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# Fabrication and microstructure of porous SiC ceramics with $Al_2O_3$ and $CeO_2$ as sintering additives

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ABSTRACT

In the present study, porous silicon carbide ceramics were prepared via spark plasma sintering at relatively low temperatures using $Al_2O_3$ and $CeO_2$ as sintering additives. Sacrificial template was selected as the pore forming mechanism, and gelcasting was used to fix the slurry in a short time. The evolution process of the microstructures during different steps was observed by SEM. The influence of the sintering temperature and sintering additives on the shrinkage and porosity of the samples was studied. The microstructures of different samples were characterized, and the mechanical properties were also evaluated.



## 1. Introduction

With low density, low thermal expansion coefficient, excellent thermal shock resistance and oxidation resistance, high specific area, porous silicon carbide (SiC) ceramics have attracted more and more attention in recent years [1–4]. Because of the combination of these superior properties, porous SiC ceramics can be used for many applications, such as catalyst supports, filters for hot gases, and filters for molten metals, et al. [5–8].

High porosity is needed, but the pore walls or struts of porous SiC ceramics should be densified to improve the properties of them. However, due to the strong covalence bond and low self-diffusion coefficient of SiC, the densification process is so difficult that the sintering temperature usually reaches 2000–2200 °C. Adding sintering additives is an effective way to decrease the sintering temperature by accelerating the mass diffusion rate [9]. Liang et al. [10] studied the wettability of $Al_2O_3$ and $CeO_2$ on the surface of SiC ceramics. Using this system as additives, SiC ceramics with relative density larger than 99% was fabricated at temperature as low as 1840 °C. Pressureless sintering was usually adopted to prepare the porous ceramics [11]. Spark plasma sintering (SPS) is an effective way to get densified samples, because it has a very fast ramping rate as well as internal heat transfer induced by electric field. It can also be used in the preparation of porous ceramics [12].

In the present study, emphasis was focused on the preparation of porous silicon carbide ceramics via SPS technique at relatively low temperatures using $Al_2O_3$ and $CeO_2$ as sintering additives. The influence of the sintering temperature on the shrinkage of the samples was studied. Sacrificial template was selected as the pore forming mechanism, and microbeads were used to control the pore size exactly. The microstructures were characterized, and the mechanical properties were also evaluated.

## 2. Experimental

SiC powders were supplied by Xinyuan Micropowders Co., Ltd., (Shandong, China), and the index was shown in Table 1. $Al_2O_3$ powders (99.8%, $D_{50}=0.6$ μm, Almatis Inc., Qingdao, China) and $CeO_2$ powders (99.95%, < 100 nm, Sigma-Aldrich Co., Sweden) were selected as sintering additives. The amounts of $Al_2O_3$ powders and $CeO_2$ powders were both 3.5 wt% of SiC powders, which was similar with [10]. Tetramethylammonium hydroxide (TMAH, 25 wt% solution) was used as dispersant. Polymethyl methacrylate (PMMA) microbeads with an average diameter of 5 μm were used as template materials. For gelcasting system, acrylamide (AM), N,N′-methyleneacrylamide (MBAM), N,N,N′,N′-tetramethylethylenediamine (TEMED) and ammonium persulfate (APS) were selected as monomer, cross linker, catalyst and initiator, respectively.

Except APS, all the raw materials and deionized water were put into a polyurethane jar, together with SiC balls. The slurry was

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**Table 1**
The index of the silicon carbide powders.

| $D_{50}$ (μm) | BET ($m^2$/g) | Composition % | | | | |
|---|---|---|---|---|---|---|
| | | SiC | $Fe_2O_3$ | $SiO_2$ | F.C. | F.Si. |
| 0.50 | 8–10 | > 99 | < 0.05 | < 0.25 | < 0.12 | < 0.2 |

