FULL PAPER

Effects of the size distribution of SiC powders on the microstructures and properties of liquid phase bonded porous SiC with neck bonding phases of $Y_4Al_2O_9$, $Y_3Al_5O_{12}$, $Y_2Si_2O_7$, and Al_2O_3

Sung Il YUN^{1,2}, Sahn NAHM^{1,†} and Sang Whan PARK^{2,‡}

¹Division of Materials Science and Engineering, Korea University, Seoul 02841, Republic of Korea

²Materials Architecturing Research Center, Korea Institute of Science and Technology, Seoul 02792, Republic of Korea

Liquid phase bonded (LPB) porous SiC with neck bonding phases consisting of yttrium aluminate ($Y_4Al_2O_9$, $Y_3Al_5O_{12}$), yttrium silicate ($Y_2Si_2O_7$), and Al_2O_3 were fabricated using varying amounts of an Al_2O_3 - Y_2O_3 - SiO_2 bonding additive in Ar at 1500 °C for 1 h. LPB porous SiC ceramics exhibited unimodal pore-size distributions, porosities of 36.6–44.8 %, and pore sizes of 7.7–8.5 μm . The particle-size distribution of SiC powders was an important factor in determining the pore characteristics, including pore-size distribution, pore shape, porosity, and pore size, and the flexural strength as well as the gas permeability of LPB porous SiC ceramics. The porosity and pore size increased, and the pore-size distribution narrowed by using SiC powders with a narrow size distribution. The flexural strength of porous SiC varied in the range of 39.7–66.7 MPa and was mainly dependent on the porosity, pore shape, pore size, and solid bonding area varied by the SiC particle-size distribution. A relatively high permeability ($1.28\text{--}1.84 \times 10^{-12} \text{ m}^2$) of LPB porous SiC was attained mainly due to the unimodal pore size distribution of pores with sizes of 7.7–8.5 μm .

©2021 The Ceramic Society of Japan. All rights reserved.

Key-words : Porous SiC, Reaction bonding, Flexural strength, Permeability, Particle size distribution

[Received June 7, 2021; Accepted August 17, 2021]

1. Introduction

Porous SiC has been widely utilized in essential industrial parts operating under harsh operation atmospheres such as in hot gas filters, including diesel particulate filters, vacuum chucks, and high-temperature catalytic supports because of its outstanding thermomechanical properties, such as high-temperature stability and mechanical strength, excellent thermal conductivity, good thermal shock resistance, and high chemical stability.^{1)–4)} Porous SiC have been developed to preserve distinctive properties such as gas permeability and thermomechanical properties tailored for specific applications. The normal sintering process of SiC usually requires a significantly high temperature above 2000 °C because of its high covalent bonded crystal structure.

Efforts have been made to develop cost-effective fabrication processes for porous SiC using oxide bonding phases such as mullite ($3Al_2O_3 \cdot 2SiO_2$), glass frits, SiO_2 , and cordierite ($2MgO \cdot 2Al_2O_3 \cdot 5SiO_2$) with relatively low melting temperatures or low-temperature sintering behavior.^{5)–18)}

Glass frit-bonded porous SiC were fabricated at temperatures below 1000 °C that exhibited high flexural strength

and high gas permeability with various pore sizes and porosities.^{5)–8)} A homogeneous porous microstructure with a unimodal pore-size distribution was developed using glass frit-bonded porous SiC fabricated with various particle sizes of SiC without using a pore-forming agent. The pore characteristics, including pore size, porosity, and pore-size distribution of glass frit-bonded porous SiC were easily varied by changing the SiC powders size and the contents of glass frit.

Mullite-bonded porous SiC has been intensively studied to improve the thermomechanical properties because mullite exhibits a high melting temperature and a thermal expansion coefficient similar to that of SiC.^{9)–18)} Mullite-bonded porous SiC was prepared using an in situ reaction bonding method in an air at a relatively low temperature (1250–1550 °C). A mullite bonding phase between SiC particles was synthesized by an in situ reaction of aluminum oxide and silicon oxide generated by oxidizing SiC particles during heat treatment in an air atmosphere. Mullite-bonded porous SiC was fabricated using various average size of SiC powders and aluminum sources with and without pore-forming agents as polymer beads and graphite powders. The pore size and porosity of the previously developed mullite-bonded porous SiC were widely diverse and seemed to be mainly controlled by the SiC particle size and pore-forming agents. The mechanical

[†] Corresponding author: S. W. Park; E-mail: spark@kist.re.kr

[‡] Corresponding author: S. Nahm; E-mail: snahm@korea.ac.kr

properties and permeability of mullite-bonded porous SiC varied widely depending on the processing conditions, aluminum sources, and SiC particle size.^{12)–18)}

