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Short communication

# Preparation and characterization of SiC/cordierite composite porous ceramics

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**Abstract**

SiC/cordierite composite porous ceramics were prepared by a reactive process, in which clay, talc and alumina were mixed with SiC powder and graphite was used as the pore former. The mixture was heated in air so that graphite was burned out and SiC particles were bonded by reaction-derived cordierite. The sintering temperature was chosen as 1360 °C according to DTA and XRD studies. The volume fraction of graphite had a strong influence on porosity and strength. A three-point bending strength of 54.6 MPa was achieved at a porosity of 27.6%, while the strength of 9.3 MPa was corresponding to a porosity of 59.6%. Moreover, as-prepared composite porous ceramics exhibited excellent resistance to cyclic thermal shock.

*Keywords:* B. Composite; B. Porosity; D. SiC; D. Cordierite; C. Thermal shock resistance

## 1. Introduction

Porous ceramics have been extensively studied and used in various areas, including filters, catalyst supporters, electrodes, surgical implants, and etc. [1–5]. Due to their excellent mechanical and chemical stability, SiC porous ceramics have been a research focus in recent years [6–9]. However, because of the covalent nature of Si–C bond, sintering of SiC ceramics needs high temperatures (normally above 2000 °C), which has limited the practical applications of SiC porous ceramics. In order to lower the processing temperature, She and co-workers [10,11] have reported an oxidation bonding technique for the preparation of SiC porous ceramics at low temperatures. In addition, we have fabricated SiC porous ceramics by using the polycarbosilane as preceramic precursor to bond SiC particles at 1100 °C [12]. It is necessary to choose a suitable inter-particle phase between SiC particles in order to reduce the processing cost while retaining the good properties of SiC porous ceramics. Reactive processing is an effective way to fabricate ceramics at relatively low temperatures.

Cordierite ($2MgO{\cdot}2Al_2O_3{\cdot}5SiO_2$) is known to have a low coefficient of thermal expansion and low cost [13,14]. In the present work, clay, talc and alumina were used as starting materials for cordierite, which then bonded SiC particles to form a composite. Graphite powder was used as the pore former. The sintering behaviors, porosity and mechanical strength of the SiC/cordierite composite porous ceramics were investigated. Thermal shock resistance behavior under cyclic water quenching of the as-prepared porous ceramics was also evaluated. The thermal shock treatments were up to four cycles.

## 2. Experiments

Commercially available clay, talc and alumina were used as the starting materials and mixed according to the chemical composition of cordierite. After being ball-milled for 12 h, the above mixture was dried and then analyzed by an X-ray fluorescence spectrometer (PW2404, Philips) to determine the composition, which is listed in Table 1. The dried powder mixture was further mixed with SiC powder (mean particle size is 20 μm, Qinzhou Micropowders, China) by a weight ratio of 40:60 and ball-milled for 12 h. Moreover, 10, 20, and 40 wt.% of graphite powder was added in the above mixture respectively and was then kept milling for 1 h. After drying and passing through a 100 mesh sieve, the powders were pressed into rectangular bars under 50 MPa pressure using a stainless die. These specimens were then put into a box furnace at a heating

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Table 1
Composition of the powder mixture for cordierite synthesis

| Component | MgO | $Al_2O_3$ | $SiO_2$ | CaO | $TiO_2$ | $Fe_2O_3$ | Others |
|---|---|---|---|---|---|---|---|
| Fraction (wt.%) | 12.8 | 36.1 | 50.1 | 0.26 | 0.25 | 0.18 | 0.31 |

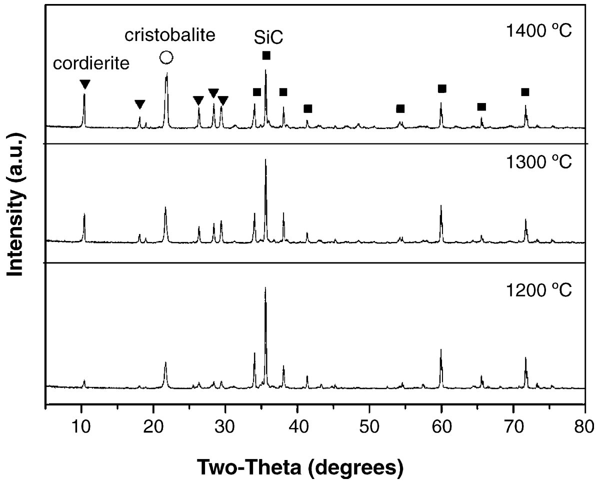

Fig. 2. XRD patterns of the specimens sintered at 1200, 1300, and 1400 °C, respectively.

rate of 1 °C/min below 900 °C to burn out graphite and 5 °C/min above 900 °C to the sintering temperature with a holding time of 2 h.

