Permeability and Nanoparticle Filtration Assessment of Cordierite-Bonded Porous SiC Ceramics

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ABSTRACT: Cordierite bonded porous SiC ceramics having pore fractions (ε) between 0.33 and 0.72 and pore sizes of 6−50 μm, flexural strength of 5−54 MPa, and elastic modulus of 6−42 GPa were prepared by oxide bonding at 1350 °C in air compacts of SiC, Al₂O₃, and MgO powders with petroleum coke (PC) as the sacrificial pore former. To test the applicability of the porous ceramics in the fluid flow field, air permeation behavior was studied with fluid superficial velocity from 0.083 to 0.90 m s⁻¹ and at 26−750 °C. The Darcian, k₁, and the non-Darcian, k₂, permeability coefficients were evaluated by fitting Forchheimer’s equation to the experimental results. The temperature dependence of the permeability coefficients was explained from structural changes occurring during test conditions. The collection efficiency of filter ceramics (ε = 0.62−0.68) operating on removal of nanosized aerosol particles with sizes varying from 7 to 300 nm was determined by counting particles before and after filtration at a fluid superficial velocity of 0.1 m s⁻¹. Experimental results showed variation of collection efficiency from 96.7 to 99.9%. The size-selective fractional collection efficiency at different porosity levels was derived by using the well-known single-collector efficiency model considering some boundary conditions, and the model data were validated with experimental results. The test results were used for examination of the applicability of the filter ceramics in nanoparticle filtration processes.

I. INTRODUCTION

Porous SiC ceramics are promising filtering media for the control of particulate matter (PM) emissions from off-gas streams in many important industrial processes, such as coal/biomass combustion and gasification for power generation, thermal remediation of contaminated soils, incineration of biomedical and industrial wastes, metal smelting, and manufacturing of cement, carbon black, and glass, etc.¹−¹¹ The long-term needs of the power generation systems have driven the development of high-temperature particle filtration technology using ceramic filters, but the focus is now shifted more and more to the chemical and process industry. SiC ceramics have low bulk density, excellent mechanical and chemical stability, high thermal conductivity, and thermal shock resistance, and they can also withstand high temperatures and hostile atmospheres. Processing routes are found to retain or create porosity without sacrificing these special properties. Oxide bonding is one of the major techniques used for synthesis of porous SiC ceramics. This technique is simple and inexpensive because it does not require any sophisticated equipment and delicate instrumentation. SiC ceramics can be processed by the oxide bonding technique at low temperatures (1300−1550 °C) under an ambient atmosphere using commercial-grade raw powders. Clay, silica, and mullite are often used as oxide bonds for SiC.¹²−¹⁷ Cordierite (2MgO·2Al₂O₃·5SiO₂) is also considered another important bond as it has a low thermal expansion coefficient and good thermal and chemical stability and can be processed at a low temperature (1350 °C).¹⁸

Studies on permeation behavior of porous ceramics are important as the results can be used to determine the optimum flow conditions and characteristic features of filtering media to ensure that the cost of fan shaft power (operational cost) and filtering media (capital cost) is kept to a minimum for effective off-gas cleaning. The face velocity and the pressure drop with which ceramic filters typically operate vary in the ranges of 1−5 cm s⁻¹ and 100−8000 Pa, respectively, depending on the particle collection efficiency and the characteristic features (porosity, pore size, thickness, and structure type, e.g., granular, fibrous, and cellular, etc.) of the filtering medium.¹⁹−²⁰ For high flow rate exhaust gases that often occur in many industrial processes, optimization of filter structure causes the operational pressure drop to decrease and the face velocity to increase without any change in the overall collection efficiency, implying useful savings of power and space for filter installation and operation.

