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Effect of bentonite addition on fabrication of reticulated porous SiC ceramics for liquid metal infiltration

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Abstract

SiC with different particles and a clay mineral bentonite (montmorillonite) were mixed in water to prepare ceramic slurry. The slurry was then infiltrated high porous polyurethane sponge. Excess slurry was squeezed out to adjust ceramic rate in the infiltrated body. The pore walls were coated with ceramic mix after the infiltrated body was dried. The polyurethane containing SiC particles and bentonite was fired in a box furnace to burn out the polyurethane from the body at 500 °C for 30 min. The remaining porous ceramic bodies were sintered at elevated temperatures to give strength. SiC particles with bentonite surface coating took polyurethane pore forms after firing the sponge. Bentonite was both used as binder for ceramic slurry at room temperatures and the sintering additives at elevated temperatures. Therefore, increasing bentonite addition gives higher strength to the resulting ceramic performs.

Keywords: B. Porosity; B. Composite; Ceramic foam; Infiltration; SiC particles

1. Introduction

The interest in porous ceramics has grown rapidly in the recent years with the demands from new fields of applications; processing routes to develop unique structures are now under extensive investigation. The use of porous ceramics in thermal insulation, substrates for catalysts and filtration systems has been assessed for many years and the materials are still expanding into other areas. Reticulated porous ceramics (RPCs) are three-dimensional network structure. The most common applications of the RPCs are molten metal and diesel engine exhaust filters, catalyst supports due to their high permeability, resistance to chemical attack and structural uniformity [1–4].

The most common method of production of RPCs is the ‘‘polymeric sponge’’ process [5], invented by Schwartzwalder and Somers [6]. In the method, green RPCs consisting of a few filled cells were fabricated by coating a polyurethane sponge with viscous slurry. In order to remove excess slurry from sponge, passing through preset rollers is the commonly used technique. RPCs produced by this process are of low strength and fracture toughness, because very thin struts of ceramic structure may remain and triangular pores exist after the organic sponge is burned out, making them sensitive to structural stresses and limiting their structural applications [7,8]. Several techniques have been tried to improve the slurry coverage [9–13]. A second coating of slurry may be applied to fill in any flaws or thin areas in the first coating. As suggested by Schwartzwalder and Somers [6] and Brown and Green [14], the second coat may be directly applied on the dried original coating. The dried body including the sponge substrate is difficult to recoat by dipping, because dipping can wash away some of the previously deposited ceramic coating and weaken the structure. At the same time, the recoat slurry floated by gravity will lead to filled cells, especially near the bottom portion of the part. Zhu et al. [15] developed a new recoating method. First, the green body prepared by preset roller method is preheated to produce reticulated foam with enough handling strength after the sponge is burnt out.

Zhao et al. [16] fabricated SiC foam (through the polymer foam replication method) SiC particles–Al composites by squeeze casting technique. They revealed that the SiC foam reinforcement has effective restriction on the thermal expansion of the aluminum matrix. Chae et al. [17] produced porous

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silicon carbide ceramics by using sacrificial polymer microbeads for the fabrication of metal matrix composites.

In this study, bentonite addition was the key factor achieving dimensionally stable porous ceramic. This technique greatly improved the mechanical properties of RPCs. Not much study has been reported on the densification with no shrinkage on the structure and properties of reticulated porous silicon carbide ceramics.

2. Experimental procedure

The Replica technique was used to fabricate high porous ceramic foam for liquid metal infiltration using polyurethane sponges, Urosan Foam A.S., Istanbul, Turkey, Commercial SiC powders (30 μm mean particle size), BM BOR Teknolojileri, Kayseri, Turkey were used for ceramic foam production. Submicron bentonite powders, Karakaya Bentonit A.S., Ankara, Turkey were also used as binder. Chemical content of the bentonite is given in Table 1.