Sintering of the composite was investigated by DTA (DTA 404 PC, Netzsch) with a heating rate of 10 °C/min in air and XRD (RAX-10, Rigaku) with Cu Kα radiation. Open porosity was measured by the Archimedes method. Mercury intrusion porosimetry (PoreSizer 9320, Micromeritics) was adopted to determine pore size distribution. Specimens were machined into the dimension of 3 mm × 4 mm × 40 mm for the three-point bending test (AUTOGRAPH AG-I, Shimadzu) with a cross-head speed of 0.5 mm/min and a support distance of 30 mm. Five specimens were tested to obtain the average strength and standard deviation. Microstructures were observed by SEM (JSM-5600LV, JEOL).

Thermal shock test was conducted by the conventional water quenching method [15]. Briefly, the specimens were heated to 1000 °C with a holding time of 30 min in a box furnace before being quenched into a container of water at 20 °C and then dried in an oven at 110 °C until constant weight. These specimens were further used for bending test or the next quenching cycle.

## 3. Results and discussion

### 3.1. Sintering behaviors

From the DTA curve of the green powder without graphite (Fig. 1), two exothermal peaks can be seen at near 1285 and 1351 °C, respectively. The peak at 1285 °C is attributed to the formation of cordierite phase. As shown in Fig. 2, SiC and cristobalite peaks exist at all the three sintering temperatures, but obvious cordierite peaks can only be seen at the temperatures of 1300 and 1400 °C. At 1200 °C, the cordierite peaks are very weak. Compared to the peaks corresponding to cordierite at 1300 °C, those at 1400 °C have higher intensity. The enhanced peak intensity is due to the better crystallization of cordierite, which is also confirmed by the exothermal peak at 1351 °C in the DTA curve (Fig. 1). However, SiC peaks were weakened as a result of excessive oxidation at 1400 °C. Based on the DTA and XRD studies, 1360 °C was selected as the sintering temperature for the SiC particles to be bonded to each other by the reaction-derived cordierite.

### 3.2. Porosity and mechanical strength

The porous ceramics with higher porosities can be obtained by increasing the content of the added graphite powder. In this work, the bending strength is tested on the specimens with various porosities. Fig. 3 shows the SEM image of a typical fracture surface of the composite porous ceramics with the

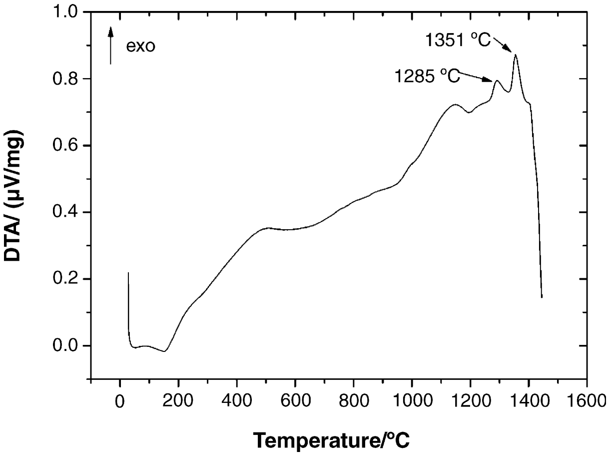

Fig. 1. DTA curve of the dried starting powder mixture without graphite at a heating rate of 10 °C/min in air.