The impact on air quality of hot gas cleaning technologies is also an important issue that needs to be considered during development of ceramic filters. The collection efficiency of a filter depends on the PM emission limit and the PM concentration of a hot aerosol stream. For stringent environmental control, acceptable limits of airborne particles finer than 10 μm (PM 10) and 2.5 μm (PM 2.5) are regulated. With the rapid development of new technologies, new processes, and

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new products based on nanoparticles, the knowledge on how these materials severely affect human health and life also grows very fast and serious considerations are being given on issues of bringing the emissions of ultrafine particles—particles finer than 0.1 μm (PM 0.1)—from industrial processes under stringent regulatory control. The researchers, thus, now consider the size of airborne particles an important ultimate parameter of processing and optimization of industrial filters. Ceramic filters must have the ability to capture ultrafine dusts, meet the stringent PM emission standards, and perform in a way similar to high-efficiency particulate air (HEPA) filters or ultralow penetration air (ULPA) filters that exhibit size-selective performance in workplace and clean room applications.

In this context, the present study is performed as a part of an Indian–Brazilian research project that aims to develop high-performance SiC ceramic filters for removal of fine and ultrafine particulate matter from hot gas streams in industrial applications. The focus of this paper is on performance evaluation of cordierite bonded SiC filters. The influences of structural parameters (porosity and pore size) on mechanical and permeability characteristics were evaluated and discussed. Experimental data of nanoaerosol filtration by the porous ceramics were acquired and compared to the existing models in order to determine the best parameters for the optimized filter compositions.

II. EXPERIMENTAL PROCEDURE

(1). Sample Preparation.

Powders of SiC (α-SiC; purity, 98.20% (w/w); 1.5406 Å). Microstructural examination was done by scanning electron microscopy (SEM, Model SE-440, Leo-Cambridge, Cambridge, U.K.). The average pore size and the pore size distribution were evaluated by mercury intrusion porosimetry (Poremaster, Quantachrome Instruments Inc., Boynton Beach, Florida, USA). Room-temperature flexural strength was determined in the three-point mode (sample dimension, 45 × 4.7 × 3.5 mm; span, 40 mm; loading rate, 0.5 mm min⁻¹) in an Instron Universal Testing Machine (Model 1123, Instron Corp., Norwood, MA, USA). Young’s modulus was determined from the slope of the load-deflection curves using standard software (Instron Bluehill-2, Bucks, U.K.).

Permeability to air flow was evaluated from room temperature to around 750 °C using a laboratory-made air permeator described elsewhere. Permeability coefficients of the samples (discs of 40 mm diameter and 10 mm thickness) were determined by the Forchheimer’s equation applicable for the compressible flow of fluids through flat membranes:

\[ \frac{P_i^2 - P_o^2}{2P_i L} = \frac{\mu v_s}{k_1} + \frac{\mu^2}{k_2} \]

where \( P_i \) and \( P_o \) are, respectively, the absolute air pressures at the entrance and exit of the sample, \( v_s \) is the outlet superficial air velocity, i.e., volumetric air flow rate at the sample exit \( Q \) divided by the cross-sectional area normal to the flow \( A \), \( L \) is the sample thickness, \( \mu \) and \( \rho \) are respectively the viscosity and the density of the permeating fluid (air), and \( k_1 \) and \( k_2 \) are, respectively, the Darcian and non-Darcian permeability coefficients, in reference to Darcy’s law. The parameters \( k_1 \) and \( k_2 \) are introduced to account for the viscous and inertial effects on the flow systems. They incorporate only the structural features of the porous medium and therefore are considered constant even if the fluid or the flow conditions change.

A dimensionless parameter, known as Forchheimer’s number, is defined by the expression:

\[ F_o = \frac{\mu v_s(k_1/k_2)}{k_1} \]

which makes it possible to rewrite Forchheimer’s equation in the following form:

\[ \frac{P_i^2 - P_o^2}{2P_i L} = \frac{\mu v_s}{k_1} [1 + F_o] \]

The percentage contributions of viscous and inertial losses on total pressure drop of a ceramic filter can thus be estimated by:

\[ \text{viscous contribution/\%} = \frac{1}{1 + F_o} \]

\[ \text{inertial contribution/\%} = \frac{F_o}{1 + F_o} \]

The permeability coefficients \( k_1 \) and \( k_2 \) were obtained by polynomial fitting of Forchheimer’s equation (eq 1) to \( v_s \) and ΔP experimental data. The density and the viscosity of air at a temperature \( T_o \) and pressure \( P_o \) (atmospheric pressure at the laboratory location (95.46 kPa)) at the exit stream were determined following the procedure described in ref 27.

