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Preparation of high porous silicon nitride foams with ultra-thin walls and excellent mechanical performance for heat exchanger application by using a protein foaming method

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Abstract

Ceramic foam with excellent mechanical performance, high thermal conductivity and high surface area-to-volume ratio is thought to have great potential in heat exchanger application. In this study, silicon nitride (Si_3N_4) foams with ultrathin cell walls were prepared by using a protein foaming method via increasing the air fraction of foamed slurry. Cell thickness of these Si_3N_4 foams was only 2–3 μm and the pores in the Si_3N_4 foams fabricated here exhibited polyhedral shapes. Thermal conductivities of these Si_3N_4 foams ranged from 2.427 $W\,m^{-1}\,K^{-1}$ to 3.154 $W\,m^{-1}\,K^{-1}$, whereas compressive strengths of these Si_3N_4 foams are in the range from 12.85 MPa to 19.99 MPa. Moreover, compressive load–displacement curves of the Si_3N_4 foams generally exhibit cellular behavior.

Keywords: B. Porosity; C. Thermal conductivity; D. Si_3N_4; E. Heat exchangers

1. Introduction

Heat exchanger, such as the bipolar plates in fuel cell stacks and the heat sinks for cooling of power, is a very important unit in many systems. Up to now, the widely used heat exchangers are generally made of metal foams, such as aluminum foams and copper foams [1–6], or carbon foams [7] with high thermal conductivity and high surface area-to-volume ratio. However, these two kinds of foams cannot meet the demands of many harsh conditions due to low thermal and chemical stability, low mechanical performance and high thermal expansion coefficient of metal foams or the low mechanical performance of carbon foams. Therefore, new kind of porous material with high thermal conductivity needs to be developed. Si_3N_4 is a kind of ceramic with high thermal conductivity, outstanding mechanical performance, low thermal expansion, extraordinary thermal shock resistance, and excellent thermal and chemical stability [8–12]. The thermal conductivity of Si_3N_4 is estimated up to 200 W/m K [13] and dense β-Si_3N_4 ceramic with a thermal conductivity up to 170 W/m K has been achieved by Zhu et al. [14]. Consequently, highly porous Si_3N_4 foams with excellent thermal conductivity are very suitable candidates for substituting the aluminum foams, the copper foams and the carbon foams in the hash conditions of this application.

Numbers of methods have been developed for preparing ceramic foams [15–18], such as replica method, sacrificial template method, direct foaming method, three-dimensional ceramic/camphene-based coextrusion (3D-CoEx) method [19] and three-dimensional printing of preceramic polymer method [20]. Among them, the direct foaming method bears some striking advantages and can be used to prepare ceramic foams with extraordinary overall performance. According the foaming way of this method, it can be classified as self-foaming route and air-incorporation foaming route. In the self-foaming route, the bubbles are formed by the released small molecules, which are derived from the pyrolysis of precursor [21–27] or the vaporization of the solvent [28]. While in the air-incorporation foaming route, the bubbles are formed by

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incorporating gas and stabilized by either particles, the surfaces of which are modified by short chain amphiphile molecules [29–31], or long chain surfactants which can be environment friendly bio-molecules, such as protein [32–35], starch [36–38] or a mixture of them [39,40]. It should be mentioned that, bubbles stabilized by particles are generally more stable than the bubbles stabilized by long chain surfactant [15,31,41,42].

As for ceramic foams, cell thickness is one of the most important microstructure features that determined their surface area-to-volume ratio. Up to now, only the particles stabilizing air-incorporation route has been reported to be able to prepare ceramic foams with ultrathin walls (only a layer of grains) [29,30]. However, the raw materials of this method are generally limited to nano-scale or submicro-scale, which results in the high cost of production for preparing ceramic foam with this kind of microstructure.

In this research, we report the preparation of Si_3N_4 foams with ultrathin cell walls by using the protein foaming method (a typical kind of long chain surfactants stabilizing air-incorporating foaming route). In comparison with the particles stabilizing air-incorporating foaming route, the protein foaming method is more economical and environment friendly. As far as we concerned, this is the first dealing with the preparation of Si_3N_4 foams with ultrathin cell walls by using the protein foaming method. The prepared Si_3N_4 foams generally have high porosities, excellent mechanical performances and thermal conductivities.

