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Low-temperature fabrication and characterization of porous SiC ceramics using silicone resin as binder

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Abstract

Commercially available silicone resin and silicon carbide (SiC) powders were adopted as the starting materials for the fabrication of porous SiC ceramics. During the heat treatment process, silicone resin experienced an organic–inorganic transformation and acted as the bonding material between SiC particles at a low temperature of 1000 °C. The mean particle size of starting SiC powders and silicone resin content can control the pore size, open porosity and fracture strength. The flexural strength of porous SiC ceramics increases with increasing silicone resin content and decreasing mean particle size of SiC powders. Larger pores can be obtained with coarser starting SiC powders and higher silicone resin content. The fracture surface of porous SiC ceramics was observed.

Keywords: Porous SiC ceramics; Silicone resin; Preceramic polymer pyrolysis; Porosity; Fracture strength

1. Introduction

Porous SiC ceramics, which exhibit a unique combination of excellent mechanical strength, thermal stability, chemical inertness as well as good thermal conductivity and electron conductivity [1–3], can be applied in many areas, such as filters, catalyst supports, heat exchangers, electrodes, sensors and so on [4]. However, due to the covalent nature of Si–C bonds, SiC ceramics normally need to be sintered at high temperature or/and with the addition of sintering agents, which have limited the application of porous SiC ceramics.

Preceramic polymer process, during which the polymer precursors are converted into ceramic materials, is a new route to overcome these problems. The process has the advantage of requiring usually low temperatures. However, very little study has been reported about the application of preceramic polymer in fabricating porous SiC ceramics [5–7]. Zhu et al. [5] and Ma and Yu [6] separately prepared porous SiC ceramics using polycarbosilane (PCS) as binder at a low temperature (1000–1100 °C). As the result of the low ceramic yield (~60 wt.%) of PCS, the fracture strength of porous SiC ceramics they made was less than 16 MPa. Silicone resin with a high ceramic yield (85 wt.%) was adopted as binder by Herzog et al. [7] to fabricate porous SiC ceramics. However, the resultant porous SiC ceramics showed low fracture strength (2–4 MPa). The authors did not give an explanation. The objective of this work is to report the method of using silicone resin as the bonding material to fabricate porous SiC ceramics with high fracture strength at a temperature as low as 1000 °C. The structures and properties of the porous SiC ceramics are discussed.

2. Experiment

Commercially available α-SiC powders (mean sizes of 0.4 and 10 μm, respectively) and DC249^{TM} silicone resin (Dow Corning 249, Flake Resin) were chosen as starting materials. The mixtures of SiC powders and DC249 with various mass ratios were ball-milled for 3 h with acetone as solvent for DC249. Then the acetone was evaporated to obtain DC249-coated SiC powders. The above powders were ground and passed through a 100 mesh sieve and then were pressed into wafers under 15 MPa. The wafers were cross-linked at 250 °C for 6 h in the oven, and then fired at 1000 °C for 1 h with a heating rate of 1 °C/min. High purity N_2 was used as the protective atmosphere.

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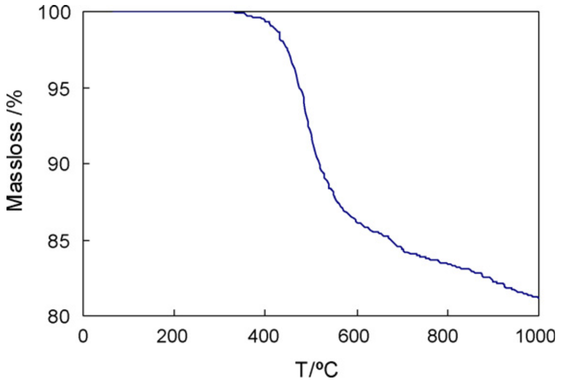

Fig. 1. Thermal gravity curve of cured DC249 (Ar, 5 °C/min).