The filtration behavior of the porous ceramics was examined by conducting the dust collection efficiency test at room temperature using NaCl nanoparticles (standard particles used for airborne dust filtration tests) of density 2165 kg m⁻³ and size ranging from 7 to 300 nm. The details of the experimental setup and the procedure of the test were described elsewhere. The filtration collection efficiency \( E_{(PM)} \) at any particle size \( i \) was determined from the ratio of the difference of the particle concentration at the inlet \( C_{(inlet,i)} \) and the outlet \( C_{(outlet,i)} \) to the
particle concentration at the inlet. The overall particle collection efficiency \( (E_{\text{overall}}) \) was computed by

\[
E_{\text{overall}} = \frac{C_{\text{inlet}} - C_{\text{outlet}}}{C_{\text{inlet}}}
\]

where \( C_{\text{inlet}} \) and \( C_{\text{outlet}} \) are the total concentrations of particles (\( \mu g \) m\(^{-3} \)) at the inlet and outlet of the filter, respectively, are obtained by

\[
C_{\text{inlet}} = \sum_{i=7\text{nm}}^{300\text{nm}} C_{\text{inlet},i}
\]

\[
C_{\text{outlet}} = \sum_{i=7\text{nm}}^{300\text{nm}} C_{\text{outlet},i}
\]

III. RESULTS AND DISCUSSION

(1). Structural and Physical Characterization. Sintered samples exhibited neither surface cracks nor distortion of shape. Negligible changes (<1%) of major dimensions were noticed. The recorded variation of apparent porosity (\( \varepsilon \)) was from 30.1 ± 1.2 to 71.7 ± 2.6%. XRD analysis detected \( \alpha \)-SiC, cordierite, and cristobalite as the main crystalline phases present in the sintered samples. The presence of a negligible quantity of \( Al_2O_3 \) was also detected. In the starting mixtures the amounts of \( Al_2O_3 \) and MgO were such that stoichiometric formation of cordierite was possible. During sintering, SiC oxidized, oxidation derived \( SiO_2 \) reacted with \( Al_2O_3 \) and MgO forming cordierite, and the residual \( SiO_2 \) finally converted to cristobalite.\(^{18} \)

Results of XRD analysis indicated that the cordierite formation reaction was nearly complete. Microstructure examination of the sintered samples indicated formation of an open-cell pore morphology. A typical micrograph of a sintered sample is shown in Figure 1. SiC particles were seen to be bonded at the contacting regions. Energy dispersive X-ray (EDX) analysis of the bond phase showed that it contained Mg, Al, Si, and O atoms, indicating formation of an oxide bond with an average Mg:Al atom ratio \((1:2)\), the same as that of the cordierite. The Si:O atom ratio was different from that of cordierite, likely because of the presence of silica in the bond phase. SiC particles were seen to be coated with a vitreous layer. At around 1350 °C formation of cordierite is accelerated due to crystallization of this phase directly from a liquid phase.\(^{30} \) The SiC particles are likely to be coated with the liquid.

The porous SiC ceramics studied in the present work also behaved mechanically like cellular solids. The flexural strength of the porous ceramics varied within a range of 5.0 ± 0.8 to 53.7 ± 1.3 MPa, showing a trend for decreasing strength with increasing porosity (Figure 2). A similar trend was also observed for elastic modulus (varied over the range of 6.2 ± 1.8 to 42.1 ± 6.8 GPa; Figure 2). These behaviors can be explained by the minimum solid area (MSA) based model that predicts an exponential relationship between porosity and mechanical property (strength \( \sigma \) or elastic modulus \( E \)) in cellular solids.\(^{31,32} \)

\[
\sigma = \sigma_0 e^{-b\varepsilon}
\]

\[
E = E_0 e^{-b'\varepsilon}
\]

where \( \sigma \) and \( E \) are the flexural strength and elastic modulus of the material at a porosity level of \( \varepsilon \) and \( \sigma_0 \) and \( E_0 \) are the flexural strength and elastic modulus data of completely dense material; \( b \) and \( b' \) are constants that depend on the shape and stacking of the pores. The mechanical property and porosity data were well-fitted to eqs 11 and 12 (Figure 2), and the values of \( \sigma_0, b, E_0, \) and \( b' \) obtained from the fitting equations were 269 MPa, 5.1, 287 GPa, 6.6, respectively. The values of these constants obtained in the present work were in the ranges of the reported values.\(^{15,33,34} \)