SiC particles with different bentonite rates were mixed within pure water in agate mortar for half an hour to prepare ceramic slurry. After adjusting viscosity, and PH value, the slurry was infiltrated to the reticulated polyurethane sponge. The infiltrated body was then rolled up to squeeze out excess slurry. All polyurethane pore surfaces were daubed with ceramic mixture after drying. The slurry infiltrated samples were placed in a chamber furnace (Protherm PLF 130/9) and fired at 500 °C for 30 min to burn out the polyurethane sponge. The remaining ceramic mixture was sintered at elevated temperatures to produce durable and highly porous ceramic perform. Final sintering causes strength increase of the ceramic foam which beneficial for liquid metal infiltration. Scanning electron microscope (SEM, JEOL, JSM-6700F, Japan) analysis was carried out to examine pore morphology and particle distribution. Elemental dispersive spectrometry (EDS) analysis was also done during SEM examination. Compressive tests were applied to the sintered ceramic foam samples with 40 mm in diameter and 60 mm in height universal testing device (DARTEC RK model max. capacity 250 kN). The phases in the sintered products were analyzed by X-ray diffraction (Rigaku, XRD D/MAX/2200/PC, Japan) using Cu Kα radiation and identified by X'Pert HighScore Report program.

3. Results and discussion

In this study, high porous (80–90 ppi) and reticulated ceramic foams (open porosity) has been fabricated and investigated. Production of high porous ceramic foams for the purpose of infiltration is also needed knowledge of the liquid metal infiltration mechanisms. It is essential to control type, rate and size of the porosity as well as surface properties during the ceramic foam production. For that purpose, it has been benefited from porous polyurethane sponge which has certain grade and type of porosity.

Table 1
Chemical content of bentonite (%).

SiO_2	Na_2O	MgO	Al_2O_3	K_2O	Fe_2O_3	CaO
60–62	2.5–3	1.8–2	17–18	0.9–9.5	3–3.5	3.5–4

SiC particles, various rate of bentonite powders and certain amount of water has been mixed to prepare ceramic slurry. Bentonite consisting of a high calcium concentration has been mixed with SiC particles. This is a non-swelling bentonite that offers substantially lower dry and hot strengths by comparison to sodium bentonite. These physical properties attributes, when used in moderation, will improve particle flowability.

3.1. Optimization of slurry infiltration process

Low viscosity slurries could easily penetrate the sponge pattern during squeezing and relaxation process, but retaining of the slurry in the pores of the polyurethane sponge is difficult. It is also difficult to infiltrate the high viscous slurry up to inner region of polyurethane sponge Therefore optimization of the slurry properties and infiltration conditions is essential. Factors affecting the slurry viscosity are water content and size of ceramic particle in mixture. Amount of ceramic powders in the slurry affects the ease of slurry infiltration as shown in Table 2. The infiltration is difficult for all slurry compositions if the water to solid rate is different from 1.

The next parameter examined is the amount of ceramic slurry penetrating into the sponge. Squeezing and relaxation method of the sponge and remaining amount of ceramic slurry within the sponge after slurry infiltration has been studied. The weight of infiltrated slurry per unit volume of the sponge given (loading of the sponge) in units of $g\,cm^{-3}$ is an important experimental variable. This is affected by the method of pressing the sponge inside the slurry both during infiltration and after infiltration, and number of repeated infiltrations. The weight of infiltrated slurry also determines like porosity, permeability and fracture strength of the ceramic foam. It has been found that loading of the sponge is independent of the size and shape of the sponge. For a given type of the sponge, it was tried to load 40 mm in diameter and 60 mm in height polyurethane foam and it has been found that when the dimension of the polyurethane changes, the slurry infiltration rates are similar.

3.2. Effect of sintering temperature

Fig. 1(a) and (b) shows the micrograph of grains sintered at 1100 °C. The grains of 1100 °C are irregular and stick loosely, and some small particles keep own shape around the large SiC

Table 2
Effect of water to solid rate in the slurry on infiltration.

Water/solid rate in slurry	Remark
0.4	High viscous slurry for infiltration
0.8	High viscous slurry for infiltration
1.0	Easy infiltration and particle is retained
1.2	Easy infiltration and particle is retained
1.4	Low viscous slurry for infiltration

