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Fabrication and properties of SiC porous ceramics using a polyurethane preparation process

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ABSTRACT

SiC porous ceramics can be prepared by introducing the polyurethane preparation method into the production process of ceramic biscuits, followed by sintering at 1300 °C for 2 h under N_2 flux after the cross-linking of polycarbosilane at 220 °C for 4 h in air. The microstructures, mechanical properties and infiltrations of the SiC porous ceramics are investigated in detail. The best dispersal effect comes from the SiC slurry with xylene as the solvent and a mixture of Silok®7096 (1 wt%) and Anjeka®6041 (4 wt%) as the dispersant. The compressive strength of SiC porous ceramics with high porosity (69.53%) reaches 16.9 MPa. The heat treatment can increase infiltration, the rate of which (4.296×10^{-7} mm^2) after the heat treatment at 750 °C in air is approximately two times faster than that before the heat treatment. The SiC porous ceramics fabricated in this study will have potential application in active thermal protection systems.

1. Introduction

SiC porous ceramic has a three-dimensional space network structure and is candidate material for high-temperature structural applications, mainly due to its high porosity, excellent high temperature oxidation resistance, mechanical properties and so on [1–5]. This porous material has been received increasing attention for use in the transpiration cooling system of spacecraft.

Currently, various methods have been successfully used to fabricate SiC porous ceramics, including directing foams from blowing agent, the sacrificial template method (impregnation of ceramic slurry with organic-inorganic template), the sol-gel process and the freezing method [6–10]. These methods have their own advantages and drawbacks. Direct foaming is a simple approach for the preparation of ceramics with high porosity but provides limited pore structural control. Alternatives such as the sacrificial template method usually result in low strut densities, which limit the increase of the mechanical strength of SiC porous ceramics [11–13], although the uniform distribution of the open pores can be controlled. To improve the infiltration and mechanical properties of SiC porous ceramics, we propose a novel fabrication process by introducing the polyurethane preparation method into the production process of ceramic biscuits, which combines the advantages of directing foams and the sacrificial template method [14,15].

In this novel fabrication process, polycarbosilane (PCS, a type of preceramic polymer containing Si/C = 1:1) is used as the binder among the SiC particles to reduce the sintering temperature. Notably, PCS is subjected to conversion from a polymer to a ceramic under heating at temperatures higher than 1000 °C in N_2 flux, in which volume shrinkage typically occurs and limits the fabrication of small dimensional porous materials. The preparation of SiC porous ceramics remains a longstanding challenge. To over this problem, we added SiC particles as aggregates in this fabrication process to prevent shrinkage. Here, the novel preparation details and characterization (microstructures, mechanical properties and infiltration) of SiC porous ceramics are reported and discussed.

2. Experimental procedure

2.1. Fabrication of porous ceramics

The fabrication procedures of SiC porous ceramics consist of the following steps. First, the ceramic slurry is prepared. A certain proportion of dispersants (Silok®7096, a high molecular weight copolymer alkylamine salt, Guangzhou Silok Polymer Co., Ltd., Guangdong, China; Anjeka®6041, Ezhou Anjeka Technology Co., Ltd., Hubei, China) and polycarbosilane (PCS, Suzhou Saifei Group Co., Ltd., Jiangsu, China), the mean molecular weight and melting point of which are 1184.5 and

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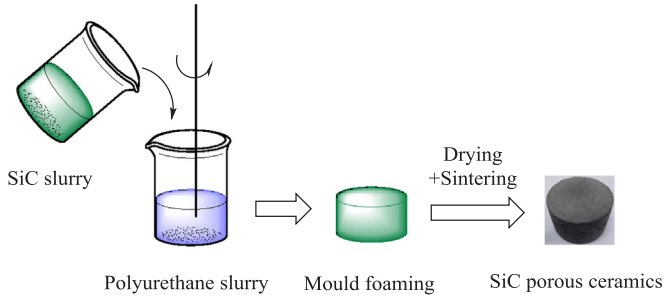

Fig. 1. Preparation flow chart of SiC porous ceramics.

217 °C, respectively, are dissolved in xylene (Tianjin Yongda Chemical Reagent Co., Ltd., China). A pinch of kaolin (Lingshou Yanxi mineral processing plant. Hebei, China) with an average particle size of 3.4 μm is added to the solution to improve the rheology and thixotropy of the slurry, which has enough viscosity under static conditions and good fluidity when stirred [16,17]. β-SiC particles (Tonghua Hongxin abrasive Co., Ltd., Jilin, China) with an average grain size of 5 μm are used as the starting powders in the slurry with continuous stirring for 30 min. Next, a polyurethane slurry is obtained from a mixture of carbon fiber (7 μm in diameter, 2 mm in length, Tianniao High Technology Co., Ltd., Jiangsu, China) and two types of polyurethanes that contain integral skin polyurethane and high rebound polyurethane (Beijing Hagibis Technology Co., Ltd., China). Subsequently, the ceramic slurry is added to the prepared polyurethane slurry under stirring for 10 min and then transferred to a cylindrical mold to foam for 24 h at room temperature. Finally, after foaming at room temperature, the green bodies are dried in a drying oven (DHG-9240A, Shanghai Huitai Equipment Manufacturing Co., Ltd., China) at a constant temperature of 80 °C before heating them for 4 h at 220 °C in air and for 2 h at 1300 °C in a pure nitrogen flow (99.99%) with a programmed heating speed of 1 °C/min, followed by natural cooling. The preparation flow chart of SiC porous ceramics is shown in Fig. 1.