The value of \( b \) can be 6 and 9 for cubic and rhombic stacking of pores.\(^{35} \) The use of the rule of mixture further indicates that the stacking was predominantly cubic for the porous SiC ceramics investigated in the present work.

The average pore size \( (d_{\text{pore}}) \) increased from 5.6 to 49.5 \( \mu m \), and the experimental \( d_{\text{pore}} \) and \( \varepsilon \) data were fitted to an exponential relation with a fairly good fitting quality (Figure 3). A porous medium bears a typical relation between \( d_{\text{pore}} \) and \( \varepsilon \) that depends on the very nature of the porous medium. For fibrous SiC ceramics synthesized by pressure-pulsed chemical vapor infiltration of carbonized cellulose powder performs, Ohzawa et al. observed that pore size varied with porosity in a non-linear fashion.\(^{36} \) Sepulveda et al. obtained a logarithmic

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**Figure 1.** SEM image of cordierite bonded porous SiC ceramics synthesized from a powder compact having \( x = 0.48, y = 0.20, \) and \( R = 2.53 \).

**Figure 2.** Flexural strength and elastic modulus of cordierite bonded porous SiC ceramics as a function of apparent porosity.
relationship between pore size and relative density ($\rho_r$) for highly porous hydroxyapatite synthesized by gel-casting of foams. Since $\rho_r = 1 - \varepsilon$, the relation indicates that pore size changes with porosity in a logarithmic or exponential manner—a trend that was also obtained in the present work.

2. Fluid Dynamic Characterization. The SiC samples of different porosities were tested for permeation initially with air flow under ambient conditions ($T_o \approx 29 \degree C, P_o \approx 95.06 \text{ kPa}; \rho = 1.1 \text{ kg m}^{-3}; \mu = 1.88 \times 10^{-5} \text{ Pa s}$). Experimental data and the respective model fitting curves are presented in Figure 4. The air velocity ranged from 0.083 to 0.67 m s$^{-1}$. The parabolic relationship between the pressure drop parameter ($P_{i2} - P_{o2}/2P_oL$) and superficial velocity ($v_s$) was confirmed through the high-quality fitting of Forchheimer’s equation for compressible fluids (correlation coefficient $R^2 > 0.999$).

The permeability coefficients $k_1$ and $k_2$ were evaluated from the curves fitted to Forchheimer’s equation in Figure 4. The data are presented in a comprehensive map proposed by Innocentini et al.$^{26,27}$ that classifies porous media according to the type and application (Figure 5). Permeability of SiC porous samples was mostly located in the range for granular filters ($k_1 \approx 10^{-13} - 10^{-15} \text{ m}^2$), corroborating literature data on ceramic filters.$^{19,27,38}$ A remarkable increase in permeability was achieved with the increase of porosity, and samples with $\varepsilon > 47.5\%$ were included in the typical range for hot aerosol filters as described in the literature.$^{19}$ The map is also useful for revealing an unequivocal relationship linking $k_1$ and $k_2$, regardless of the type of material. In cases in which only $k_1$ is available, $k_2$ can be fairly estimated by the expression:

$$k_2 = \exp\left(-\frac{1.71588}{k_1^{0.008095}}\right)$$

Permeability coefficients $k_1$ and $k_2$ are also usually related to porosity and to a characteristic length ($d_{eq}$) of the filter structure through Ergun-like equations, given by$^{27,39}$

![Figure 3. Dependence between average pore size and apparent porosity of SiC samples.](image1)

![Figure 4. Permeation curves obtained with airflow at room temperature for samples of different porosities.](image2)