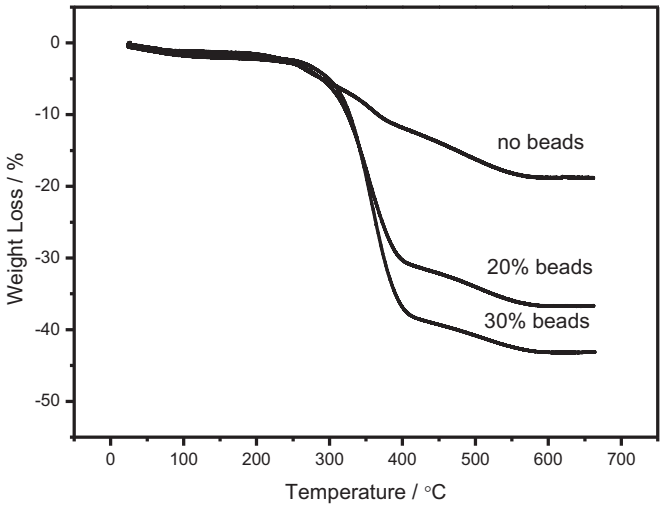


**Fig. 1.** Weight loss of the green bodies with different amounts of microbeads.

obtained after milling for 60 min at the rate of 300 r/min. After that, APS (30 wt% solution) was added into the stirring slurry. Several seconds later, the slurry was poured into a plastic mold and solidified at room temperature for 24 h. The solidified samples were dried under controlled conditions (at 30 °C and 90% relative humidity for 24 h, and then at 70 °C and 100 °C in common drying cabinet for 24 h and 24 h, respectively). Then the dried samples were located into a muffle furnace to debind the organics. After debinding, the rapid pressureless sintering was conducted in an SPS set-up (Dr. Sinter 2050, Sumitomo Coal Mining Co., Tokyo, Japan) under vacuum. The green bodies were located in a covered cylindrical graphite crucible containing powder bed (the mixtures of 7 wt% $Al_2O_3$–7 wt% $CeO_2$–86 wt% SiC powders). After the chamber was evacuated, the samples were heated to 1600 °C at the rate of 100 °C /min, and then to the final temperature at the rate of 40 °C /min and maintained for 5 min

To avoid cracks, thermal gravimetric analysis (TGA) was performed by TG Instruments (Perkin Elmer TGA 7) to set a good ramping schedule for debinding process. The porosity of the samples was measured by Archimedes method. The linear shrinkage after sintering process was calculated by measuring the lengths. The compressive strength of the sintered samples was measured. The microstructure of the sintered samples was characterized by scanning electron microscopy (JSM-7000F, JEOL, Tokyo, Japan).

## 3. Results and discussion

The rapid exhaust of microbeads and the organics for gelcasting might induce crack in the samples during the debinding process. Therefore, thermogravimetry was used to analyze the weight loss of the samples with different amounts of microbeads in air atmosphere, as depicted in Fig. 1. For all the four samples, the weight loss from room temperature to 250 °C was only about 3%. The differences were very evident from 250 °C to 400 °C, the trends of weight loss from 400 °C to 580 °C were very similar, and the weight losses between 580 °C and 660 °C were all nearly zero. That was to say, the exhaust of the organics for gelcasting took place between 250 °C and 580 °C, and the weight loss was mild and steady. The exhaust of the microbeads mainly occurred between 250 °C and 400 °C.

According to this, the ramping schedule for debinding was designed. First, the samples were heated to 200 °C at the rate of 1 °C/min. Second, they were heated to 400 °C at the rate of 0.5 °C/min and maintained for 30 min Then, they were heated to 600 °C

**Table 2**
Linear shrinkage, porosity and compressive strength of the samples.

| Sintering temperature (°C) | Linear shrinkage (%) | Porosity (%) | Compressive strength (MPa) |
|---|---|---|---|
| 1600 (20% beads) | 0.42 | 80.1 | $2.3 \pm 0.6$ |
| 1700 (20% beads) | 1.41 | 79.9 | $5.6 \pm 1.1$ |
| 1800 (20% beads) | 6.85 | 76.1 | $29.2 \pm 3.8$ |
| 1800 (30% beads) | 7.12 | 80.7 | $23.4 \pm 3.2$ |
| 1800 (20% beads, no additives) | 0.40 | 80.1 | $2.8 \pm 0.9$ |