Porous SiC for high-temperature applications have been fabricated by a partial liquid phase sintering (LPS) process using various sintering processes such as pressureless sintering, spark plasma sintering, and hot pressing in the temperature range of 1700–2000 °C.^{19)–29)} Fine SiC particles and various sintering additives such as Al₂O₃–Y₂O₃, Al₂O₃–CeO₂, AlN–Y₂O₃, and yttrium aluminate (Y₃Al₅O₁₂, YAG) were used as starting materials for the LPS porous SiC. LPS porous SiC with high porosity (>42 %) and an average pore size less than 1 μm prepared by hot pressing and SPS exhibited excellent flexural strengths over 95 MPa.^{22),24)} However, the appropriate control of pore size and porosity in LPS porous SiC is usually difficult because the fabrication temperature is too high to control the grain growth and the beta to alpha phase transition of SiC properly while maintaining the pore structure.

In this study, two different SiC powders, an as-received powders and a sieved powder, with different particle-size distributions were used as starting materials. Liquid phase bonded (LPB) SiC was prepared at 1500 °C using various amounts of an Al₂O₃–Y₂O₃–SiO₂ bonding additive. The effects of the size distribution of SiC powders on the microstructure, porosity, the pore size, and size distribution were investigated. The dependence of pore characteristics on the flexural strength and gas permeability was investigated to examine the effect of the particle-size distributions of SiC powders on the properties of LPB porous SiC.

2. Materials and method

α-SiC powders (DENSIC C #400, average particle size of 37 μm, Showa Denko, Japan) were used as starting materials. The bonding additives consisted of 0.5 μm of Al₂O₃ (Al-160GS-3, Showa Denko, Japan), 3 μm of Y₂O₃ (JB-3, Yeeyoung Cerachem. Ltd., Korea) and 9 μm of SiO₂ (Sukgyung AT, Korea) powders. Two different SiC powders were used to examine the effects of the size distribution of SiC powders on the pore characteristics, mechanical properties, and gas permeability of porous SiC: as-received and sieved SiC powders obtained by using 400 mesh (38 μm) and 500 mesh (25 μm). As presented in **Fig. 1**, the size distribution of SiC powders after completing the sieving procedure narrowed, and the average particle size (D_{50}) was reduced slightly from 37 to 35 μm, and particle-size distribution (D_{90}/D_{10}) decreased from 2.14 to 1.85. The composition of the Al₂O₃, Y₂O₃, and SiO₂ powders used as a bonding additive was 58.8:23.0:18.2 wt.%, which was selected according to the phase diagram of Al₂O₃–Y₂O₃–SiO₂ with a low melting temperature. **Table 1** shows the composition of AYS and SP-AYS fabricated by using as-received SiC powders and sieved SiC powders, respectively. The volume percent of Al₂O₃–Y₂O₃–SiO₂ was varied from 15 to 25 % in the green bodies. The Al₂O₃, Y₂O₃, and SiO₂ powders were preferentially mixed by ball milling for 12 h in a polyethylene jar with isopropyl alcohol

Fig. 1. The particle size distributions of as-received SiC powders and sieved SiC powders through 25 and 38 μm screen.

Table 1. The compositions of AYS and SP-AYS specimens

Specimen	Bonding additives (Vol.%)	Green body composition (wt.%)			
		SiC (37 μm)	Al ₂ O ₃ (0.5 μm)	Y ₂ O ₃ (3 μm)	SiO ₂ (9 μm)
AYS15	15	82.62	10.26	4.01	3.12
AYS20	20	77.04	13.55	5.29	4.12
AYS25	25	71.56	16.78	6.56	5.1
SP-AYS15	15	82.62	10.26	4.01	3.12
SP-AYS20	20	77.04	13.55	5.29	4.12
SP-AYS25	25	71.56	16.78	6.56	5.1

using Al₂O₃ balls, and then the mixed powders were dried at 70 °C for 12 h. The mixed bonding additive powders was obtained by sieving through a 325-mesh screen, and then SiC powders and the mixed bonding additive powders were simply mixed with a mortar and pestle with 5 wt.% of polyvinyl alcohol (PVA) solution (1 wt.% of aqueous solution). The mixtures of SiC and varying amounts of the bonding additive powders were uniaxially pressed into cylindrical green bodies (50 mm in diameter and 5 mm in thickness) at 30 MPa and then dried to remove the residual solvent in PVA solution at 80 °C for 12 h. The LPB porous SiC was fabricated by heat treatment of the green bodies at 1500 °C using a heating rate of 10 °C/min in an Ar atmosphere for 1 h in a graphite furnace.

