Expansionless oxidation-bonded SiC microfiltration membrane by controlling the oxidation of SiC particle mixtures

Syed Zaighum Abbas Bukhari, Jang-Hoon Ha, Jongman Lee and In-Hyuck Song

Engineering Ceramic Department, Ceramics Materials Division, Korea Institute of Materials Science (KIMS), Changwon, Gyeongnam, Republic of Korea

ABSTRACT
Oxidation bonding is a technique used to produce porous SiC ceramics at low temperatures. The oxidation behavior of SiC particles depends on various factors, including the oxidation environment, temperature, time, particle size, and impurities. The key properties required for a porous ceramic membrane are a controlled pore morphology, high strength, and high permeability. In this study, SiC powders with different particle sizes (0.55 and 7 μm) were used to fabricate porous ceramic membranes. First, the oxidation behavior of the SiC powders was evaluated. Then, the feasibility of using their mixture to create supports for microfiltration applications was analyzed. Through this study, not only were the ideal conditions for fabricating microfiltration supports quantified but also the conditions where specimens could be made with zero size change. Finally, a membrane fabricated from a powder mixture composed of 92% of the 7 μm powder and 8% of the 0.55 μm powder and sintered at 1450°C was proposed, which had a 37% porosity, 1.42 μm pore size, 49.6 MPa flexural strength, and L cm⁻² min⁻¹ bar⁻¹ air permeability.

1. Introduction
Over the past several decades, the principles of SiC oxidation have been studied extensively [1–10], and two distinct oxidation phenomena have been reported for SiC: active and passive oxidation. The operating conditions for passive oxidation are a high oxygen partial pressure and a low temperature. In the passive oxidation of SiC, an amorphous silica oxide layer is formed, which results in an increase in the overall SiC matrix mass per the following reaction:

\[ \text{SiC} (s) + 1.5\text{O}_2(g) \rightarrow \text{SiO}_2(s) + \text{CO}(g) \]  

(1)

In contrast, active oxidation occurs when there is a limited oxygen supply and a higher temperature. In addition to SiO₂, SiO gas is formed. This leads to the evolution of gaseous SiO and thus causes a faster rate of SiC consumption per the following reaction:

\[ \text{SiC} (s) + \text{O}_2(g) \rightarrow \text{SiO}_2(g) + \text{CO}(g) \]  

(2)

The phenomenon of passive oxidation is generally dependent on the dispersion of oxygen molecules and ions through the thin oxide film [11–15]. Above 600°C in air, the reaction of SiC with air results in the formation of a dense silica surface layer. It has been observed that SiC follows a parabolic law above 1000°C, which means that the oxidation reaction depends on the rate of diffusion [13–19]. However, at temperatures below 1300°C, the transfer of molecular oxygen leads to SiC oxidation, while at temperatures above 1300°C, the process occurs through the transport of ionic oxygen [19–22].

In addition to temperature and oxygen supply, SiC oxidation depends on the quality of the starting powder (SiC crystallographic phase and presence of impurities), presence of additives, varying oxidation environment (air, O₂, or H₂O atmospheres), dwell time, and particle size [22,23].

Several studies have been conducted on the oxidation of SiC powder and completely dense single-crystalline forms. Shi et al. [24] performed the first study on SiC porous preforms bonded by a SiC oxidation reaction. She et al. [25–28] expanded the research on oxidation bonding of SiC by controlling the oxidation phenomenon by varying the sintering temperature, particle size, and pore formers as well as adding alumina as an additive. Moreover, Dey et al. [29] and Chun et al. [30] incorporated petroleum coke and microbeads in an oxidation-bonding technique as pore formers, whereas Liu et al. [31] utilized carboxymethyl cellulose and starch.