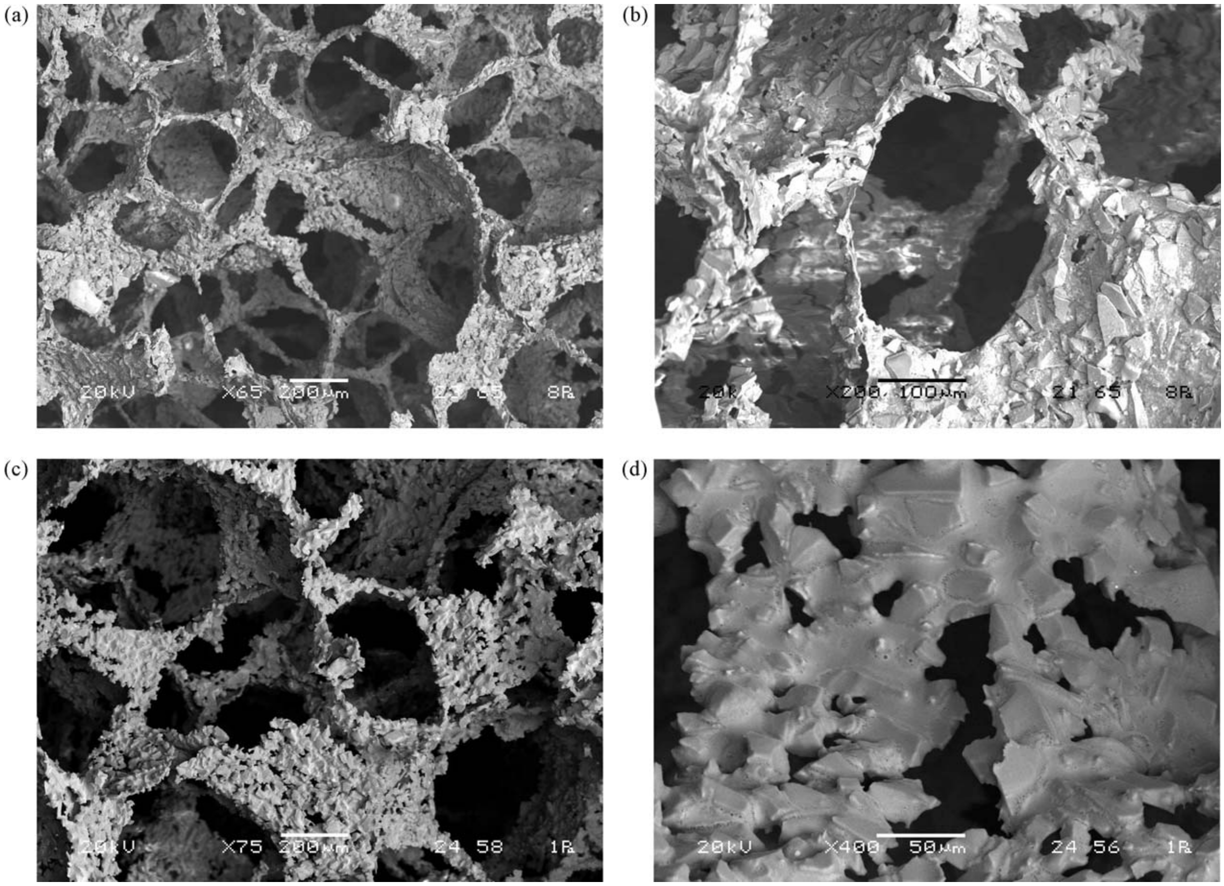

Fig. 1. SEM micrographs of the SiC foams sintered at 1100 °C and 1300 °C for 2 h. (a) SiC foam sintered at 1100 °C [65×], (b) SiC foam sintered at 1100 °C [200×], (c) SiC foam sintered at 1300 °C [75×], and (d) SiC foam sintered at 1300 °C [400×].

particles, which indicates that the sintering degree is incomplete at 1100 °C. With the increase of sintering temperatures, the small particles disappear gradually and the large grains approach and compact tightly. Strong liquid phase sintering occurs as shown in Fig. 1(b) and (c) when the foam is sintered at 1300 °C. Bentonite addition plays important role for liquid phase sintering. Ca-bentonite starts to form liquid phase between the SiC particles at 1100 °C and this liquid phase increases at 1300 °C. Densification of the SiC ceramic powders is achieved by liquid phase sintering. It is interesting that because of the liquid phase sintering no distortion or shrinkage have not been observed for the sintered ceramic foams. It is thought that sintering shrinkage is compensated by weak network connection.