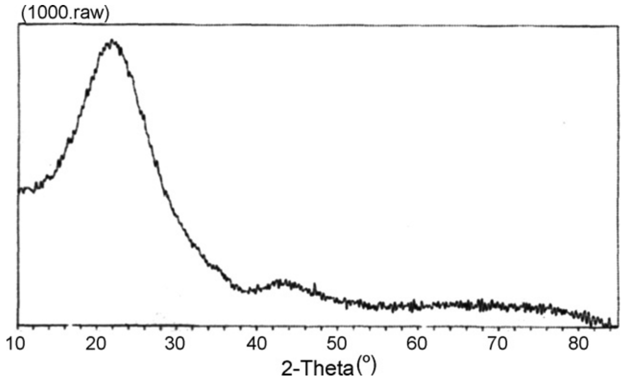

Fig. 2. XRD pattern of DC249 pyrolyzed at 1000 °C.

DC249 pyrolysis conversion process was investigated by TG (STA 449C, Netzsch) and XRD (Siemens-D500). Open porosity of the porous specimens was determined according to the standard GB/T1966–1996. Pore size distribution was measured by mercury intrusion porosimetry (PoreSizer 9320, Micromeritics). Wafers were machined into bars of 3 mm (thickness) × 4 mm (width) × 40 mm (length) for three-point bending test with a cross-head speed of 0.5 mm/min, and a support span of 30 mm. Three bars were tested to obtain the average fracture strength. SEM (JSM-5600LV, JEOL) observations were conducted to examine the microstructures.

3. Results and discussion

Fig. 1 shows the thermal gravity curve of cured DC249 (Ar, 5 °C/min), the starting preceramic polymer. DC249 is a kind of preceramic polymer with a Si–O backbone and its side groups are –CH_3, –OH and –Ph. When heated in an inert atmosphere, the low molecule weight species decompose, which causes weight loss. As can be seen from the TG curve, there is a total weight loss of ~18% from room temperature to 1000 °C. The major weight loss occurs from 400 to 800 °C, during which a low heating rate (1 °C/min) is necessary to avoid stress accumulation. Fig. 2 is the XRD pattern of DC249 pyrolysis products. DC249 treated at 1000 °C in inert atmosphere yields the residue which is in an amorphous state. According to refs. [8–11], the pyrolysis products at 1000 °C of DC249 consisted of Si, C, O atoms and its structure was a mixture of free carbon and a three-dimensional random network of silicon–oxygen tetrahedral with some silicons bonded to one or two carbons substituted for oxygen which were in turn tetrahedrally bonded to other silicon atoms. In the current route, the use of α-SiC powders as starting materials was considered not to influence the pyrolysis behavior of DC249 because SiC powder was an inert filler for DC249 [7,12].

Table 1 shows open porosity, fracture strength and average pore size of porous SiC ceramics changing with DC249 content and mean particle size of SiC powders. From these figures, the influence of DC249 content is apparent. Higher DC249 content results in lower open porosity and higher fracture strength. It can be seen from Fig. 3 that the SiC particles have been bonded each other by DC249 pyrolysis products at the contact sites and the residual pores are obvious. According to refs. [5] and [13], the joining mechanism may involve the direct formation of chemical bonds between SiC particles and the pyrolysis residual materials. As shown in Fig. 3, when DC249 content was increased in the starting mixtures, the bonding areas between SiC particles are larger, leading to higher strength. On the other hand, with more DC249, more space in the bulk specimens was occupied by the pyrolysis residue and, consequently, open porosity drops. Table 1 also shows the comparison between the figures in ref. [5] with our results. Although open porosity in this study is a little higher than that in ref. [5], the fracture strength of porous SiC ceramics in this study is 1.31 times of that in ref. [5], which is due to the higher ceramic yields of DC249 compared with that of PCS.