The weights and dimensions of green bodies were measured to calculate bulk density. The theoretical densities of each specimen were obtained from the total weight and volume of SiC, Al₂O₃, Y₂O₃, and SiO₂. The volume of starting materials were calculated by dividing the weight by theoretical density of each starting materials. The relative densities of green bodies were calculated by dividing the bulk density by theoretical density. After the heating process, the weights and dimensions of sintered specimens were measured to evaluate the linear shrinkage rate and weight loss. The crystalline phases of sintered specimens were analyzed using an X-ray diffractometer (XRD, Bruker D8, Bruker) with Cu Kα radiation. The apparent porosity was evaluated using the Archimedes method in deionized water. Five samples (3 mm × 4 mm × 35 mm)

per each specimen were prepared to evaluate the flexural strength using the four-point bending test (Instron 4465, Instron Co., Ltd.) with a crosshead speed of 0.5 mm/min and an outer/inner span of 20 mm/10 mm. The microstructures of the fractured and polished surfaces were observed using field emission-scanning electron microscopy (Inspect F50, FEI) and backscattered electron (BSE) images. The composition of the bonding phase was analyzed by energy dispersive X-ray spectroscopy (EDS). The pore-size distributions were evaluated using a mercury porosimeter (MA-3000, Nippon Instruments Corporation, Japan). The gas permeability of sintered specimens (30 mm in diameter and 5 mm in thickness) were conducted by using capillary flow porosimetry (CFP-1200-AEL, Porous Materials Inc., USA) and capillary flow porometer software. By increasing the gas pressure after inserting the specimen between the top and bottom O-rings in the capillary flow porosimetry chamber, the gas flux and gas pressure drop were measured. The gas permeability was calculated using Darcy's law as follows:

$$K = \frac{v \cdot \mu \cdot X}{\Delta P} \quad (1)$$

where K is the permeability, μ is the dynamic viscosity of air, v is the fluid flow velocity of the gas, X is the thickness of the specimen, and ΔP is the pressure drop caused by the flow of gas through the specimens.

3. Result and discussion

The relative densities of AYS and SP-AYS green bodies sequentially increased from 59.6 to 62.5 % and from 57.9 to 62.2 %, respectively, as the amount of bonding additive increased, as shown in Fig. 2. The relative densities of AYS green bodies were higher than that of SP-AYS green bodies, because the wide size distribution of the SiC powders improved the packing density of green body.

Figure 3 presents the linear shrinkage rate and weight loss of AYS and SP-AYS after heat treatment at 1500 °C. The linear shrinkage rate increased with increasing amounts of bonding additive because the liquid phase

Fig. 2. The relative density of AYS and SP-AYS green bodies with a variation of the volume percent of bonding additives.

formed from the bonding additives shrinks in volume. As the amount of bonding additive increased, the amount of liquid phase formed between the SiC particles increased, promoting the shrinkage of the specimens to achieve a strong bonding between SiC particles. However, the shrinkage of specimens was limited by the close stacking of SiC particles, which resulted in the formation of pores among stacked SiC particles. The weight loss of AYS and SP-AYS was less than 2.24 wt.%, and it increased slightly as the amount of bonding additive increased. According to a previous study,³⁰⁾ the weight loss of the LPS SiC fabricated using the Al_2O_3 – Y_2O_3 – SiO_2 sintering additives was mainly owing to the evolution of gaseous species such as $SiO(g)$ and $CO(g)$ by the chemical reactions of SiC and SiO_2 in the liquid phase in the temperature range of 1500–1700 °C. The weight loss of AYS increased slightly compared to that of SP-AYS because the formation reaction of gaseous phases was enhanced by the increased interfacial area between SiC and the liquid phase as a result of the wide size distribution of the SiC powders used in AYS specimens.

Figure 4 presents the XRD patterns of AYS and SP-AYS specimens prepared using as-received SiC powders and sieved SiC powders at 1500 °C. LPB porous SiC ceramics with neck bonding phases consisting of yttrium aluminate ($Y_4Al_2O_9$, $Y_3Al_5O_{12}$), yttrium silicate ($Y_2Si_2O_7$), and Al_2O_3 were fabricated. Yttrium aluminate ($Y_4Al_2O_9$, $Y_3Al_5O_{12}$) and yttrium silicate ($Y_2Si_2O_7$) as secondary

Fig. 3. The linear shrinkage rate and weight loss of (a) AYS specimens and (b) SP-AYS specimens with a variation of the volume percent of bonding additives.