![Figure 5. Classification of permeability data of cordierite based SiC filters tested in this work. (Adapted from Innocentini et al.$^{26,27}$)](image3)


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Ergun equations work for dispersed systems of fibers, where the solid matrix is continuous and the constitutive particles/grains/fibers are not individually recognizable, the average pore or cell size \((d_{\text{pore}})\) is considered as the characteristic length \(d_{\text{eq}}\) on the other hand, if the filter is made of packed columns of loose multisided particles/grains/fibers, the characteristic length is customarily selected by the Sauter mean diameter \(d_{\text{sp}}\) of the particle/grain/fiber size distribution. The constitutive particles of the filter are spherical, then a simple relationship between \(d_{\text{as}}\) and \(d_{\text{pore}}\) can be derived:

\[
d_{\text{as}} = 1.5 \left(1 - \varepsilon \right) d_{\text{pore}}
\]

For the knowledge of \(k_1\) obtained from Ergun equation (eqs 12 or 15), one viable approach proposed in this work for estimation of \(k_2\) from porosity and pore/particle size is based on the relationship between \(k_1\) and \(k_2\) derived from a large experimental data set, as shown in the permeability map of Figure 5, and is represented by eq 11. Substitution of Ergun equations for \(k_1\) into eqs 12 and 15 yields

\[
k_2 = \exp \left( - \frac{2.57394(1 - \varepsilon)0.16186}{\varepsilon^{0.24279} d_{\text{pore}}^{0.16186}} \right)
\]  

(16)

Equations 12 and 15 defining the Darcian coefficient \(k_1\) have been tested and validated for a variety of unconsolidated porous media and also for several systems of partially sintered porous ceramics on the condition that the size distribution of ceramic raw materials is representative in the consolidated product. However, if the experimental data and studies in the literature are considered, one definite conclusion that can be drawn is that while Ergun equations work fine for \(k_1\), they are not reliable for \(k_2\) over a wide range of pore size or particle size, indicating that the Ergun approach does not fit the relationship between \(k_1\) and \(k_2\) well to the permeability map shown in Figure 5. This point is well-documented in the literature, as many empirical equations have been suggested to predict \(k_2\) based on the knowledge of \(k_1\) obtained from Ergun equation (eqs 12 or 15). One viable approach proposed in this work for estimation of \(k_2\) from porosity and pore/particle size is based on the relationship between \(k_1\) and \(k_2\) derived from a large experimental data set, as shown in the permeability map of Figure 5, and is represented by eq 11. Substitution of Ergun equations for \(k_1\) into eqs 12 and 15 yields

\[
k_2 = \exp \left( - \frac{2.41044}{\varepsilon^{0.08093} d_{\text{pore}}^{0.16186}} \right)
\]  

(17)

To check the applicability of this approach, the experimental \(\varepsilon\) and \(d_{\text{pore}}\) data of SiC filters were used in eq 18 to estimate \(k_2\), and the results were plotted in Figure 6, together with the values of \(k_2\) predicted by Ergun eq 16 using the same \(\varepsilon\) and \(d_{\text{pore}}\) data. The \(y = x\) line indicates the hypothetical agreement between predicted and experimental values obtained from the airflow tests. Plots in Figure 6 confirm that (i) as expected, Ergun eq 12 predicted very well the Darcian permeability \(k_1\) for the sintered SiC filters of average pore size in the micrometric scale (from 5.6 to 45.5 \(\mu\)m) and a wide ranging porosity from 30.1 to 71.7%; (ii) Ergun eq 13 overestimated the non-Darcian permeability \(k_2\), and the deviation increased progressively with a decrease in \(k_2\), also confirming the trends reported in the literature. (iii) the proposed equation derived on the basis of the permeability map (eq 18) displayed a satisfactory and consistent predictability of \(k_2\) in the entire range of experimental data.

