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Development of manufacturing method for NITE-porous SiC ceramics using decarburization process

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Abstract

The objective of this work is to develop manufacturing method of porous SiC ceramics with mechanical and chemical stability. The nanoinfiltration transient eutectic (NITE)-porous SiC ceramics were fabricated by decarburization of the hot-pressed NITE-SiC including carbon particles. Porosity could be controlled at less than ±0.5% by the amount of carbon particles. The NITE-porous SiC ceramics exhibited a substantially high strength in comparison with other conventional porous SiC ceramics, due to its robust microstructure consisted of spherical pores.

Keywords: SiC; Porosity; Mechanical properties; NITE

1. Introduction

Recently, there has been an increasing interest in the applications of porous ceramics as hot-gas or molten-metal filters, catalyst supports, battery electrodes, heat insulators, ion exchangers, gas sensors and water cleaners. In particular, porous SiC ceramics are considered as functional materials of sustainable and advanced energy systems, such as perforated containment wall or flow channel inserts (FCIs) for blanket module of fusion reactor and inner/outer tube of a coated particle type fuel compartment for horizontal flow cooling concept with directly cooling system on gas-cooled fast reactor, because of their low thermal-expansion coefficient, low thermal conductivity and good thermal-shock resistance as well as excellent physical and chemical stability at elevated temperature.[1–3] FCIs made of a SiC fiber reinforced SiC matrix composite were first proposed by Tillack and Malang[4] as a means for electrical insulation between the flowing liquid metal and the load-carrying channel walls to reduce the magnetohydrodynamic pressure drop in the dual-coolant lead lithium blanket channels of a fusion power reactor. The main attraction of the FCIs is that SiC fiber reinforced SiC matrix composite has relatively low electrical conductivity, allowing for sufficient reduction of the induced electric currents by decoupling the liquid metal flow from the walls. Another potential advantage of the FCIs is related to low thermal conductivity of the SiC fiber reinforced SiC matrix composite, which allows for the reduction of heat losses from the breeder and therefore high bulk temperatures at the blanket exit, making the overall thermal efficiency of the blanket higher.[5–9] However, manufacturing method for a SiC fiber reinforced SiC matrix composites is very difficult. On the other hand, porous SiC is manufactured through a simple procedure than the SiC fiber reinforced SiC matrix composite. In addition to, porous SiC ceramics has relatively low electrical and thermal conductivity. For these reason, porous SiC ceramics is one of a candidate material as the FCIs in the dual-coolant lead lithium blanket module.

A number of manufacturing approaches have been applied to fabricate porous SiC including polymer pyrolysis, oxidation bonding and reaction bonding.[10–15] However, their processes are complicated and conventional porous SiC shows insufficient physco-chemical stability under high temperature environment. Therefore, from the view point of safety and stability, it is necessary to develop an uncomplicated manufacturing method and to investigate mechanical properties of porous SiC ceramics. In this study, porous SiC ceramics have been manufactured based on the nanoinfiltration transient eutectic process (NITE process), which is developed as a processing technique for high performance a SiC fiber reinforced SiC matrix composite.[16] The manufactured

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NITE-porous SiC ceramics had mechanical and chemical stability. As for this material, an application to several advanced energy systems is expected.

2. Experimental procedure

2.1. Materials

SiC nanopowder (β-SiC, purity > 99.0%, mean particle size 50 nm, HeFei, China) and carbon powder (mean particle size 80 nm, Cancarb, Canada) were used as the starting materials. For sintering additives, we used Al_2O_3 (purity > 99.99%, mean particle size 0.3 μm, Kojundo Chemical Lab. Co. Ltd., Japan) and Y_2O_3 (purity > 99.99%, mean particle size 0.4 μm, Kojundo Chemical Lab. Co. Ltd.). Powders were wet-milled in isopropanol for 12 h using Planetary Ball Mill (Fritsch, Germany). The slurry was dried and sieved through a 100 μm screen. Mixed powders were hot-pressed under a pressure of 20 MPa at 1900 °C for 1 h with a heating rate of 10 °C/min. A typical hot-pressing schedule is reported in Fig. 1. Sintering atmosphere was argon.