crystalline phases were newly synthesized, and a residual Al_2O_3 crystalline phase was observed in all specimens. The amount of these secondary crystalline phases increased with increasing the amounts of bonding additives. The formation of $\text{Y}_2\text{Si}_2\text{O}_7$ and $\text{Y}_3\text{Al}_5\text{O}_{12}$ was slightly enhanced in SP-AYS specimens, while the formation of $\text{Y}_4\text{Al}_2\text{O}_9$ was slightly increased in AYS specimens. The difference in XRD patterns between AYS and SP-AYS might be owing to the change in the distribution of Al_2O_3 , Y_2O_3 , and SiO_2 particles in green bodies with different particle size distributions of SiC powders. The formation of the yttrium aluminate and yttrium silicate was reported in LPS SiC using Al_2O_3 – Y_2O_3 sintering additives at temperatures as low as 1500°C ^{31)–33)} because SiO_2 on the SiC powders

surface was included in formation of the liquid phase. The liquid phase was primarily developed by the reaction of SiO_2 with Y_2O_3 at temperatures below 1500°C by using relatively large amount of SiO_2 ,³²⁾ and residual Al_2O_3 remained because of the limited dissolution of Al_2O_3 in the liquid phase at 1500°C .

Figure 5 presents the SEM microstructures of the polished surfaces of AYS and SP-AYS. The SiC particles were tightly bonded by the uniformly distributed bonding phase between the particles, and relatively uniform pore structures developed in the spaces produced by the stacking of SiC particles. With increasing the amounts of bonding additive, the pore size slightly decreased because of the increased neck-bonding area between the SiC particles, whereas the porosity significantly decreased, mainly because of the increased liquid phase, which filled small pores between the SiC particles. However, the SEM microstructures of the neck-bonding area and pores developed in SP-AYS specimens appeared different from those in AYS specimens. In SP-AYS specimens, thin neck-bonding phase was developed between large SiC particles because of the use of SiC particles with a narrow size distribution. Relatively sharp edges (mark A) around the pore boundary were formed in SP-AYS, which was mainly resulted from the change in the stacking of sieved SiC particles with a narrow particle size distribution. The pore size and porosity of SP-AYS appeared to be higher because of the reduced thickness of the neck-bonding area. **Figure 6** shows a high-magnification BSE microstructure image and EDS analysis of AYS25 specimen. The neck-bonding phase consisted of a continuous phase produced from the liquid phase formed in the Al_2O_3 – Y_2O_3 – SiO_2 bonding

Fig. 4. XRD patterns of AYS and SP-AYS specimens fabricated at 1500°C in an Ar atmosphere.

Fig. 5. BSE mode SEM images of polished surface of AYS and SP-AYS specimens.

additives (Part A, white) and dispersed residual Al_2O_3 particles (Part B, gray). The sizes of the residual Al_2O_3 particles increased significantly because of grain growth in the liquid phase consisting of Al_2O_3 , Y_2O_3 , and SiO_2 during the heat treatment procedure.

Figure 7 shows the pore-size distributions and average pore size of AYS and SP-AYS specimens with varying amounts of bonding additive. Unimodal pore-size distributions were obtained because the pore-wall structure was simply developed by infiltration of the liquid bonding phase into the green body structure consisting of stacked SiC particles. SP-AYS specimens using sieved SiC, which showed a narrower size distribution of pores and large pore size than AYS specimens. With increasing amounts of bonding additive, the average pore sizes of AYS were

reduced slightly from 7.9 to 7.7 μm , whereas those of SP-AYS were almost constant at approximately 8.5 μm . The difference in the pore characteristics of AYS and SP-AYS was mainly owing to the SiC particle-stacking structure in the green body as a function of the size distribution of the SiC particles used as starting materials. From these results, it appears that the average pore size and size distribution can be tailored by modifying the size distribution of SiC powders used as starting materials.

Table 2 shows the pore size, the porosity, and the flexural strength of LPB porous SiC prepared in this study. The porosity of AYS and SP-AYS steadily decreased from 41.9 to 36.6 % and from 44.8 to 38.9 %, respectively, as the amount of bonding additive increased from 15 to 25 vol.%. The porosity of AYS was lower than that of SP-AYS

Fig. 6. BSE mode SEM image with high magnification and EDAX analysis of polished surface of AYS25.

Fig. 7. The pore size distributions of (a) AYS specimens, (b) SP-AYS specimens and (c) the variation of average pore size as a function of the amount of bonding additives.

Fig. 8. BSE mode SEM images of fracture surface of AYS and SP-AYS specimens.

