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Fabrication of pure SiC ceramic foams using SiO_2 as a foaming agent via high-temperature recrystallization

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ABSTRACT

A novel method was developed to produce the pure silicon carbide foams via the high-temperature recrystallization with the presence of a novel foaming agent-SiO_2. In this method, SiO_2 reacts with SiC to produce the gases in the silica liquid at high temperature, which leads to the formation of foams. The foams consist of the directional and interconnecting SiC crystals, and numerous intercommunicating pores that are located between them. The phase of foams was identified as 6H–SiC without the presence of SiO_2 since SiO_2 particles could react completely with SiC particles and vaporize from the sample at high temperature. The total porosity, weight loss and volume expansion rate can be increased with increasing SiO_2 contents while the three-point bending strength decreasing. The porosity of SiC foam with 25 wt.% SiO_2 as a foaming agent exhibits the maximum value while the three-point bending strength shows minimum value correspondingly. The sintered samples presented the porosities of 61–81%, the bending strength from 1.5 MPa to 4.8 MPa, and the volume expansion rate from 17.4% to 65%. This research can develop the theory for the preparation of SiC ceramics foams with controlled structure.

1. Introduction

Ceramic foams are a class of porous materials with large voids, and have a wide range of applications due to their unique properties such as low density, low thermal conductivity and thermal shock resistance [1,2]. Among a number of oxide and non-oxide ceramic foams, silicon carbide foams have unique properties such as high thermal conductivity, extremely high chemical inertness and high temperature strength making them suitable for industrial applications such as catalyst supports, hot gas filtration, metal–ceramic composites, filters in the metal casting industry, and high efficiency combustion burners [3–8]. Silicon carbide foams can be produced by different methods such as the impregnation of polyurethane foams with a ceramic suspension followed by the pyrolysis and pressureless sintering at elevated temperatures [4,9–13], the gel casting process which combines the foaming of ceramic suspension and in situ polymerization [14], and the direct blowing of a preceramic polymer [15]. It is well known that silicon carbide is very difficult to densify without sintering aids due to the covalent nature of Si–C bonding and low self-diffusion coefficient [16]. The sintering aids such as AlN, Si_3N_4 and Al_2O_3 are usually used to decrease the sintering temperature and manufacturing costs of SiC ceramics [6,9,10,17,18]. However, for the fabrication of SiC ceramic foams, the additives will induce the high shrinkage, which results in the destruction of the foam ceramics during sintering [9]. The residual carbon, silicon and additives such as AlN, Si_3N_4 and Al_2O_3 will decrease the thermal strength, oxidation resistance and chemical stability of SiC foams [3].

In this work, pure SiC foams were directly synthesized using SiC and SiO_2 powders via the high-temperature recrystallization. SiO_2 particles first play a role of binder to bond SiC particles during sintering [19]. With increasing the temperature higher than 1950 K, SiO_2 will melt, vaporize, and react with SiC to produce SiO, Si and CO gases and play a role of a foaming agent [20–22]. The recrystallized SiC crystals are generally formed between 2073 K and 2800 K [23,24]. During the high-temperature recrystallization at 2573 K in this study, the feature of silicon carbide foams sintering is the active participation of the gaseous phases, which leads to the formation of silicon carbide foams with unique microstructure. The effects of SiO_2 contents on the porosity, pore size, weight loss, volume expansion rate, and mechanical property of SiC foams have also been investigated.

2. Experimental

Starting materials were commercial SiC powders (α-SiC, average particle size of 112.1 μm, purity of 99.9%, Zaozhuang Liyuan Silicon Carbide Co., Ltd., China) and SiO_2 powders (α-SiO2, quartz

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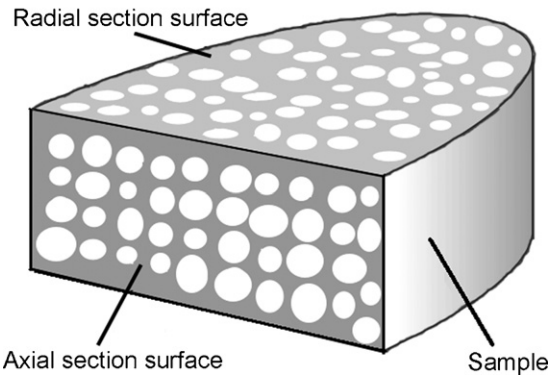

Fig. 1. The schematic distribution of the fields of view for the measurement of pores size.

sand, purity of 99.9%, average particle size of 107.5 μm, Zaozhuang Jinzhu Quartz Sand Plant, China). The weight ratios of SiO_2 and SiC particles were 5:95, 10:90, 15:85, 20:80, and 25:75, respectively. 1 wt.% Polyvinyl butyl (PVB) and 0.1 wt.% stearic acid were added into the mixture as a binder and lubricant. The powders were milled in ethanol using agate balls as the milling media. The milled powders were dried in an evaporator and sieved through a 50-mesh screen, and then pressed into the pellets with diameter of 30 mm and height of 10 mm under 1.5 MPa pressure. Subsequently, the green pellets were put into a graphite crucible and sintered in a medium frequency vacuum induction furnace (Model ZGRS-160/2.55 Jinzhou Electric Furnace Co., Ltd., Jinzhou, China). The specimens were firstly heated until the melt point of SiO_2 with the heating rate of 20 °C/min. Then, the specimens were sintered in nitrogen atmosphere (0.5×10^5 Pa) at 2300 °C for 5 min with a heating and cooling rate of 10 °C/min. After sintering, the sintered samples were first decarburized at 750 °C for 10 h, and then grinded and cleaned.