2.2. Decarburization

Carbon particles can be removed by reaction with oxygen according to the following equations:

$$C(s) + O_2(g) \rightarrow CO_2(g) \quad (1)$$

$$2C(s) + O_2(g) \rightarrow 2CO(g) \quad (2)$$

In this research, carbon particles which were added to sintered body were burned out by decarburization process in air at 700 °C. A schematic of the fabrication process for NITE-porous SiC ceramics is given in Fig. 2.

2.3. Microstructure and mechanical properties

Bulk density of NITE-porous SiC ceramics was measured by the Archimedes method, using distilled water. Porosity was calculated from relative density and theoretical density, which were obtained by the rule of mixture. The pore shape and size distribution were examined by optical and scanning electron microscopy

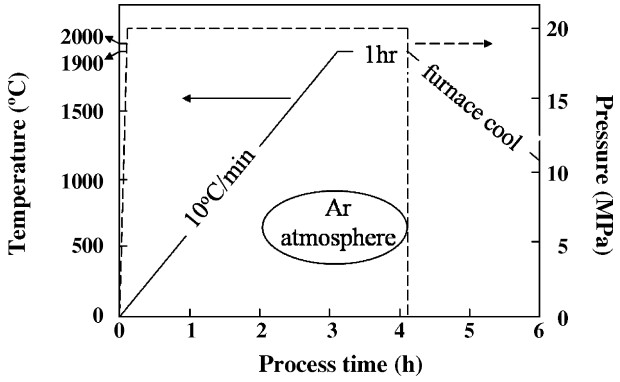

Fig. 1. Typical hot-pressing schedule.

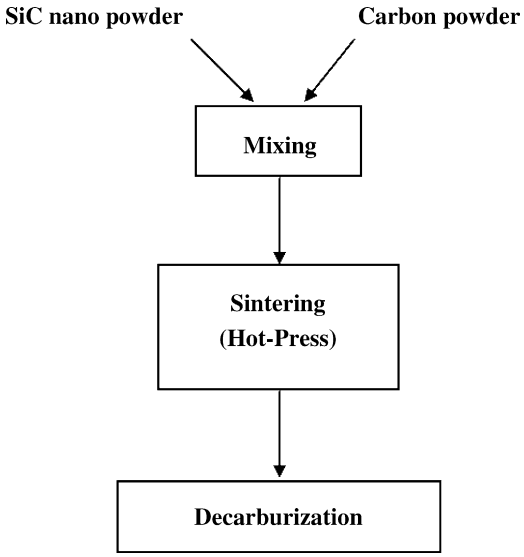

Fig. 2. Schematic of the fabrication process.

(FE-SEM, Model JSM-6700F, JEOL, Japan). Flexural strength of NITE-porous SiC ceramics was measured using three-point bend testing. Rectangular bar 25.0 mm × 4.0 mm × 2.5 mm were prepared by grinding and cutting. The tensile faces of the bars were subsequently polished down to 1 μm diamond polish. The tensile edges were beveled to decrease the effect of edge cracks. All tests were conducted on a conventional screw-driven loading frame (Model 5581, Instron, UK), with a crosshead speed of 0.5 mm/min, using a three-point bending jig of 18 mm support span, at room temperature. Elastic modulus of the NITE-porous SiC ceramics was measured using tensile testing which was also conducted on the Instron machine.

3. Results and discussion

3.1. Weight loss during decarburization

Weight change of the carbon particles during decarburization process is shown in Fig. 3. Decarburization speed became fast with increase of amount of carbon particles. In the case of the specimen which carbon particles of 10 vol.% was added in, a

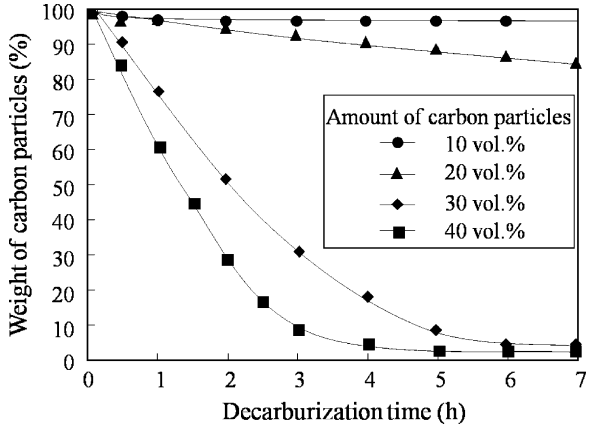

Fig. 3. Weight change of the carbon particles during decarburization process.