In our previous studies [32,33], we investigated the application of oxidation-bonded SiC ceramics as a membrane-coating material for the first time and observed that a controlled oxidation behavior is critical for the fabrication of a filtration layer. In that study, we used a clay-bonded SiC support. To avoid any mismatch between the support and the membrane, in
this study, we investigated an oxidation-bonded SiC ceramic support for an oxidation-bonded SiC ceramic membrane. Membrane supports must have high strength and high permeability. Because large SiC particles (which have low sinterability) lead to high permeability and low strength, while fine-sized SiC particles (which have high sinterability) lead to high strength and low permeability, we proposed the use of powder mixtures, which could help overcome the issues of low strength and low permeability. Moreover, our previous study [32,33] demonstrated that the oxidation of SiC is accompanied by volume expansion, which can be problematic in fabricating the support. Therefore, in this study, a combination of SiC particles of different sizes was used to prepare a porous SiC ceramic support without volume expansion. This study is divided into two parts: (i) evaluation of the effect of two different particle sizes on the oxidation behavior and pore structure of SiC, and (ii) verification of the effect of using a mixture of two different particle sizes.

2. Materials and methods

SiC powder pellets were prepared separately using two different SiC powders (based on the average particle size). To prepare the pellets, SiC powder with large particles (d_{50}: 7 μm) was obtained from Dalian Zhengxing Abrasive Co., Ltd., China; these are referred to as the L-SiC specimens. SiC powder with small particles (d_{50}: 0.55 μm) was procured from H.C. Starck Ceramics GmbH, Germany; these are referred to as the F-SiC specimens. These powders were then uniaxially pressed using a cylindrical die (diameter: 3.6 cm), both separately and in powder mixtures with various ratios, under a uniaxial load of 20 MPa. The compositions of the powder mixtures are presented in Table 1. Average values are reported from the results obtained by testing five samples for each experimental condition. After pressing, the specimens were sintered at 800–1700°C for 2 h, by using conventional pressureless MoSi₂ furnace made by Ajeon Heating Industrial Co., Ltd., South Korea, was used for the experiments.

As reported by Shi et al. [23], if the SiC particles used to fabricate specimens have identical sizes and a spherical shape, then under a certain sintering condition, the volume fraction of the silica layer over the SiC particles after oxidation can be calculated by the following equation:

\[
\text{Volume fraction of SiO}_2 = \frac{3\Delta W/\rho_{SiO}_2}{(W_g - 2\Delta W)/\rho_{SiC} + 3\Delta W/\rho_{SiO}_2}
\]

where

\[
\Delta W = \frac{W_s - W_g}{W_g}
\]

Here, \(\Delta W\) denotes the relative change in mass, and \(W_s\) and \(W_g\) are the weights of the sintered and green bodies, respectively. \(\rho_{SiC}\) is the density of SiC (3.16 g cm\(^{-3}\)), and \(\rho_{SiO}_2\) is the density of SiO\(_2\) (2.19 g cm\(^{-3}\)).

The linear shrinkage of the pellets was measured by determining the difference in diameter before and after sintering. Open porosity was measured using Archimedes’ method with water as the liquid medium. To measure the flexural strength, the samples were tested by the three-point bending method using an Instron 4206 (Instron, USA). The microstructure was investigated using scanning electron microscopy (SEM, JSM-5800 JEOL, Japan), and the pore size was measured using by AutoPore IV 9510 (Micromeritics, USA). X-ray diffraction (XRD) measurements were performed using a D/max-2200 PC (Rigaku Co., Ltd., Japan). Finally, the air permeability was determined using capillary flow porosimetry (CFP-1200-AEL, Porous Materials Inc., USA).

3. Results and discussion

The study was conducted in two parts: the oxidation behavior of SiC powders with two different particle sizes at various sintering temperatures, and the effect of mixing fine particles with large particles to evaluate the change in pore morphology for microfiltration applications.

3.1. Oxidation behavior of different sized SiC powders

This section discusses the effect of temperature on the oxidation of SiC powders with various particle sizes. Compacted specimens made from powders with large (L-SiC) and fine (F-SiC) particle sizes separately sintered at various temperatures were first analyzed using XRD to observe any crystallographic changes (shown in Figure 1). For the F-SiC specimens, a broad peak was observed at \(2θ = ~22°\), indicating the amorphous behavior of SiO\(_2\) as the temperature reached 1200° C. A further increase in the oxidation temperature (1300°C) showed an intense sharp peak of cristobalite. Ogbuju et al. [34] also observed the crystallization of SiO\(_2\) at 1300°C owing to the formation of cristobalite.
Furthermore, at higher temperatures, the amount of cristobalite increased, while the SiC peak showed a corresponding decrease.

