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Journal of the European Ceramic Society 34 (2014) 3819–3825

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Effect of particle size distribution of raw powders on pore size distribution and bending strength of Al_2O_3 microfiltration membrane supports

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Received 14 March 2014; received in revised form 11 May 2014; accepted 2 June 2014
Available online 3 July 2014

Abstract

To lower the sintering temperature of Al_2O_3 microfiltration membrane support, Al_2O_3 powders with particle size distribution of tri-modal are chosen. The results show that the function of fine Al_2O_3 grains depends on their agglomeration state: if fine Al_2O_3 grains distribute discretely, the bending strength of the support increases along with a slight increase in porosity; however, the aggregated fine grains are harmful to both bending strength and pore size distribution of the support. The bridging of medium Al_2O_3 grains between coarse grains contributes to increase the bending strength, but has less effect on porosity. The addition of medium (and/or fine) Al_2O_3 powder has less effect on the pore size distribution of the support if only coarse Al_2O_3 grain forms the support's framework, which suggests a new way to prepare the support with both high bending strength and high porosity at low temperature.

Keywords: Porous ceramic; Alumina membrane support; Particle size distribution; Pore size distribution; Bending strength

1. Introduction

Ceramic membranes have been gaining more attentions in industrial application due to their excellent thermal, chemical and mechanical stability.[1–4] Generally, ceramic membranes have an asymmetric structure consisting of a porous thin layer with separation function on a support. An excellent microfiltration membrane support should possess high mechanical strength, high permeability, and narrow pore size distribution, which mainly depend on the raw materials, sintering temperature and fabrication methods.[5] To achieve the high permeability, coarse alumina powder is generally chosen for the commercialized microfiltration membrane support. However, the poor sintering-activity of coarse alumina grains results in high cost of this kind support because high sintering temperature is required to obtain sufficient bending strength.[6] Though some raw materials with low melting temperature had been used to prepare the ceramic support,[6–8] alumina support is still popular due to its good bending strength and excellent acid/alkali resistance.[8] Some sintering aids, such as TiO_2,[4,7] boehmite,[9] kaolin,[10] and clay,[11] were added to decrease the sintering temperature. However, these materials weaken the bending strength and acid/alkali-resistance of the support in some degree.[11]

Ceramic microfiltration support is a kind of porous ceramic, whose bending strength is proportional to the neck areas among the grains. The particle size has an important effect on the neck area among the grains through sintering. To a pair of grains with different particle sizes, it has been proved that fine grains disappear and coarse grains coarsen through surface diffusion or grain boundary diffusion.[12–16] To an array of coarse-fine-coarse grains, the neck area among coarse Al_2O_3 grains increases due to the immigration of fine grains. Taking this rule into account, the support is designed using alumina powders with the particle size distribution of tri-modal as raw materials. The support sintered at

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http://dx.doi.org/10.1016/j.jeurceramsoc.2014.06.001
0955-2219/

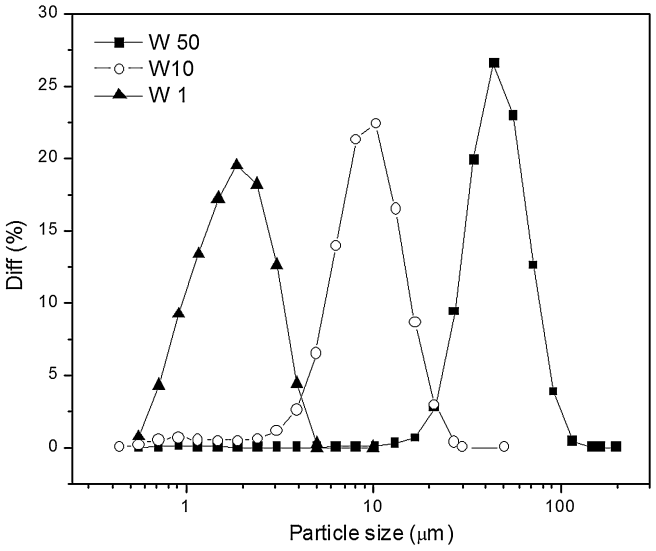

Fig. 1. Particle size distributions of W50, W10 and W1.