Table 2. Summarization of flexural strength, porosity, and average pore size of AYS and SP-AYS specimens

Specimens	Flexural strength (MPa)	Porosity (%)	Average pore size (μm)
AYS15	56.1	41.9	7.9
AYS20	64.7	40.3	7.8
AYS25	66.7	36.6	7.7
SP-AYS15	39.7	44.8	8.5
SP-AYS20	42.1	43.5	8.5
SP-AYS25	46.3	38.9	8.5

because of the formation of a thick pore wall consisting of a liquid phase that bonded tightly packed SiC particles with a wide particle size distribution. The flexural strengths of AYS and SP-AYS varied from 56.1 to 66.7 MPa and from 39.7 to 46.3 MPa, respectively, as the amount of bonding additive increased from 15 to 25 vol.%. As the amount of bonding additive increased, the flexural strength of AYS and SP-AYS increased owing to the decreased porosity accompanying with the increased bonding area. The flexural strength of LPB porous SiC fabricated using Al_2O_3 – Y_2O_3 – SiO_2 bonding additives at 1500 °C appeared higher than those of LPS porous SiC with pore sizes of 7.7–8.5 μm and similar porosities prepared using fine SiC particles and pore-forming agents as starting materials at high temperature (1850–1950 °C).^{26),29)} The flexural strengths of AYS were much higher than those of SP-AYS specimen, which was mainly due to the decreased porosity and pore size in AYS specimen depending on the amount of sintering addi-

tives. However, it appeared that the flexural strengths of SP-AYS specimen with similar porosity were much lower than those of AYS, although the amount of sintering additives used in SP-AYS specimen was higher than that in AYS specimens. The slight difference in the compositions of the neck bonding phase between AYS and SP-AYS might affect the flexural strength. However, the reduced flexural strengths of SP-AYS cannot be simply explained by the variation in the composition of the neck bonding phase, because both of $\text{Y}_2\text{Si}_2\text{O}_7$ with a low strength³⁴⁾ and $\text{Y}_3\text{Al}_5\text{O}_{12}$ with a high strength³⁵⁾ crystalline phases were simultaneously increased in the neck bonding phase of SP-AYS specimens. According to the previous study,^{36),37)} the pores with sharp edge severely reduced the flexural strength of porous SiC due to the stress concentration, and it was well known that the flexural strength of porous materials decreased by increasing pore size. As shown in Fig. 5, relatively large pores with sharp edges were produced in SP-AYS specimens. From these results, the flexural strengths of SP-AYS were reduced by the formation of relatively large size pores with sharp edges, rather than the slight differences in the composition of neck bonding phase. **Figure 8** presents the fracture surface of AYS and SP-AYS. The liquid phase developed from bonding additives was homogeneously distributed in the neck bonding area, and was also observed on the surface of SiC particles as droplets. The fracture of LPB porous SiC was occurred not only through the neck bonding interface, but also through the SiC particles. Although it was not easy to

Fig. 9. The exponential fitting of the flexural strengths of AYS and SP-AYS specimens as a function of porosity.

analyze precisely the solid bonding area from the fracture surface, the solid bonding area inferred from the fracture surface appeared to be increased in AYS compared to those observed in SP-AYS. Therefore, the lower flexural strength of SP-AYS specimens was partly due to the reduced solid bonding area as suggested by Li.³⁸⁾

The flexural strengths of AYS and SP-AYS exponentially decreased with increasing porosity (Fig. 9). According to previous studies based on the minimum solid area model in porous materials,³⁹⁾ the strength of porous ceramics is dependent on the porosity as an exponential equation.

$$\sigma = \sigma_0 \cdot \exp(-b \cdot P) \quad (2)$$

where σ and σ_0 are the strengths of a porous body at a particular porosity P and a pore-free dense body, respectively, and b is an empirical constant, which is dependent on pore characteristics such as pore structure, pore shape, and pore-forming process.

For the idealized porous materials, the value of b for brittle polycrystalline porous material is 3 for spherical pores with cubic stacking, 6 for spherical solids with cubic stacking and 9 for spherical solids with rhombohedral stacking owing to the change in the minimum solid contact area in the porous material.³⁹⁾ It was reported that the value of b for porous SiC ceramics varied widely from 3 to 8 depending on the fabrication process, porosity range, and neck-bonding materials.⁴⁰⁾ By linear regression fit of the variation of flexural strength versus porosity according to Eq. (2), the values of b for AYS specimens and SP-AYS specimens were estimated to be 7.45 and 2.93, respectively, indicating that the pore structures of the prepared AYS specimens and SP-AYS specimens were extremely dissimilar. From these results, the size distribution of SiC powders used as starting materials seemed to be a key factor in determining not only the pore characteristics but also the pore structure by changing the particle-stacking arrangement.