In contrast to the trend of the cristobalite peak for the F-SiC specimens at 1300°C, the XRD patterns of the L-SiC specimens sintered at 1300°C exhibited a very low-intensity cristobalite peak. The intensity of the cristobalite peak increased at higher temperatures, with a corresponding decrease in the intensity of the SiC peak. This difference in silica formation is believed to be related to the particle size because reactivity is directly proportional to the surface area of the reactants.

Figure 2 shows the effect of the sintering temperature and SiC powder size (F-SiC and L-SiC) on the oxidation behavior of the SiC specimens in terms of (a) changes in the mass of the specimens and the volume fraction of SiO₂ formed after oxidation of SiC and (b) the resultant changes in the linear expansion of the specimens. In Figure 2(a), because the volume fraction of SiO₂ was calculated from the mass change data using Equation 3, similar trends are observed for the same starting particle size. Thus, based on the changes in the oxidation behavior of the specimens with temperature, the trends in Figure 2(a,b) are divided into four different stages.

In stage I (800–1200°C) in Figure 2(a), the volume fraction of SiO₂ due to oxidation in the L-SiC specimens increased at oxidation temperatures >1000°C (it is nearly zero at 1000°C). In contrast, the volume fraction of silica in the F-SiC specimens oxidized at 1000°C was already 23.06% because the oxidation began at <800°C; the extent of oxidation increased rapidly with temperature. Below 1200°C, the oxidation of the F-SiC specimens (in terms of mass change) occurred rapidly, whereas for the L-SiC specimens, the increase in the rate of mass increase was not very sharp. Thus, at 1200°C, the volume fraction of silica in the F-SiC specimens was 63%, while for the L-SiC specimens it was 18.10%. This difference in the oxidation rate was mainly due to the larger surface area of the fine particles because the oxidation rate is directly proportional to the surface area of the powder.

Similar to the increase in volume fraction of silica observed in stage I in Figure 2(a), an increase in linear expansion was observed for both the L-SiC and F-SiC specimens, as shown in Figure 2(b). In addition, as observed in Figure 2(a), this increase in linear expansion was smaller for the L-SiC specimens compared to that of the F-SiC specimens. The higher expansion for the F-SiC specimens was due to the larger amount of oxidation-based silica that formed on that SiC particles, which increased the particle size and thus the specimen size.

Stage II in Figure 2(a) (1200–1500°C) shows that as the temperature increased ≥1300°C, the slope (weight change % vs temperature) for the F-SiC specimens decreased. The oxidation rate of SiC at 1300°C decreased when the oxide layer was fully crystallized (as shown in Figure 1). As explained by Viejo et al., this is because the permeation of molecular oxygen through crystallized silica is considerably more difficult.
than that through amorphous silica [35]. Thus, the permeation of molecular oxygen through the SiC/SiO$_2$ interface was hindered, which affected the supply of oxygen required to sustain the high rate of oxidation. At this stage, the mechanism of oxidation changed from the passive to active state, which produced gaseous SiO. The produced SiO gas diffused out of the specimens, leading to further oxidation with mass loss. This mass loss is believed to be accompanied by an increase in the oxide layer and a decrease in the SiC-based core. In contrast, at >1200°C, the passive mode of oxidation was still ongoing in the L-SiC specimens. Stage II in Figure 2 shows that for the L-SiC specimens at high temperatures, the oxidation was

**Figure 2.** Effect of sintering temperature on (a) relative mass change and volume fraction of silica for SiC specimens with different starting powder sizes, and (b) linear expansion of the specimens with different starting powder sizes.
comparatively lower, which was due to the smaller surface area of the SiC powder.

However, stage II in Figure 2(b) shows that when the temperature increased from 1200 °C to 1400°C, the linear expansion of both L-SiC and F-SiC pellets began to decrease. This was due to the formation of a denser cristobalite oxide layer over the surface of the specimens because excessive oxidation-derived silica leads to high sinterability and reactive sintering. It is noteworthy that for the F-SiC specimens at ≥1300°C, the expansion was negative, which indicates shrinkage in the specimens. This is because at such temperatures, there is a high amount of silica which increases the sinterability of the specimens and causes shrinkage of the specimens. In contrast, the linear expansion of the L-SiC specimens decreased as the formation of an amorphous silica phase over the SiC particles continued, causing a slight decrease in the trend of expansion.