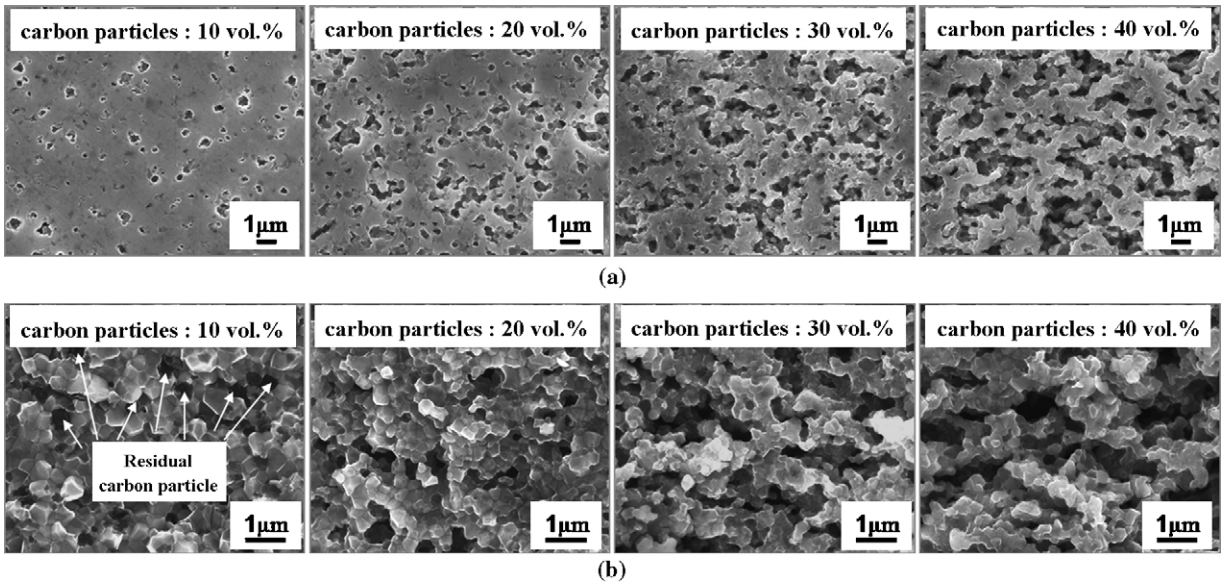

Fig. 4. Microstructure of NITE-porous SiC ceramics: (a) surface of specimen and (b) inside of specimen.

weight loss of carbon particles was not able to be found after 1 h. A rate of weight loss was about 3%. It means that decarburization process was performed at only surface of specimen. In other words it is predicted that carbon particles of the specimen inside are left. In the case of 20 vol.% carbon particles, it took about 100 h for carbon particles to decarburize more than 95%. However, in the case of specimens added carbon particles of 30 and 40 vol.%, a rate of weight loss exceeded 95% after only 6 and 5 h, respectively.

Fig. 4 shows the microstructure of decarburized specimen during 100 h. Residual carbon particles could not be observed at the surface of all specimens. However, in the case of the specimen which carbon particles of 10 vol.% was added in, residual carbon particles were able to be found at the inside of specimen. During decarburization process, residual carbon particles at the inside of specimen could not contact with oxygen, because the carbon particles were surrounded by SiC matrix. On the other hand, in the case of the specimens added carbon particles of 20, 30 and 40 vol.%, residual carbon particles were not observed at not only surface but also inside of specimens. Additionally, it was confirmed that most of the pores in the NITE-porous SiC ceramics were formations of the open pore.

3.2. Porosity

Porosity of the NITE-porous SiC ceramics is shown in Fig. 5. Bulk density of NITE-porous SiC ceramics was measured by the Archimedes method, using distilled water. The relative density was obtained by normalizing the bulk density with the theoretical density of the material. The theoretical density was calculated by applying the rule of mixtures to the volume fraction for the constituents. As shown in Fig. 5, porosity of the P30SiC, P40SiC and P50SiC were typically 30.2 ± 0.4, 40.3 ± 0.3 and 49.7 ± 0.5 g/cm^3, respectively. In other words porosity of the NITE-porous SiC ceramics could be controlled at less than $\pm 0.5\%$ by the change of amount of carbon particles.