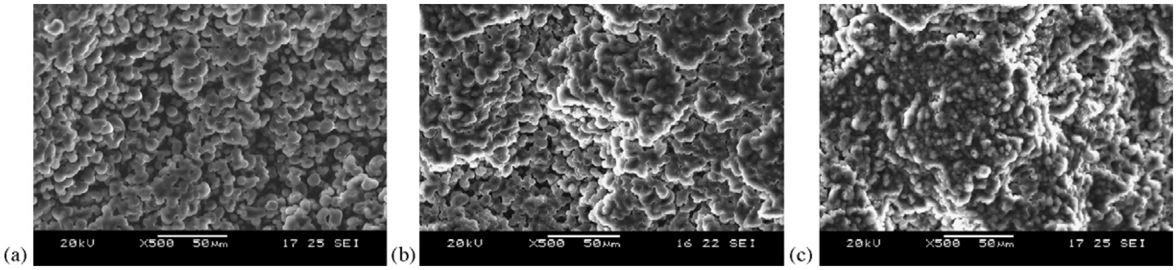

Fig. 3. SEM images of fracture surfaces of porous SiC ceramics with (a) 5 wt.%, (b) 10 wt.%, (c) 15 wt.% DC249 in starting mixtures (10 µm SiC powders and 15 MPa shaping pressure).

Table 1
Comparison of the properties of porous SiC ceramics with different content of DC249 and different particle size of SiC powders in the starting mixture

DC249 content (wt.%)	Mean size of SiC powders (μm)	Open porosity (%)	Fracture strength (MPa)	Average pore size (μm)
5	0.4	45.7	6.07 ± 0.19	–
	10	49.8	3.33 ± 0.18	1.21
10	0.4	43.1	18.59 ± 2.47	0.31
	10	46.2	14.10 ± 0.19	1.23
15	0.4	41.2	21.49 ± 1.35	–
	10	45.1	20.62 ± 1.12	1.39
	0.9[a]	44.7[a]	15.79 ± 2.35[a]	–

[a] From ref. [5] with a shaping pressure of 40 MPa and PCS as binder.

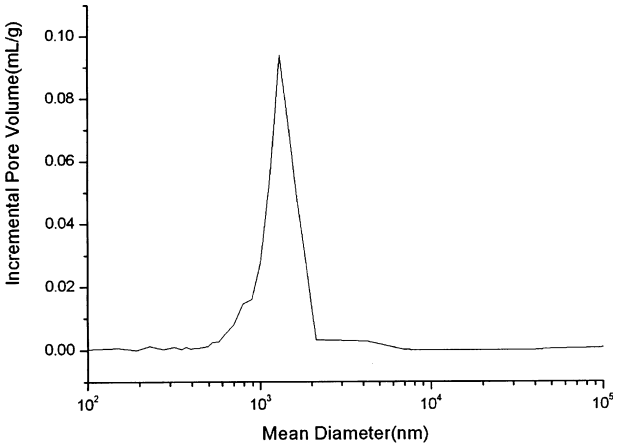

Fig. 4. Pore size distribution curve of porous SiC specimens fabricated by using starting SiC powders with a mean particle size of 10 μm where DC249 content is 5 wt.%.

Porous ceramics with designed pore structures are necessary for concrete application. As can be seen from Table 1, the average pore diameter in porous SiC ceramics becomes larger with increasing DC249 content and mean particle size of SiC powders. Fig. 4 shows the pore size distribution of porous SiC ceramics fabricated by using starting SiC powders with a mean particle size of 10 μm where DC249 content is 5 wt.%, which is representative of all the samples. It is clear that the pore size distribution is narrowly distributed.

4. Conclusion

In summary, Porous SiC ceramics were fabricated through the DC249 reaction bonding route at a temperature as low as 1000 °C. In this process, SiC particles were bonded to each other by DC249 pyrolysis products. Average pore size, open porosity and fracture strength of porous SiC ceramics were found to be functions of DC249 content and mean particle size of SiC powders.

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