Stage III in Figure 2(a) (1400–1650°C) shows that the F-SiC specimens exhibited a slight change in mass. As discussed in the previous stage, the active oxidation mode had already begun, and the surface of the particles in the F-SiC specimens were fully covered by the cristobalite phase; therefore, the supply of air at the SiC/SiO$_2$ interface was blocked. A similar trend was observed for the linear expansion of the F-SiC specimens in Figure 2(b); that is, the development of a very thick cristobalite layer over the surface of the SiC particles hindered further oxidation of the F-SiC specimens, yielding an almost constant volume shrinkage. In contrast, amorphous silica was converted into the cristobalite phase in the L-SiC specimens at this stage, as shown in Figure 1. The trend in oxidation shows an increase in mass change (i.e., passive oxidation), but after the formation of cristobalite, it converted into active oxidation. At this stage, owing to the higher temperatures and active oxidation phenomenon, the lattice diffusion of ionic oxygen becomes significant. Some researchers [17,36,37] hypothesized that the out-diffusion of the by-products (carbonaceous species, such as CO$_{(g)}$) is the rate-controlling step for oxidation. Owing to this out-diffusion, bubbles were observed in the oxide layer formed on the sintered polycrystalline SiC [37]. Thus, at >1400°C, alternative rate-controlling mechanisms have been suggested: the diffusion of oxygen ions and the diffusion of CO$_{(g)}$ away from the growth interface. This out-diffusion of CO$_{(g)}$ causes porosity on the oxidized SiC surface (also indicated in Figure 3(f)), which assists in continuous oxidation at the interparticle level as well. This continuous oxidation of L-SiC with increasing temperature also caused an increase in the expansion, as shown in Figure 2(b).

Stage IV in Figure 2(a) (≥1650°C) shows a sharp decrease in the slope for the L-SiC specimens. Stage IV in Figure 2(b) also shows that at 1700°C, there is a decrease in the trend for both specimens because at this temperature, cristobalite is melted, which induces higher liquid-phase sintering and decreases oxidation by preventing oxygen flow from the outside to the inside of the surface [38]. This behavior is similar to that studied for the F-SiC specimens.

Figure 3 shows the fractured microstructures of the F-SiC and L-SiC specimens. Three temperature conditions were chosen to compare the microstructures. At 1200°C, it can be seen that particle necking had begun for the F-SiC specimens (Figure 3(a)), while for the L-SiC specimens (Figure 3(b)), there was insufficient oxidation-derived silica to form a neck between the SiC particles. This was due to the larger surface area of the F-SiC particles, which allowed them to begin sintering at lower temperatures. In contrast, the L-SiC specimens appear to have begun sintering after the temperature increased to 1450°C (Figure 3(d)) because the interparticle bonding area had just begun to develop. In contrast, the F-SiC specimens were observed to be well sintered at the same temperature (Figure 3(c)). At higher temperatures (≥1600°C), the F-SiC specimens exhibited higher amounts of oxidation-derived silica and higher densification in the microstructure (Figure 3(e)), whereas the L-SiC specimens had a non-smooth, porous surface, which is indicative of SiO$_{(g)}$ and CO$_{(g)}$ out-diffusion from the SiC/SiO$_2$ interface (Figure 3(f)). Because the structure remained porous, further oxidation would be possible.

Figure 4 shows the mercury porosimetry results to analyses the pore-size distributions of the F-SiC and L-SiC specimens sintered at various temperatures. These data show unimodal pore-size distributions for all the specimens. Because the pore size depends on the initial particle size, the F-SiC specimens had a smaller average pore size relative to that of the L-SiC specimens. Pore formation in the oxidation-bonded SiC samples followed a combination of two phenomena: Ostwald ripening [39] and volume expansion of SiC particles. In the former, as the sintering temperature increased, the generation of SiC oxidation-based silica also increased. This SiC oxidation-based silica is usually present in higher amount in fine-sized SiC particles. These highly oxidized, fine SiC particles with more amorphous silica were attached to large SiC particles, thus leaving large pores in the specimens. However, in the second phenomenon, as the sintering temperature increased, the volume expansion of SiC particles occurred owing to the formation of a low-density silica layer over the SiC particles, initiating a decrease in the pore size.