As shown in Fig. 10, the specific flow of AYS and SP-AYS prepared at 1500 °C were gradually increased with increasing pressure. The specific flow of AYS specimens

Fig. 10. The specific flow of (a) AYS specimens and (b) SP-AYS specimens as a function of pressure.

decreased slightly with increasing amounts of bonding additive up to 20 vol.%, after which the change was negligible. For SP-AYS specimens, the specific flow varied little regardless of the increase in the amount of bonding additives. It appeared that the specific flow of SP-AYS specimens with higher porosity and larger pore size increased compared to those of AYS.

Figure 11(a) shows the permeability of the specimens calculated from the results of the gas-flow experiments according to Eq. (1). The permeability of AYS specimens gradually decreased from 1.64×10^{-12} to $1.28 \times 10^{-12} \text{ m}^2$ as the amount of bonding additive increased, but the permeability of SP-AYS specimen was not dependent on the amount of bonding additive, and it remained at approximately $1.8 \times 10^{-12} \text{ m}^2$. The variations in permeability and pore size were replotted as a function of the porosity to assess the effects of porosity and pore size on the permeability in Fig. 11(b). The permeability of AYS was improved from 1.28×10^{-12} to $1.64 \times 10^{-12} \text{ m}^2$ with increasing porosity from 36.6 to 41.9 %. The permeability of AYS was increased mainly by increase of porosity. However, the permeability of SP-AYS remained constant at approximately $1.8 \times 10^{-12} \text{ m}^2$ regardless of the porosity variation from 38.9 to 44.8 %. The permeability of SP-AYS increased compared to that of AYS. The increase in

Table 3. Summarization of the permeability, porosity, and pore characteristics of porous SiC in previous studies

Materials	Sintering aid or bonding phase	Porosity (%)	Average pore size (μm)	Permeability (10^{-12}m^2)	Pore size distribution
This study SiC ⁽⁴³⁾	YAG, YAM, $\text{Y}_2\text{Si}_2\text{O}_7$	44.8	8.5	1.83	unimodal
	Glass	56	6	1.9	unimodal
		45	12	2.5	
SiC ⁽¹⁸⁾	Mullite	62	20	1.1	bimodal
SiC ⁽²⁶⁾	Al_2O_3 , Y_2O_3 , CaO	54.7	7	1.0	—
SiC ⁽⁴⁴⁾	Polysiloxane, Al_2O_3 , Y_2O_3	75	6.6	0.82	bimodal
SiC ⁽⁴⁵⁾	ZrO_2 , Mullite fiber	46.8	20.0	16.4	unimodal
SiC ⁽⁴⁶⁾	Glass	38	8.2	1.04	unimodal
SiC ⁽¹⁵⁾	Al_2O_3 , Magnesium carbonate/Mullite, Corundum	35.8	47.1	4.8	unimodal

Fig. 11. The variation of permeability versus the amount of bonding additives (a) and the variation of permeability and average pore size versus porosities (b).

the permeability of SP-AYS specimen was mainly due to the increase in the pore size. It is known that permeability is subject to the porosity, pore size, tortuosity, and other pore characteristics, such as pore interconnectivity and pore shape.^{(41),(42)} Recently, it was reported that the permeability of porous SiC with a unimodal pore-size distribution, porosities of 35–65 %, and pore sizes of 6–47 μm ,^{(15),(18),(43)} was reliant on the average pore size rather than porosity, whereas other studies have suggested that porosity was the main primary parameter to determine the permeability with a wide range of pore-size distributions. However, the dependency of the permeability of porous SiC on the porosity and pore size has not been systematically analyzed.

Table 3 summarizes the permeability value, porosity, pore size and the type of size distribution of porous SiC ceramics. The permeability of AYS and SP-AYS with similar pore sizes and porosities appeared to be higher than those of porous SiC fabricated using small SiC particles with pore-forming agents, but it was comparable to those of porous SiC prepared using large SiC particles without using pore-forming agents. Depending on the fabrication process, the pore structure of porous SiC developed in previous studies was characterized into two types: a uniform pore structure with a low deviation and unimodal pore-size distribution and a complicated pore structure with a wide and multimodal pore-size distribution. As shown in Table 3, porous SiC with unimodal pore-size distribution exhibited higher permeability compared to porous SiC with a wide and multimodal pore-size distribution, although the porous SiC ceramics had similar pore sizes and porosities. It could be suggested that the pore-size distribution is another significant factor for determining the permeability of porous SiC because the pore-size distribution might affect the interconnectivity of pores in porous SiC with relatively high porosity. The unimodal pore-size distribution observed in the prepared AYS and SP-AYS specimens might provide a beneficial effect to improve the permeability. Furthermore, it was suggested that the narrower pore size distribution of SP-AYS was partially involved in the increased permeability compared to those of AYS.