There is several benefit of this type of sintering in terms of liquid metal infiltration when the produced ceramic foams are used as preforms for metallic composites. Reticulation occurring from every regions of the foam is very important. Some closed porosity may remain within the cell walls and struts. After sintering, micro porosity occurs within the cell walls due to the liquid phase sintering and volatile species as shown in Fig. 1(c) and (d). This micro pores behave like channel to connect closed porosity coming from strut voids. That is to say that the liquid can flow through macro pores first then enter to the micro pores and matrix fill all kind of pores resulting in a pore free metal matrix composite (MMC). Increase in foam strength resists pressure infiltration front and deflects cracks under mechanical loads.

3.3. Phase determination

XRD analysis of as received bentonite and SiC foam fired at 1100 °C was performed and given in Figs. 2 and 3, respectively. The XRD analysis showed that the used bentonite was sodium–calcium–bentonite. Trace amount of

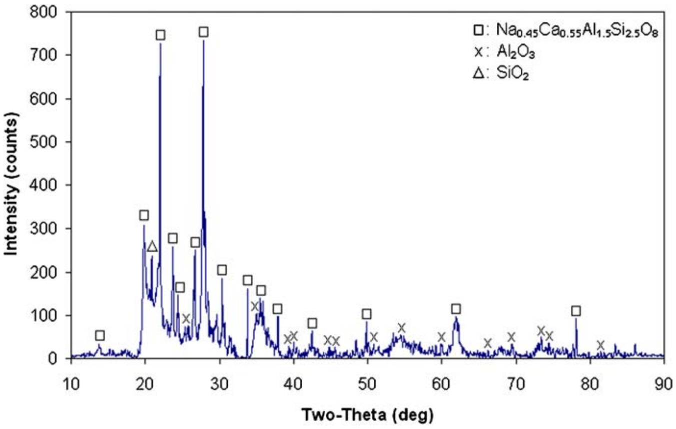

Fig. 2. XRD analysis of bentonite.

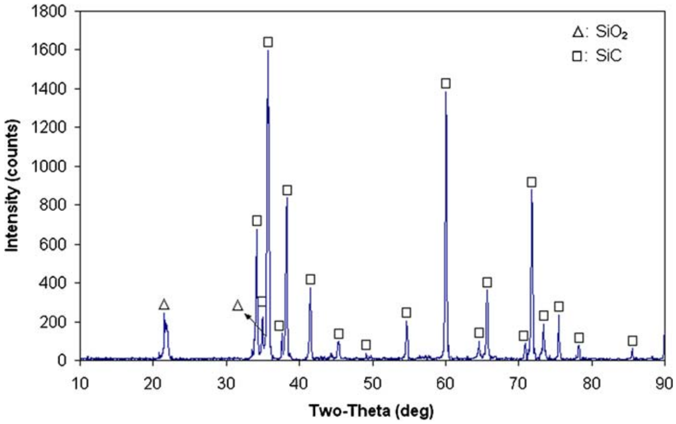

Fig. 3. XRD analysis of SiC foam.

alumina and silica were also observed within the as received bentonite. As shown in Fig. 3, SiC and SiO_2 phases were only developed from the SiC foam. Any other phase like cristobalite was not observed in the sample. The sodium and calcium either formed glassy phase or volatilized. Therefore any phase related to sodium and calcium was not found or insufficient to detect from XRD examination.

3.4. Compressive strength

The foams produced at low temperatures (700–1000 °C) have low strength and low fracture toughness as a result of the high porosity and thin skeletons, making them sensitive to structural stress and limiting their structural application. Because sintering temperatures determine porosity and sintering degree, which then affect the strength of ceramic foams. We manage to improve the foams strength by controlling sintering temperatures.

In order to observe effect of bentonite addition on sintering and mechanical properties, bentonite rate in the slurry was increased gradually. The resulting sample properties after sintering are given in Table 3. Bentonite addition does not change density and porosity much but it increases compressive strength sharply. Fig. 4 shows that 10% bentonite addition to the SiC particles gives foam maximum strength (2.44 MPa). Bentonite provides not only sintered strength but also green strength which is very important during polyurethane firing.

Table 3
Effect of bentonite amount on bulk density and porosity of SiC foams sintered 1100 °C.