Calculations based on experimental permeability coefficients \(k_1\) and \(k_2\) of samples of different porosities confirm that even for a relatively high face velocity of \(v_f = 0.1\) m s\(^{-1}\), the contributions of viscous effects and inertial effects to the total pressure drop shown by filter samples for air flow at 101.3 kPa and 30 °C are
97 and 3%, respectively. This proves that regardless of the porosity level, the airflow through porous SiC samples under typical ambient aerosol filtration conditions follows reasonably Darcy’s law.

The influence of gas temperature on the permeation behavior of cordierite based SiC filters was investigated using samples of porosity at six different levels. Figure 7 illustrates typical permeation curves for a sample of $\varepsilon = 41.6\%$ (airflow test was done with face velocity ranging from 0.083 to $\sim$0.90 m s$^{-1}$ and at three temperatures). The results showed that the resistance to fluid flow increased with temperature, as reported by other authors. The other samples tested under the same conditions also showed similar trends. The results also confirmed that the experimental data fitted to Forchheimer’s equation (eq 1) exceptionally well.

The variations of permeability coefficients $k_1$ and $k_2$ with temperature for different samples are shown in Figure 8, which indicates a slight increase of $k_1$ and a decrease of $k_2$ with an increase of temperature from 30 to 750 °C. Similar qualitative trends were observed by Innocentini et al. for refractory castables. By Barg et al. for freeze casting foams, and by Biaetto et al. for porous lanthanum carbide discs.

The permeation behavior of SiC filter samples (Figure 7) was thus affected not only by the changes of properties of air (increase of 131% in $\mu$ and decrease of 70% in $\rho$, for an increase of temperature from 30 to 750 °C) but also by the variation of permeability coefficients (average increase of 71% in $k_1$, and average decrease of 94% in $k_2$). All of these factors account for the change of pressure drop by the temperature. The situation can be better analyzed by the data generated on the basis of simulation of airflow through the SiC samples at $v_s = 0.05$ m s$^{-1}$ and temperature at two levels (30 and 750 °C; Table 1). Equal sample thickness ($L = 0.01$ m) was considered. There are three aspects of the data presented in Table 1: (i) with temperature remaining the same, an increase in $\varepsilon$ caused a clear reduction in filter pressure drop ($\Delta P$); (ii) with porosity remaining constant, an increase in temperature caused the pressure drop to increase; (iii) the contribution of inertial effect on pressure drop was very small for all samples at 30 °C (<1.2%), and it was further reduced at 750 °C (<0.2%). Thus it is considered that clean or virgin filtering elements are used at 300–1500 °C with $v_s = 0.05$ m s$^{-1}$ and they show pressure drop values in the range of 1000–5000 Pa, several conclusions can be drawn on the basis of the data shown in Table 1: (i) SiC samples of porosity $>55\%$ are the good candidates to be used as hot aerosol filters, as they exhibited pressure drop values comparable to those of commercial products; (ii) the increase of viscosity with temperature was the main factor responsible for the rising pressure drop levels of SiC filters; and (iii) at increasing airflow temperatures, tendencies of the pressure drop curves are to obey Darcy’s law.

### Dust Filtration Behavior

The mechanisms by which particles are arrested by filter media are well-documented in the published literature. The expressions were derived in the literature on the basis of semiempirical correlations, considering the process of filtration to be time-independent—a condition valid for the first stage of a filtration process when no dust cake is present in the filter. In the present work, the fractional collection efficiency of a filter in the first filtration stages was calculated using the single-collector efficiency model derived for granular filters:

$$E_{\text{frac}} = 1 - \exp\left[-\frac{aKL(1 - \varepsilon)\eta_T}{d_c}\right]$$  

(19)

where $L$, $\varepsilon$, and $d_c$ represent respectively the filter thickness, filter porosity, and the average collector diameter (represented here as the average Sauter diameter of the raw SiC powder used to prepare the filter sample, $d_{sa}$); $a$ is the fitting constant (in this work $a$ is considered to be unity); and $K$ is given by Yoshida and Tien as

$$K = \left(\frac{6}{(1 - \varepsilon)^{2/3}}\right)^{1/3}$$  

(20)

