracture load, L is the length of the span between two points, b and w are related to the width and thickness, respectively, a is the depth of specimens center crack, and Y is a geometrical constant.

$$Y = 1.93 - 0.37\frac{a}{w} + 14.53\left(\frac{a}{w}\right)^2 - 25.07\left(\frac{a}{w}\right)^3 + 25.08\left(\frac{a}{w}\right)^4 \tag{3}$$

To test the mechanical properties of supports, the rectangular bars were grounded and beveled in advance to eliminate surface stress. The cross section of macroporous Al_2O_3 support was observed by scanning electron microscopy (SEM, Quanta 200, FEI, The Netherlands). The phase composition of ZrO_2 in samples after sintering was analysed by X-ray diffraction (XRD, D8 Advance, Bruker Instrument Co., Ltd. Germany) and the phase content was calculated based on the relative diffraction intensity of t-ZrO_2(1 1 1), m-ZrO_2(1 1 1) and (1 1 $\bar{1}$) peak, the relative content of t-ZrO_2 and m-ZrO_2 was obtained from [20]:

$$X_m = \frac{I_m(1\,1\,1) + I_m(1\,1\,\bar{1})}{I_m(1\,1\,1) + I_m(1\,1\,\bar{1}) + I_t(1\,1\,1)} \times 100\% \tag{4}$$

and

$$\phi_m = \frac{1.311X_m}{1 + 0.311X_m} \tag{5}$$

where X_m is the integrated intensity ratio of m-ZrO_2 volume, the subscripts m and t represent the intensities of the monoclinic and tetragonal phases after the peak separation and fitting procedures, $I_t(1\,1\,1)$, $I_m(1\,1\,1)$ and $I_m(1\,1\,\bar{1})$ represent the intensity of t-ZrO_2(1 1 1), m-ZrO_2(1 1 1) and m-ZrO_2(1 1 $\bar{1}$) peak, respectively, and ϕ_m is the volume fraction of the monoclinic phase.

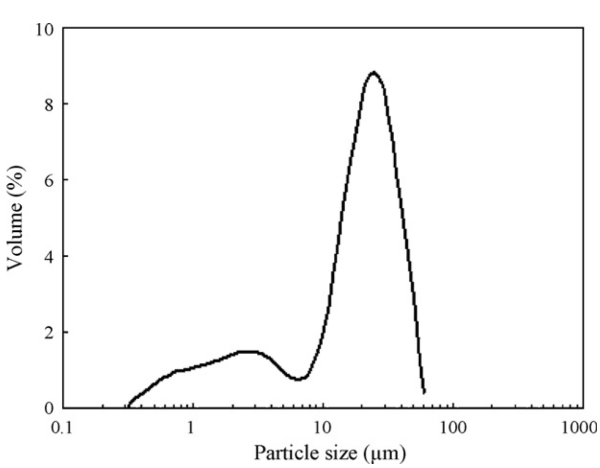

Fig. 1. Particle size distribution (D_{50}) of alumina powders.

3. Results and discussion

3.1. Effects of YSZ content

3.1.1. Porosity

According to the phase diagram of Al_2O_3–ZrO_2 [21], there is not any compound obtained in the Al_2O_3/YSZ system.