As shown in Figure 4(a), an increase in the sintering temperature resulted in an increase in the pore size for the F-SiC specimens owing to the dominance of Ostwald ripening. The average pore size increased from 63 to 87 nm with increasing sintering temperature. In contrast, as shown in Figure 4(b), the L-SiC
specimens were dominated by the particle volume expansion phenomenon, that is, with increasing sintering temperature, there was a slight decrease in pore size. However, at 1700°C, the pore size increased as the bonding silica phase melted, causing liquid-phase sintering; thus, the average pore size increased, similar to the F-SiC specimens. Figure 4 shows another significant factor of the mercury intrusion volume. In mercury porosimetry, the intrusion volume of mercury is directly related to open porosity. Thus, the decrease in mercury intrusion volume with increasing sintering temperature indicates a decrease in open porosity due to high sinterability. This decrease in open porosity leads to low permeability in microfiltration applications.

In summary, as described in Table 2, the difference in the volume change of SiC powders with different particle sizes at similar temperatures can be used to produce porous SiC ceramics with controlled volume changes. More importantly, significant differences were observed at sintering temperatures in the range of 1200 to 1600°C. Thus, this study used a mixture of the two powders, L-SiC and F-SiC, to produce beneficial results, as is discussed in Section 3.2.

3.2. Oxidation behavior of the mixed SiC powders

In the previous section, the oxidation behavior of the individual SiC powders was evaluated; in this section, the oxidation behavior of a mixture of the two SiC powders is evaluated with a focus on fabricating a porous support for microfiltration applications. The main requirements for a support are high permeability, high strength, and a smooth surface. The permeability of a support depends on the pore morphology, i.e. open porosity and pore size. Because the two main properties (i.e. strength and permeability) oppose each other, an optimum condition between the two was sought to create the ideal conditions.
The effects of sintering temperature and starting material mixing ratio on the linear expansion are shown in Figure 5. Specimen with low dimensional changes during sintering is industrially desirable. Samples with a high quantity of fine particles exhibited high linear expansion below 1300°C and vice versa, whereas the condition was reversed above 1300°C, i.e. samples with a high quantity of fine particles.
Table 2. Summary of the effect of temperature on various properties of pellets fabricated from SiC powders with different sizes.

| Oxidation Mode | Stage I (800–1200 °C) | Stage II (1200–1400 °C) | Stage III (1400–1650 °C) | Stage IV (at 1700 °C) |
|----------------|------------------------|--------------------------|--------------------------|----------------------|
| Oxide Layer Composition | F-SiC | Passive oxidation | Active oxidation (transition temp. = 1200 °C) | Formation of a thick Crystalline silica layer blocks the supply of oxygen, leading to stability | Silica melts at approx. 1700 °C, this severely affects the oxygen supply from pellet surface. |
| | L-SiC | Passive oxidation | Passive oxidation | Out-diffusion at >1500 °C leads to porosity, which increases oxidation | Silica melts at approx. 1700 °C, this severely affects the oxygen supply from pellet surface. |
| Mass Change | F-SiC | + + + | - | No change | - |
| | L-SiC | + + | + + | + + + | - |
| Volume Change | F-SiC | + + + | - - | No change | - |
| | L-SiC | - | + + + | - |

Figure 5. Effect of sintering temperature on linear expansion ratio of oxidation-bonded SiC pellets prepared with various powder size ratios.

exhibited less linear expansion. This is because the F-SiC specimens underwent expansion below 1250°C and shrinkage above 1300°C, as observed in Figure 2. In addition, between 1350 and 1500°C, almost all the mixture samples exhibited negligible expansion or shrinkage. This is because, in this temperature range (1350–1500°C), the fine particle shrinkage ceases and the large particles have very little expansion, as observed in Figure 2.