4. Conclusion

LPB porous SiC with neck bonding phases consisting of yttrium aluminate ($\text{Y}_4\text{Al}_2\text{O}_9$, $\text{Y}_3\text{Al}_5\text{O}_{12}$), yttrium silicate ($\text{Y}_2\text{Si}_2\text{O}_7$), and Al_2O_3 were fabricated varying amounts of an Al_2O_3 – Y_2O_3 – SiO_2 bonding additive in Ar at 1500 °C for 1 h. LPB porous SiC ceramics exhibited unimodal pore-size distributions, porosities of 36.6–44.8 %, and pore sizes of 7.7–8.5 μm . The particle-size distribution of the SiC powders used as the starting material was an important factor in determining the pore characteristics including pore-size distribution, porosity, average pore size and pore shape. The flexural strength and gas permeability closely depended on the pore characteristics. The flexural strength of SP-AYS decreased significantly compared to that of AYS, mainly due to the formation of large pores with relatively sharp edges, increased porosity, and reduced solid

bonding area. The relatively high permeability ($1.28\text{--}1.84 \times 10^{-12} \text{ m}^2$) was attained in LPB porous SiC with a unimodal pore size distribution. The permeability of SP-AYS increased compared to that of AYS because of the increased pore size and the narrow pore size distribution.

Acknowledgments This study was supported by a grant from the Fundamental R&D Program of the Ministry of Trade, Industry, and Energy, Republic of Korea and Korea Institute of Science and Technology, Republic of Korea.