The bulk density and total porosity was determined by the Archimedes method, and kerosene was used as the liquid medium. The microstructure of SiC ceramic foams such as the size and shape of pores was observed by scanning electron microscopy (SEM, VEGA3 XMU, TESCAN, Czech Republic). The distribution of pore diameter was estimated by quantitative image analysis software Image-Pro Plus (Version 7.0, Media Cybernetics, USA). The distribution of the fields of view is shown schematically in Fig. 1. The fields of view were randomly distributed over the radial section surface and systematically shifted 2 mm in horizontal and vertical directions on the axial section surface, thus allowing the analysis and measurement of the distribution of pores size. The distribution of particle size was characterized by laser particle size analyzer (Model Rise-2008, Jinan Rise Science & Technology Co., Ltd., Jinan, China). The phase formation of specimens was analyzed using X-ray diffractometry (XRD, X'Pert PRO, PANalytical, Netherlands) using Cu Kα radiation. XRD patterns of samples were obtained in the 2θ range from 15° to 85° with a step of 0.01° and scan speed of 10°/min. The specimens were cut into the dimension of 6.0 mm × 6.0 mm × 25.0 mm to measure the flexural strength via the three-point bending test (Model WDT-10, Tianshui Hongshan Test Machine Accessories Factory, Tianshui, China) with the support distance of 20.0 mm and cross-head speed of 0.5 mm/min. More than four specimens were tested as a group and three groups to obtain the strength.

3. Results and discussion

3.1. Macrostructure and microstructure

The macro-appearances of the green pellet and SiC foam with 15 wt.% SiO_2 as a foaming agent were shown in Fig. 2. As can be seen, the volume of SiC foam was much higher than that of the green pellet, which exhibited the "cake" shape. The surface of SiC foam was coated by the thin shell of fine SiC particles. The volume expansion rates of foams were discussed in Section 3.3.

Fig. 3 shows the typical microstructures of SiC foams in the radial and axial sections, respectively. The samples with different SiO_2 contents exhibited the similar microstructures. Hence, we only discussed the microstructure of the typical sample with 15 wt.% SiO_2. Fig. 3 shows that the foams consisted of the directional, enlarged SiC crystals which were interconnected, and numerous pores which were located between them. Most of pores are intercommunicating, and exhibit a preferential orientation to some extent, in the axial section (Fig. 3c and d). The morphologies of pores are different in the radial and axial sections, such as the irregular pores in the radial section surface, while the regular and directional pores in the axial section surface.

Fig. 4 is X-ray diffraction patterns of sintered sample with 25 wt.% SiO_2 and raw SiC powders. It shows that all the peaks of sample can be identified as pure 6H–SiC, and the peaks corresponding to SiO_2 were not observed even for the addition of SiO_2 content up to 25 wt.% since SiO_2 particles react completely with SiC particles and vaporize from the sample. Comparing the intensities of the (0 0 0 1) peaks of foams in the axial and radial sections, and raw SiC powder, the preferential orientation in (0001) direction was exhibited, forming *c*-axis oriented grains. It suggests that the oriented growth of SiC crystals was in the axial direction during sintering

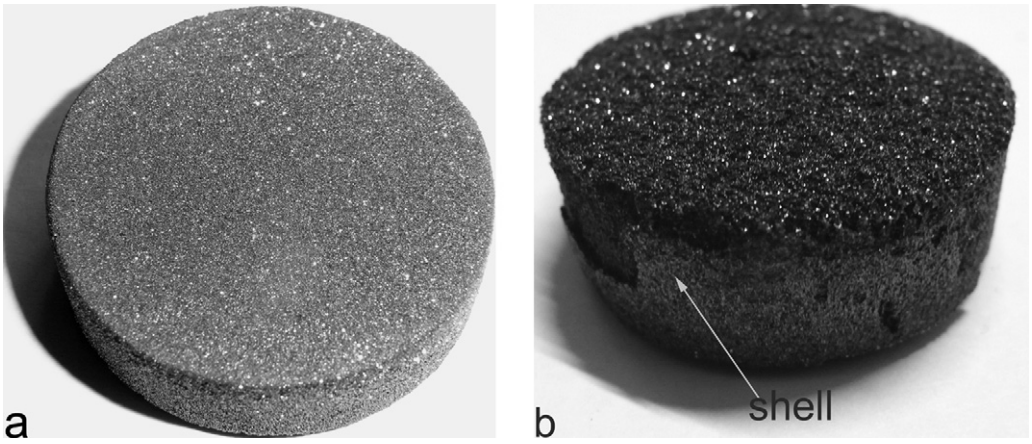

Fig. 2. The macro-appearance of samples with 15 wt.% SiO_2: (a) green pellet; (b) SiC foam.