Pore morphology is very important for membrane properties. Figure 6 shows the densities and porosities of the mixture samples. It was observed that, as fine particles were mixed with large particles, the fine particles filled the spaces between the large particles, leading to an increase in the bulk density of the resultant mixture and consequently, a reduction in the effective porosity. In addition, the porosity showed a continuous decrease in the temperature range of 1200 to 1350°C, whereas the density showed a continuous increase. This is because in this temperature range, the addition of fine particles to the large particle matrix induced traditional sintering shrinkage. The thin layer of fine particles surrounding the large particles will oxidize to the maximum extent, helping join the large particles. Thus, the effectiveness of that high sinterability depends on the ratio of fine particles. At temperatures >1300°C, the density had a negligible effect until 1600°C, but the decrease in porosity was observed to continue.

The pore size distributions of the powder mixtures are shown in Figure 7. At 1450°C, the addition of fine particles to large particles caused the pore size to decrease, as shown in Figure 7(a). The average pore size decreased as the percentage of fine particles in the powder mixtures increased. This is because the fine particles filled the interstices between the particles,
and because of the high sinterability of the fine particles, they became part of the surfaces of the large particles during sintering. In contrast, Figure 7(b) shows that the sintering temperature had no effect on the pore size for powder mixtures with the same composition. As previously explained in reference to Figure 4(a,b), the dominant mode of increase or decrease in pore size with increasing sintering temperature was opposite for the F-SiC and L-SiC specimens. Thus, it can be concluded that in powder mixtures, the fine particles are sintered early, covering and smoothing the surface of the large particles, which tends to increase the pore size; at the same time, the expansion of the large particles tends to decrease the pore size. As a result, the overall effect is balanced, and the pore size remains constant with increasing sintering temperature for powder mixtures with the same ratio of large to small particles.
Figure 8 shows the morphologies of the fracture surfaces of the sintered specimens with an increasing ratio of fine particles at the same sintering temperature (1450°C). With increasing ratio of fine particles in the powder mixture, the interparticle bonding area increased. This was due to the high sinterability of the fine particles, leading to densification and a decrease in the porosity of the specimens. Moreover, the high sinterability and low porosity also led to a higher flexural strength.

Figure 9 shows the fracture morphologies of the oxidation-bonded SiC pellets sintered at various temperatures using the same composition of powder mixture (i.e. 8% fine particles). An increase in temperature caused an increase in the sinterability of the fine particles, which helped to increase the particle-to-particle bonding area. This indicates that, as the sintering temperature increased, the oxidation of the fine particles increased, the morphology became smoother, and the particle size appeared to expand, leading to a decrease in porosity.
Figure 8. SEM micrographs of oxidation-bonded SiC pellets prepared with various powder size ratios: (a) L100F0, (b) L92F8, and (c) L86F14, sintered at 1450°C.
Figure 9. SEM micrographs of oxidation-bonded SiC pellets prepared with 8% fine particles and sintered at (a) 1200°C, (b) 1300°C, and (c) 1600°C.
Figure 10 displays the air permeability data, which shows that with an increase in the sintering temperature up to 1300°C, air permeability decreased. This decrease in air permeability was due to the higher linear expansion in the fine SiC particles, which led to a decrease in the porosity and average pore size. From 1350°C, an increase in air permeability was observed, which reached a maximum at 1450°C. At this stage, the oxidized layer on the fine SiC powder (whose surface was all cristobalite) was well spread over the large SiC particles to create a smooth surface, thus causing the highest permeability at 1450°C. A further increase in temperature caused a decrease in the porosity, which in turn led to a decrease in air permeability. When considering the fine particle content in the matrix, it was observed that with an increase in the proportion of fine particles, the permeability decreased because the addition of fine particles led to a decrease in porosity and pore size.