Bentonite in ceramic content (%)	Density (g m^{-3})	Porosity (%)
1.00	0.515	85.9
2.00	0.528	85.5
2.50	0.530	85.4
5.00	0.541	85.1
7.50	0.558	84.5
10.00	0.563	84.3
12.50	0.559	84.5
15.00	0.554	84.7
20.00	0.545	84.9

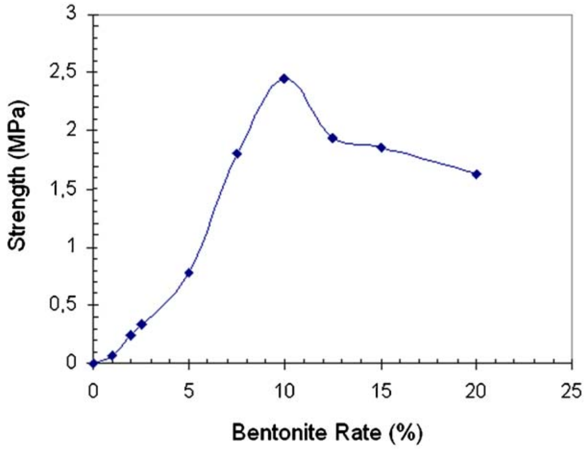

Fig. 4. Strength increases with bentonite rate.

After determining ideal bentonite in terms of foam strength, different sintering temperature has tested to observe effect of sintering temperature on densification and strength. The strength increase as result of temperature increase is shown in Table 4. At 1100 °C strength increase reaches maximum value 2.44 MPa and drops slightly above the 1100 °C. These results are also plotted in Fig. 5. Temperature increase gives strength up to 1100 °C but do not shows same affect for 1200 °C and 1300 °C. These results show consistency with Zhu et al. [15] and Yao et al. [18]. Density and porosity increase are expected due to the sintering but surprisingly, in this study, notable shrinkage has not been observed. This can be explained by densification behavior of bentonite.

As shown in Fig. 1(c) and (d), micro porosity occurs within the cell walls. Because of that shrinkage take place locally not centroid. This also explains why strength drops at high temperature sintering. As a result of liquid phase sintering densification occurs around the SiC particles and microporosity comes out between the particles. This is also beneficial during liquid metal infiltration since closed porosity at the pore struts is open by this sintering mechanism. Therefore liquid metal follows macro pores first then enter to the micro pores providing pore free metal matrix composites.

Table 4
Effect of sintering temperature on strength of SiC foam (10% bentonite).

Sintering temperature (°C)	Strength of SiC foam (Mpa)
600	0.066
700	0.238
800	0.337
900	0.776
1000	1.808
1100	2.446
1200	1.939
1300	1.933

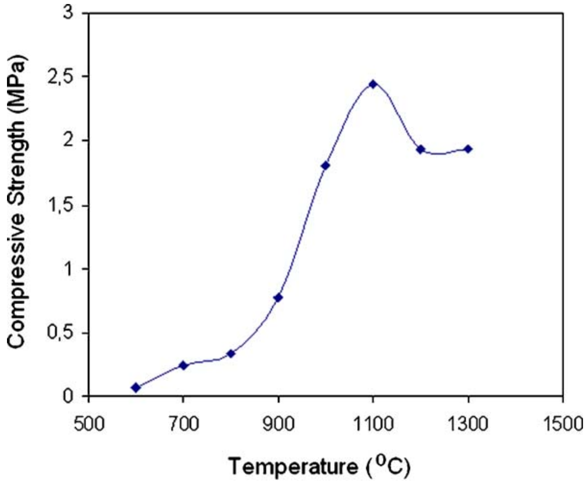

Fig. 5. Strength increases with sintering temperature for 10% bentonite addition.

4. Conclusions

Highly porous SiC foams were fabricated by a slurry infiltration process with using the polymeric sponge as the substrates. The slurry used for coating on the sponge had higher viscosity. This type of slurry rheology behavior was required by the coating on the sponge. Bentonite addition into the slurry eased to slurry infiltration and gave the strength before and after sintering with increase in viscosity, the loading content of SiC reticulated porous ceramics (RPCs) increased, leading to a decrease in the cell size and an increase in the strut thickness of SiC RPCs. The microstructure of SiC RPCs was improved by the over sintering. Instead of shrinkage micro porosity took place within the cell walls which is beneficial for dimension control and liquid metal infiltration.

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