References

- 1) S. Schaafhausen, E. Yazhenskikh, S. Heidenreich and M. Müller, *J. Eur. Ceram. Soc.*, **34**, 575–588 (2014).
- 2) T. Ohji and M. Fukushima, *Int. Mater. Rev.*, **57**, 115–131 (2012).
- 3) U. F. Vogt, L. Györfy, A. Herzog, T. Granule and G. Plesch, *J. Phys. Chem. Solids*, **68**, 1234–1238 (2007).
- 4) J. Adler, *Int. J. Appl. Ceram. Tec.*, **2**, 429–439 (2005).
- 5) A. Preston and G. Mueller, *Int. J. Appl. Ceram. Tec.*, **9**, 764–771 (2012).
- 6) S. H. Chae, Y. W. Kim, I. H. Song, H. D. Kim, J. S. Bae, S. M. Na and S. I. Kim, *J. Korean Ceram. Soc.*, **46**, 488–492 (2009).
- 7) C. Y. Liu, W. H. Tuan and S. C. Chen, *Ceram. Int.*, **41**, 4564–4568 (2015).
- 8) J. H. She, Z. Y. Deng, J. Daniel-doni and T. Ohji, *J. Mater. Sci.*, **37**, 3615–3622 (2002).
- 9) J. H. She, T. Ohji and Z. Y. Deng, *J. Am. Ceram. Soc.*, **85**, 2125–2127 (2002).
- 10) J. H. She, J. F. Yang, N. Kondo, T. Ohji, K. Shuzo and Z. Y. Deng, *J. Am. Ceram. Soc.*, **85**, 2852–2854 (2002).
- 11) J. H. She, T. Ohji and K. Shuzo, *J. Eur. Ceram. Soc.*, **24**, 331–334 (2003).
- 12) Y. H. Choi, Y. W. Kim, I. S. Han and S. K. Woo, *J. Mater. Sci.*, **45**, 6841–6844 (2010).
- 13) J. H. Eom, Y. W. Kim, S. K. Woo and I. S. Han, *J. Ceram. Soc. Jpn.*, **117**, 421–425 (2009).
- 14) Y. Jing, X. Y. Deng, J. B. Li, C. Y. Bai and W. K. Jiang, *Ceram. Int.*, **40**, 1329–1334 (2014).
- 15) Z. Y. Luo, W. Han, X. J. Yu, W. Q. Ao and K. Q. Liu, *Ceram. Int.*, **45**, 9007–9016 (2019).
- 16) S. Z. Abbas Bukhari, J. H. Ha, J. M. Lee and I. H. Song, *J. Eur. Ceram. Soc.*, **38**, 1711–1719 (2018).
- 17) C. Y. Bai, X. Y. Deng, J. B. Li, Y. N. Jing and W. K. Jiang, *Mater. Charact.*, **90**, 81–87 (2014).
- 18) S. Q. Ding, S. M. Zhu, Y. P. Zeng and D. L. Jiang, *J. Eur. Ceram. Soc.*, **27**, 2095–2102 (2007).
- 19) E. Gomez, J. Echeberria, I. Iturriza and F. Castro, *J. Eur. Ceram. Soc.*, **24**, 2895–2903 (2004).
- 20) B. Yuan, G. Wang, H. X. Li, L. F. Liu, Y. S. Liu and Z. Z. Shen, *Ceram. Int.*, **42**, 12613–12616 (2016).
- 21) S. Q. Ding, S. M. Zhu, Y. P. Zeng and D. L. Jiang, *Ceram. Int.*, **32**, 461–466 (2006).
- 22) M. Hotta, H. Kita, H. Matsuura, N. Enomoto and J. Hojo, *J. Ceram. Soc. Jpn.*, **120**, 243–247 (2012).
- 23) B. K. Jang and Y. Sakka, *Sci. Technol. Adv. Mat.*, **8**, 655–659 (2007).
- 24) Y. H. Park, J. S. Park, T. Hinoki and A. Kohyama, *J. Eur. Ceram. Soc.*, **28**, 657–661 (2008).
- 25) C. Soto, C. García-Rosales and J. Echeberria, *J. Eur. Ceram. Soc.*, **39**, 3949–3958 (2019).
- 26) J. H. Eom, Y. W. Kim and I. H. Song, *J. Eur. Ceram. Soc.*, **32**, 1283–1290 (2012).
- 27) J. H. Eom and Y. W. Kim, *J. Mater. Sci.*, **44**, 4482–4486 (2009).
- 28) J. H. Eom and Y. W. Kim, *Met. Mater. Int.*, **16**, 399–405 (2010).
- 29) J. H. Eom and Y. W. Kim, *J. Ceram. Soc. Jpn.*, **116**, 1159–1163 (2008).
- 30) F. Hue, Y. Jorand, J. Dubois and G. Fantozzi, *J. Eur. Ceram. Soc.*, **17**, 557–563 (1997).
- 31) P. Zhou, X. B. Yu, L. Z. Yang, S. P. Yang and W. J. Gao, *J. Lumin.*, **124**, 241–244 (2007).
- 32) Z. Sun, Y. C. Zhou and M. S. Li, *J. Mater. Res.*, **21**, 1443–1450 (2006).
- 33) A. Can, M. Herrmann, D. S. McLachlan, I. Sigalas and J. Adler, *J. Eur. Ceram. Soc.*, **26**, 1707–1713 (2006).
- 34) Z. Sun, Y. Zhou, J. Wang and M. Li, *J. Am. Ceram. Soc.*, **90**, 2535–2541 (2007).
- 35) H. Nozawa, T. Yanagitani, T. Nishimura and H. Tanaka, *J. Ceram. Soc. Jpn.*, **116**, 649–652 (2008).
- 36) S. I. Yun, M. R. Youm, S. Nahm and S. W. Park, *Ceram. Int.*, **47**, 11979–11988 (2021).
- 37) V. R. Salvini, V. C. Pandolfelli and D. Spinelli, *Rec. Adv. Porous Ceram.*, 171–199 (2018).
- 38) J. Li, H. Lin and J. Li, *J. Eur. Ceram. Soc.*, **31**, 825–831 (2011).
- 39) R. W. Rice, *J. Mater. Sci.*, **31**, 102–118 (1996).
- 40) R. W. Rice, *J. Mater. Sci.*, **31**, 1509–1528 (1996).
- 41) N. Nishiyama and T. Yokoyama, *J. Geophys. Res.-Sol. Ea.*, **122**, 6955–6971 (2017).
- 42) Y. B. P. Kwan, D. J. Stephenson and J. R. Alcock, *J. Porous Mat.*, **8**, 119–127 (2001).
- 43) B. Wang, H. Zhang, H. T. Phuong, F. Jin, J. F. Yang and K. Ishizaki, *Ceram. Int.*, **41**, 2279–2285 (2015).
- 44) I. H. Song, I. M. Kwon, H. D. Kim and Y. W. Kim, *J. Eur. Ceram. Soc.*, **30**, 2671–2676 (2010).
- 45) F. Han, Z. X. Zhong, Y. Yang, W. Wei, F. Zhang, W. H. Xing and Y. Q. Fan, *J. Eur. Ceram. Soc.*, **36**, 3909–3917 (2016).
- 46) S. C. Kim, Y. W. Kim and I. H. Song, *J. Eur. Ceram. Soc.*, **37**, 1225–1232 (2017).