Finally, Figure 11 shows important data for determining the ideal support composition, in which the effect of the ratio of the two powders in the mixture and the sintering temperature are linked to the flexural strength of the specimens and the air permeabilities for the same sintering conditions. She et al. studied the effect of an increase in sintering temperature for oxidation-bonded SiC specimens and observed that with an increase in sintering temperature, the oxidation of SiC and crystallization of the amorphous SiO₂ to cristobalite increases, resulting in an increase in the necking area between SiC particles [25]. A similar trend can be observed in Figure 11: as the sintering temperature increased, the flexural strength of the samples also increased. Moreover, the strength of the specimens increased with increasing proportion of fine particles in the powder mixtures. It was observed that with respect to the large particles, the fine particles were oxidized earlier, thereby increasing the overall amorphous/crystalline phase at a specific temperature. Thus, a greater the proportion of fine particles yields a higher strength.

In addition, a high sintering temperature and increased proportion of fine particles resulted in a decrease in the porosity of the SiC ceramics. R. W. Rice has reported that the strength of the porous ceramics increases exponentially with a decrease in the porosity [40]. As a result, the flexural strength of the porous oxidation-bonded SiC ceramics increased with increasing sintering temperature and increasing proportion of fine particles. However, when the flexural strength data were linked with the air permeability data, it was observed that as the flexural strength increased, the air permeability decreased, and vice versa. This is because both properties are dependent on open porosity. However, at 1450°C, an enhancement in permeability and strength was noted for a mixture containing 8 wt.% fine particles, which is believed to be the mixture with a smoother microstructure, and it did not undergo a volume change.

Table 3 shows the data for the previous studies which were focused on evaluating different aspects of oxidation-bonded SiC materials. From literature we have found that flexural strength and pore size both depend on starting SiC powder, pore former and sintering temperature. It is important to note that our
Figure 11. Effect of (a) sintering temperature of L92F8 specimens and (b) ratio of fine particles sintered at 1450°C on the flexural strength and air permeability of oxidation-bonded SiC pellets.

Table 3. Summary of literature on oxidation-bonded SiC.

| SiC       | Pore former material         | Sintering Temperature | Open Porosity | Pore Size | Flexural Strength | Reference |
|-----------|------------------------------|-----------------------|---------------|-----------|-------------------|-----------|
| -         | (particle size, µm)          | (particle size, µm)   | (%)           | (µm)      | (MPa)             |           |
| 5, 0.6, 2.3 | -                            | 1100–1400            | No data       | No data   | 20–80             | [24]      |
| 0.3, 2.3, 6, 10, 27, 58 | Graphite                    | 1100–1500            | 28–40         | 0.2–1     | 60–200            | [25]      |
| 2.3       | Graphite                     | (5, 10, 20)          | 31.5–40.5     | 2.5–11.6  | 40–133            | [26]      |
| 2.3       | Carbon (5)                   | 1400–1500            | 36–46         | 3–7.5     | 18–60             | [27]      |
| 0.34      | Microbeads                   | 1300–1400            | 19–77         | <30       | 2–202             | [30]      |
| 0.55, 7   | Petroleum coke (11.4)        | 1300                 | 35–56         | 7–9       | 5–30              | [29]      |
| 40        | Carboxy-methyl-cellulose     | 1150–1400            | 34–37         | No data   | 10–19             | [31]      |
| 0.55, 7   | Microbeads                   | 1200–1600            | 26–52         | 1.27–1.47 | 4–90              | Present study |
study is unique in two ways; firstly, the study was focused on mixture effect of two different SiC powders on the basis of particle size, which was not the focus of any of these studies. Secondly, in these studies no one has given the results of expansion caused by oxidation of SiC after sintering in air at high temperature.

4. Conclusion
In this study, the differences in the oxidation behavior of two SiC powders with different particle sizes were established, and the temperature at which oxidation began as well as the change in the transition temperature change with the particle size were identified. Furthermore, the pore structure was controlled by controlling the sintering temperature and the proportion of fine particles in the powder mixture. It was observed that the pore morphology affected the flexural strength and air permeability of the resultant samples. It was also established that a mixture of SiC powders with different particle sizes could prevent the occurrence of any volume change in the final product. Finally, 1450°C was the temperature at which maximum permeability was observed. When considering the relationship between strength and permeability, L92F8 sintered at 1450°C produced membranes with high air permeability (1.1 L cm⁻² min⁻¹ bar⁻¹) and a reasonably high strength (49.6 MPa).

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