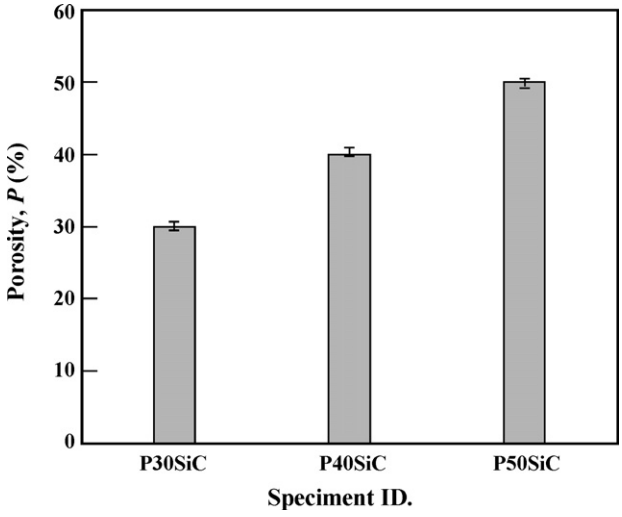

Fig. 5. Porosity of NITE-porous SiC ceramics.

3.3. Microstructures and pore-size distribution

Pore-size distributions of the NITE-porous SiC ceramics were determined by the image analysis method. Microstructures of the NITE-porous SiC ceramics were observed by scanning electron microscopy. Fig. 6 shows the microstructures and pore-size distributions of the NITE-porous SiC ceramics. Microstructures are cross-section surface of the specimens. All specimens were not able to observe residual carbon particles after decarburization process. It is means that most of the pores in the NITE-porous SiC ceramics are formations of the open pore.

Furthermore, as shown in Fig. 6, it was confirmed that mean pore size of the NITE-porous SiC ceramics is 0.5 μm. The mean pore size of the NITE-porous SiC ceramics was not changed by the porosity. Accordingly, pore-size of the NITE-porous SiC ceramics depends on the size of carbon particles instead of quantity of the carbon particles.