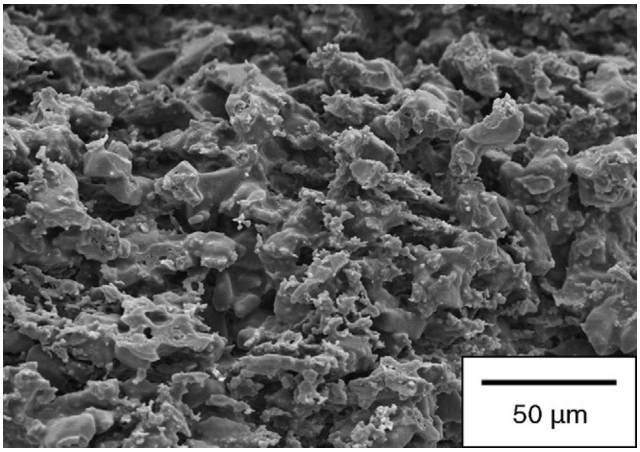

Fig. 3. SEM image of a porous specimen with the porosity of 59.6%, which was prepared by adding 40 wt.% of 9.7-μm sized graphite powder into the starting mixture.

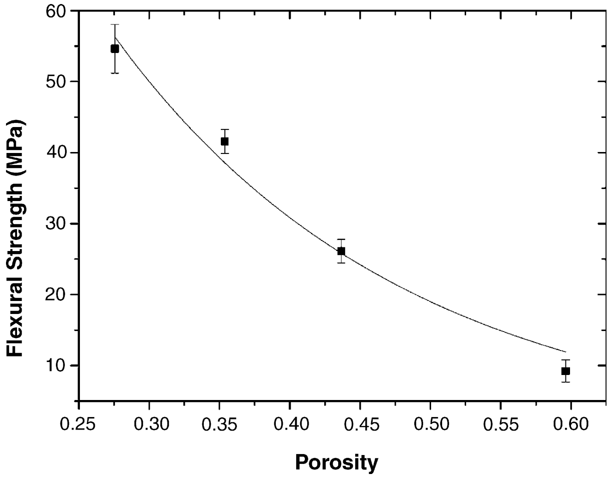

Fig. 4. Strength as a function of porosity for the porous specimens, in which a 9.7-μm sized graphite powder was used as the pore former.

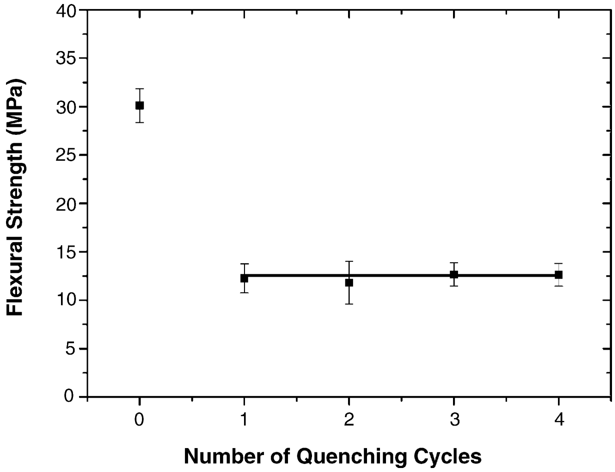

Fig. 6. Flexural strength of the porous specimens under cyclic thermal shock treatments from 1000 °C to 20 °C.

porosity of 59.6%. The strength of 54.6 MPa is achieved at a porosity of 27.6%, and that of 9.3 MPa is corresponding to a porosity of 59.6%. As shown in Fig. 4, when the porosity increases, the strength drops exponentially. The relation between strength and porosity of the specimens fits well with the equation given by Rice as follows [16]:

$$\sigma = \sigma_0 \exp(-bP) \tag{1}$$

where $\sigma$ is the strength of specimens at a porosity $P$, $\sigma_0$ the strength of those without porosity, and $b$ a constant depending on the pore structure and the material composition. In this work, $\sigma_0 = 213.4$ MPa and $b = 4.8$ are obtained by fitting the experimental data with Eq. (1).

Furthermore, it is interesting that the SiC/cordierite composite porous ceramics have a bimodal pore size distribution curve in Fig. 5. The smaller pores with the size of 1.9 μm are obtained by the stacking of ceramic powder, and the larger ones of 4.9 μm result from the burnout of graphite. However, the size of larger pores is smaller than that of graphite powder, which has a mean particle size of 9.7 μm. Since the specimens have almost no shrinkage after sintering, the above result might be attributed to the fact that the pore sizes measured by mercury porosimetry are those of pore necks rather than cavities [17].