a relative low sintering temperature can own both high bending strength and high porosity. Simultaneously, the broader particle size distribution contributes to the sintering because of the high green compact density.[12,17–19] In another words, the sintering of the porous ceramic can be realized by the sintering-induced immigration of the fine grains at a relatively lower temperature without sintering of coarse grains. This method gives a promising way to obtain ceramic membrane support with high porosity without degrading bending strength. However, the broad particle size distribution may also lead to the difference of localized densification,[17] which results in a broad pore size distribution.[20] The case should be avoided for Al_2O_3 microfiltration membrane supports.[17]

In the present work, the powders with tri-modal particles size distribution were prepared by mixing three different powder sizes (d_{50} = 37 μm, 8.2 μm, 1.6 μm). The effect of particle size distribution on the pore size distribution, the porosity and bending strength of ceramic membrane supports were investigated in detail.

2. Experimental

2.1. Preparation of ceramic membrane supports

Al_2O_3 powders, labeled as W50, W10 and W1 (purity of 99.5%, Henan White-dove Group, China) were purchased without further treatment prior to the preparation of microfiltration membrane supports. The d_{50} of W50, W10 and W1 were 37 μm, 8.2 μm and 1.6 μm, respectively. Their particle size distributions are shown in Fig. 1.

Alumina powders with different average particle size were prepared by mixing W50, W10 and W1 in different ratios by ball milling for 2 h at 150 rpm as shown in Table 1. The particle size distribution and loose density of the mixed powders were measured.

To prepare the support, alumina powder, 2 wt% TiO_2 were mixed with 1 wt% poly vinyl alcohol (PVA, molecular weight of 1750 ± 50, was solved into 5 wt% aqueous solution) of the total alumina powder in a ball mill at 150 rpm for 2 h to avoid the breakage of alumina grains. The mass ratio of powder:zirconia ball:alcohol is 1:1:2. The obtained suspensions were dried in an oven at 70 °C for overnight. The mixed powders were compressed into the rectangle shape of 40 mm × 10 mm × 10 mm ($L \times h \times w$) by dry pressing under the applied pressure of 12 MPa. The samples were calcined in a programmable furnace at a heating rate of 3 °C/min to 1650 °C and annealing for 2 h.

Table 1
Compositions of W50, W10 and W1 used for preparing supports.

Powder	W50/wt%	W10/wt%	W1/wt%
Sample label			
S1	80	16	4
S2		10	10
S3		6	14
S4	70	24	6
S5		15	15
S6		9	21
S7	60	32	8
S8		20	20
S9		12	28

2.2. Characterization

The particle size distributions were determined by laser scattering particle size analyzer (Bettersize2000, Dan Dong, China).

The loose densities of the powders were measured by Hall flow meter (FT-01, Ningbo, China). The densities of the green compacts were measured by mass-volume method. The dimensions were measured by a caliper and the weight was measured by the electronic balance with the accuracy of 0.0001 g.

The pore size distributions and the porosity of the sintered compacts were measured by Mercury Intrusion porosimeter (Autopore IV9500, Micromeritics, USA).

The bending strength was determined by three-point bending method at room temperature using universal material testing machine (WDW-30, Xi'an Letry Machine Testing Co. Ltd., China). The span length is 20 mm and the loading speed is 0.2 mm/min. Three samples were tested to determine the bending strength.

Fracture surfaces of the sintered ceramic supports were observed by means of field emitting scanning electron microscope (JSM-6700F, JEOL, Japan). To test the evolution of the shape of alumina grain, the green compact of sample S4 was observed by SEM.

3. Results and discussion

3.1. Relationship of particles size distribution with pore size distribution of supports

The particle size distributions of the mixed powders S1–S9 are shown in Fig. 2. All particle sizes distribution of the mixed powders shows the tri-modal distribution. The difference in mass