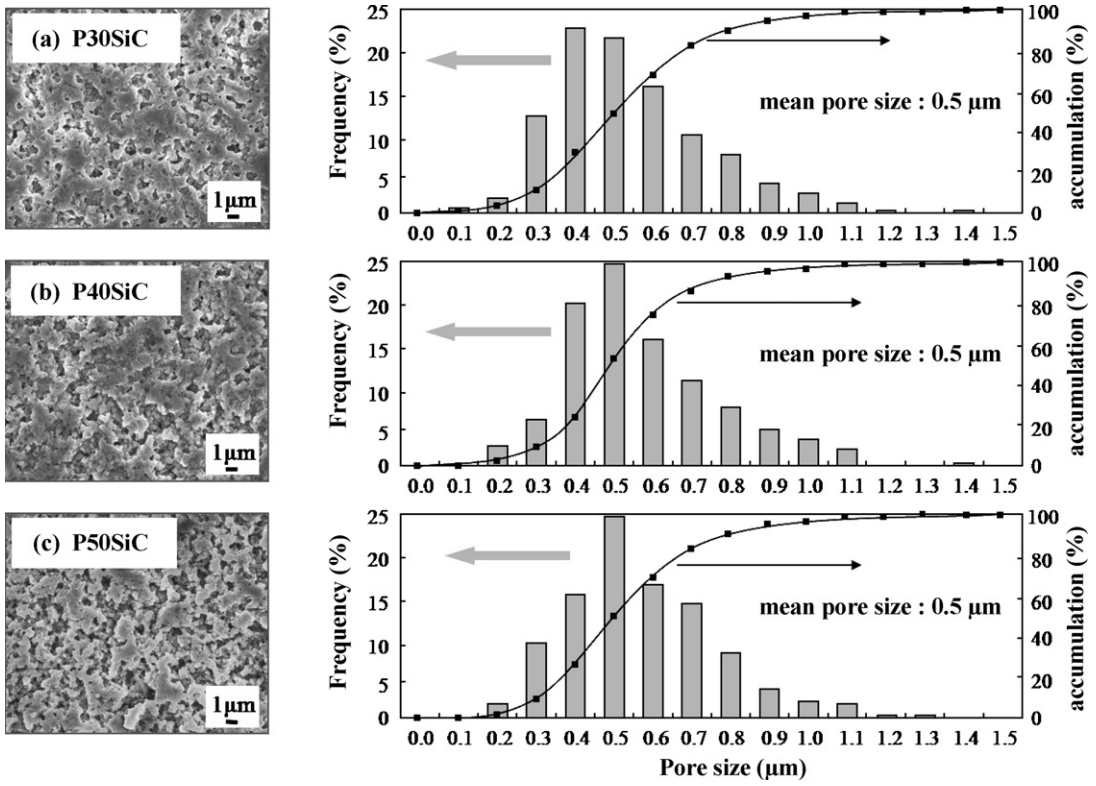

Fig. 6. Microstructures of NITE-porous SiC ceramics and pore-size distribution.

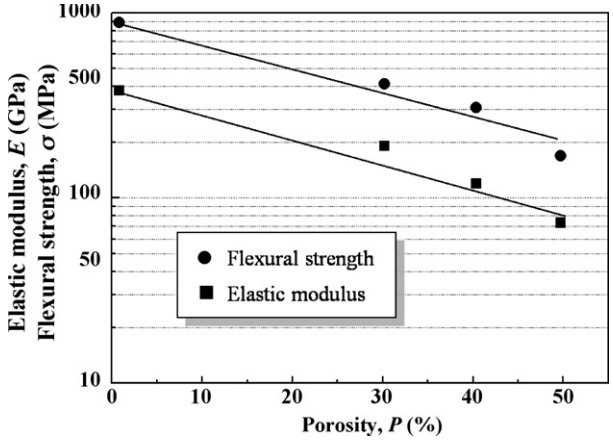

Fig. 7. Flexural strength and elastic modulus of NITE-porous SiC.

3.4. Mechanical properties

Flexural strength and elastic modulus of the NITE-porous SiC ceramics are shown in Fig. 7. Flexural strength of the NITE-porous SiC ceramics, which has porosity of 30, 40 and 50% was about 410, 305 and 170 MPa, respectively. Moreover, elastic modulus of the NITE-porous SiC ceramics, which has porosity of 30, 40 and 50% was about 190, 120 and 75 GPa, respectively. Generally, the mechanical properties of porous ceramics depend strongly on porosity. The mechanical properties–porosity dependence can be approximated by an exponential equation as follows,

$$\sigma = \sigma_0 \exp(-bP) \tag{3}$$

$$E = E_0 \exp(-bP) \tag{4}$$

where σ_0 and E_0 is the strength and elastic modulus of a non-porous structure, σ and E the strength and elastic modulus of the porous structure at a porosity P and b is a constant that is dependent on the pore shape. The values of b were reported to be 1.4 for cylindrical pores, 3 for spherical pores and 5 for solid spheres in cubic stacking.[17] The fit of this equation to the results in flexural strength gave $\sigma_0 = 972$ MPa and $b = 3.19$. By the elastic modulus, E_0 and b was 423 GPa and 3.24, respectively. The mechanical properties–porosity behavior of the NITE-porous SiC ceramics can be described by the following equation,

$$\sigma = 972 \exp(-3.19P) \tag{5}$$

$$E = 423 \exp(-3.24P) \tag{6}$$

As discussed above, main shape of the pores in the NITE-porous SiC ceramics can be viewed as a spherical pore. Consequently, the NITE-porous SiC ceramics exhibited a substantially high strength in comparison with other conventional porous SiC ceramics, due to its robust microstructure consisted of spherical pores.

4. Conclusion

Porous SiC ceramics have been manufactured based on the nanoinfiltration transient eutectic process, which is developed as a processing technique for high performance a SiC fiber reinforced SiC matrix composite. It is a simple and an economical manufacturing method. When the NITE-porous SiC ceramics was fabricated, carbon particles of more than 20 vol.% were required for the decarburization more than 95%. Porosity of the NITE-porous SiC ceramics could be controlled at less than ±0.5% by the change of amount of the carbon particles. Flexural strength of the NITE-porous SiC ceramics, which has porosity of 30, 40 and 50% was about 410, 305 and 170 MPa, respectively. Moreover, elastic modulus of the NITE-porous SiC ceramics, which has porosity of 30, 40 and 50% was about 190, 120 and 75 GPa, respectively. In other words, the NITE-porous SiC ceramics exhibited a substantially high strength in comparison with other conventional porous SiC ceramics, due to its robust microstructure consisted of spherical pores.

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