### *3.3. Thermal shock resistance*

The specimens with initial strength of 30.1 MPa at a porosity of 43.4% were adopted for thermal shock tests. Fig. 6 shows the flexural strength of the composite porous ceramics after cyclic thermal shock treatments by being quenched from 1000 °C to 20 °C. After the first quenching cycle, the strength dropped to 12.3 MPa, but the strength does not continue to decrease with the increase of quenching cycles.

The similar phenomenon was previously observed in oxidation bonded SiC porous ceramics [18]. The reason can be attributed to the porous nature of the materials. During the

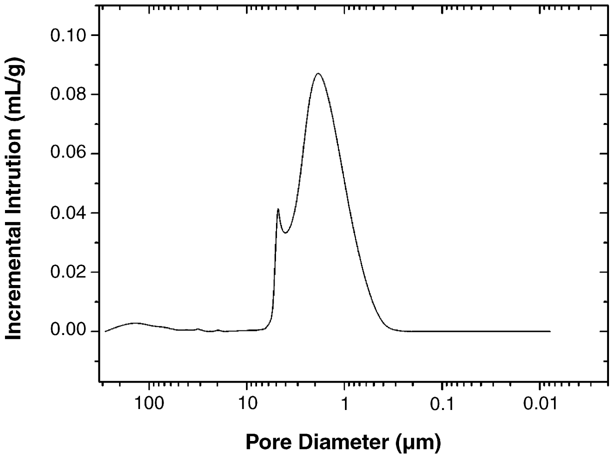

Fig. 5. Pore size distribution of a porous specimen prepared using a 9.7-μm sized graphite powder as the pore former and sintered at 1360 °C for 2 h.

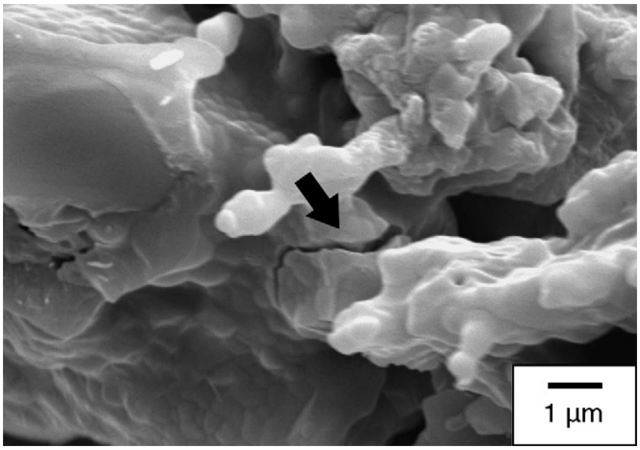

Fig. 7. SEM image of a specimen with 43.4% porosity after four cycles of water quenching from 1000 °C to 20 °C.

first quenching cycle, cracks caused by thermal stress originate at the weaker necks of the porous skeleton. When further quenching treatments are applied with same temperature difference, the thermal stress is no larger than that exists in the first cycle. The stronger necks cannot be cracked and the existing cracks will not propagate in the porous body, which is different from the situation in dense ceramics. As shown in Fig. 7, the crack terminates when it reaches the pore, despite being quenched repeatedly. Because the increasing of quenching cycles does not affect the mechanical strength of the samples, the as-prepared porous ceramics are promising to be used under cyclic thermal shock conditions.

## 4. Conclusions

A reactive processing method has been adopted for the preparation of SiC/cordierite composite porous ceramics. SiC particles are bonded to each other by corderite, which is synthesized from the starting materials of clay, talc and alumina. Graphite powder is used as the pore former to change the porosity of the specimens. Sintering of the porous ceramics is carried out at 1360 °C for 2 h. The as-prepared porous ceramics have a strength value up to 54.6 MPa at 27.6% porosity. It has been shown that the SiC/cordierite composite porous ceramics have an excellent resistance to cyclic heat shock treatment due to the unavailability of crack propagation in the porous body.

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