2. Experimental procedure

Commercial Si_3N_4 powder (d_{50}= 2.0 μm, purity of 99.9%, produced by Beijing XinRongYuan Technology Co., Ltd., China) was dispersed into distilled water by ball milling in a 500 ml bottle at a solid content of 34.7 vol%, together with 6 wt% Y_2O_3 (purity of 99.99%, Sinopharm Chemical Reagent Co., Ltd., China) and 3 wt% Al_2O_3 (200–300 mesh, Sinopharm Chemical Reagent Co., Ltd., China). Morphology of the Si_3N_4 particles was shown in Fig. 1. 0.35 wt% polyacrylate (PAA, M_W=3000, Aladdin Industrial Corporation) was selected as dispersant (based on the weight of Si_3N_4 powder) to disperse Si_3N_4 particles in suspension. The mixture was ball-milled in a planetary mill (QM-3SP2, produced by Nanjin University, China) for about 20 h before adding the egg albumen powder (protein content >86.5%, produced by Taiyo food (Tianjin) Co., Ltd., China) to acquire well-dispersed suspension. Certain mount of egg albumen powder (based on the weight of distilled water) was added into the suspension. The suspension together with the egg albumen powder was rotated at 300 rpm in the planetary ball milling. It took about 45 min to finish the foaming procedure. Air fraction was controlled by adjusting the volume of suspension and it was calculated according to the following formulate in this research:

$$\text{Air fraction} = \frac{V_{bottle} - V_{balls} - V_{slurry}}{V_{bottle} - V_{balls}} \times 100\% \quad (1)$$

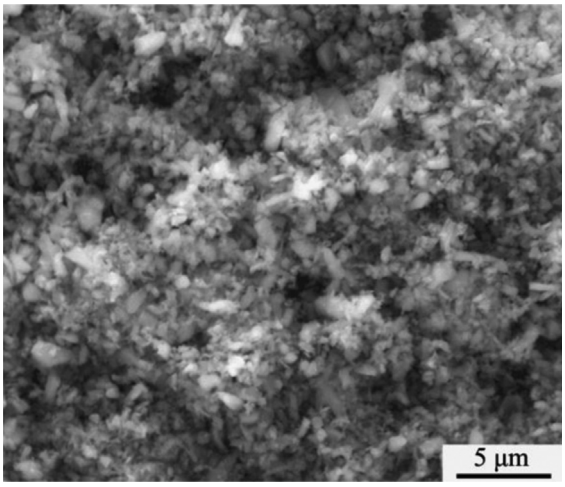

Fig. 1. Morphology of the Si_3N_4 particles.

where V_{bottle}, V_{balls}, *and* V_{slurry} were the volume of bottle, volume of balls and volume of slurry, respectively. In this research, the air fraction was controlled to be about 74.4%.

After the foaming procedure, the foamed slurry was casted into a cylindrical mold made of polytetrafluoroethylene (PTFE) for thermal consolidation. The thermal consolidation of foamed slurry was similar to the making of steam bread and details about the thermal consolidation procedure were reported elsewhere [43,44]. After the consolidation procedure, the as generated green body was cooled to room temperature and then dried in air at room temperature. Protein was removed by burnout in air at the temperature of 600 °C for 1 h with a heating rate of 1 °C /min before final sintering. The green bodies were sintered at the temperature of 1820 °C for 1 h under 10 MPa nitrogen pressure in a graphite furnace (FCT FP W 25-PT, Germany) or at the temperature of 1750 °C for 1 h under 0.3 MPa nitrogen pressure in a graphite furnace (High-multi 5000, Japan) with a heating rate of 10 °C/min. During the cooling process, the cooling rate was controlled to be 10 °C/min above 1050 °C. Powder bed was not used during the sintering of Si_3N_4 foams.

2.1. Characterization of Si_3N_4 foams

Rheology of the foamed slurry was characterized by modular compact rheometer MCR320 (Anton Paar company). The as-prepared Si_3N_4 foams were cut into a dimension of $10 \times 10 \times 10$ mm^3 before the measurement of porosity, density and compressive strength. These Si_3N_4 foams cubes were boiled in distilled water for about 3 h to completely remove air in their pores before the measurement of porosity and density by using the Archimedes method. Three specimens were used to determine the average porosity and density. The compressive strength was evaluated in a universal testing machine (WDW^{-1}00, China) using a crosshead displacement speed of 0.5 mm/min. Plotted compressive load–displacement curves were used to determine values for yield stress (σ_y) corresponding to the maximum compressive strength. At least seven specimens were used to determine the average compressive

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