2.2. Characterization of porous ceramics

The foam morphology of SiC porous ceramics is characterized by scanning electron microscopy (SEM, S-4800, Hitachi, Japan) after polishing and ultrasonic cleaning. The bulk density of SiC porous ceramics is determined from the ratio of the mass and volume of the specimens. The thermal diffusion coefficient and thermal conductivity of SiC porous ceramics at room temperature are measured using a thermal conductivity tester (DRE-III, Xiangtan Xiangyi Instrument Limited Company., Hunan, China). Testing specimens with dimensions of 20 mm × 20 mm × 20 mm are cut from the original samples, and then, the crushing strength of the SiC porous ceramics is determined at room temperature by compressive testing using a microcomputer controlled spring tension and compressive testing machine (Jinan Liangong Testing Technology Co., Ltd., Shandong, China) at a cross-head speed of 1 mm/min. The infiltration of the test pieces is obtained using a piece of in-house simple equipment that consists of a beaker and circular tube filled with clean water.

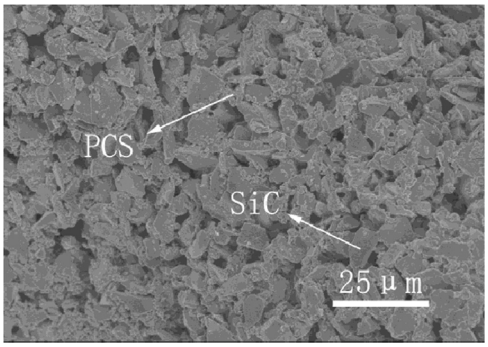

Fig. 3. Microstructure of SiC particles cohered by PCS.

3. Results and discussion

It is hard to form a stable SiC suspension in xylene due to the low polarity of xylene. Therefore, the stability of SiC particles in xylene containing different types and amounts of dispersants is studied by means of the sedimentation test. It is reported in Fig. 2 that all of the SiC particles in the presence of dispersant 7423 sink to the bottom of the test tube within 1 min. Conversely, more uniformly dispersed SiC particles are obtained after chemical absorption for 190 min in xylene with a mixture of Silok®7096 (1 wt%) and Anjeka®6041 (4 wt%) as the dispersant.

Fig. 3 shows the microstructure of the SiC porous ceramic and that the SiC particles are evenly coated by PCS.

It is estimated from Figs. 2 and 3 that the functional groups at one end of the dispersant adsorb to the SiC particles surface and that the functional groups at the other end of the dispersant stretch into the solvent medium to form a steric hindrance layer to block the aggregation and precipitation of SiC particles [18–21]. However, the blockage subsides when the dispersants are supersaturated on the surface of SiC particles owing to the wrapping of the organic chains of the dispersants.

The microstructure of SiC porous ceramics with different contents of polyurethanes is shown in Fig. 4. A large amount of closed pores is observed due to insufficient CO_2 production when the mass ratio of the polyurethane slurry/SiC ceramic slurry is 30%, as shown in Fig. 4(a). SiC particles tightly aggregate when the buoyancy from the bubbles barely overcomes the gravity of the SiC powders. On the contrary, breakage of the struts leads to a lower strength due to the production of an excessive number of voids when the mass ratio of polyurethane and

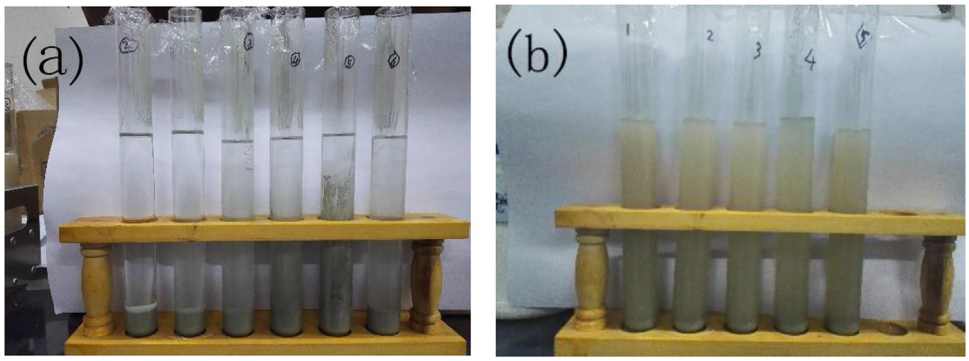

Fig. 2. Settlement test of SiC ceramic slurries with different types and contents of dispersants: (a) dispersant 7423 for 1 min, from left to right: 0%, 1% , 2%, 3%, 4%, and 5%; (b) dispersant 7096/6041 for 190 min, from left to right: 0, 1/4, 2/4, 3/4, and 4/4.