According to the model, the total collection efficiency of a single collector ($\eta_T$) is the combination of the individual contributions due to different collection mechanisms, diffusion ($\eta_d$), inertia ($\eta_i$), direct interception ($\eta_{SI}$), gravity ($\eta_g$), and electrophoresis ($\eta_{el}$). The single collector combined efficiency ($\eta_T$) is given by

![Figure 7. Typical permeation curves obtained for airflow in three temperature levels. Data presented are for a sample with porosity of 41.6%](image)

![Figure 8. Influence of airflow temperature on permeability coefficients of cordierite bonded SiC samples of different porosity (where, for example, 1E-3 represents $1 \times 10^{-3}$).](image)
Table 1. Pressure Drop Parameters Estimated by the Forchheimer’s Equation for Airflow with $v_t = 0.05 \text{ m s}^{-1}$ through Samples of Different Porosities at Two Temperature Levels (30 and 750 °C)

<table>
<thead>
<tr>
<th>$\epsilon$ (%)</th>
<th>$\Delta P$ (Pa)$^a$</th>
<th>$\Delta P_{\text{drop}}$ (%)$^b$</th>
<th>$\Delta P_{\text{inertial}}$ (%)$^c$</th>
<th>$\Delta P_{\text{vacuum: } 30-750 \degree C}$ (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>30.2</td>
<td>55911</td>
<td>98.8</td>
<td>1.2</td>
<td>1.2</td>
</tr>
<tr>
<td>41.6</td>
<td>14750</td>
<td>98.8</td>
<td>1.2</td>
<td>1.2</td>
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<tr>
<td>47.9</td>
<td>5740</td>
<td>99.0</td>
<td>1.0</td>
<td>1.0</td>
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<td>55.0</td>
<td>2963</td>
<td>98.9</td>
<td>1.1</td>
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<td>1144</td>
<td>98.6</td>
<td>1.4</td>
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</tr>
<tr>
<td>66.7</td>
<td>343</td>
<td>99.1</td>
<td>0.9</td>
<td>0.9</td>
</tr>
<tr>
<td>72.5</td>
<td>702</td>
<td>99.9</td>
<td>0.1</td>
<td>0.1</td>
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<tr>
<td>76.8</td>
<td>584</td>
<td>99.9</td>
<td>0.1</td>
<td>0.1</td>
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<td>80.2</td>
<td>420</td>
<td>99.9</td>
<td>0.1</td>
<td>0.1</td>
</tr>
<tr>
<td>83.5</td>
<td>320</td>
<td>99.9</td>
<td>0.1</td>
<td>0.1</td>
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<tr>
<td>86.6</td>
<td>254</td>
<td>99.9</td>
<td>0.1</td>
<td>0.1</td>
</tr>
<tr>
<td>88.8</td>
<td>201</td>
<td>99.9</td>
<td>0.1</td>
<td>0.1</td>
</tr>
</tbody>
</table>

$^a$Calculated by eq 1 using $k_1$ and $k_2$ from Figure 8, $\mu$ and $\rho$ are as found in ref 27, and $L = 0.01 \text{ m}$. $^b$Calculated by eqs 2 and 4. $^c$Calculated by eqs 2 and 5.

\[ \eta^* = 1 - (1 - \eta_D)(1 - \eta_{\text{diff}})(1 - \eta_i)(1 - \eta_e) \]  

(21)

The diffusional collection arises from the random movement of suspended small particles in a gas—known as Brownian diffusion—and the single-collector diffusional efficiency is given by\(^{27}\)

\[ \eta_{\text{diff}} = 4(1 - \epsilon)^{2/3}A_S^{1/3}N_{pi}^{-2/3} \]  

(22)

where the Happel’s parameter ($A_S$) is given by

\[ A_S = \frac{2(1 - (1 - \epsilon)^{5/3})}{2 - 3(1 - \epsilon)^{5/3} + (1 - \epsilon)^{5/3} - (1 - \epsilon)^2} \]  

(23)

The Peclet number ($N_{Pe}$) and the diffusivity coefficient ($D$) are given by

\[ N_{Pe} = \frac{v_d \pi_i}{D} \]  

(24)

\[ D = \frac{k_B T F_s}{3 \pi \mu \pi_i} \]  

(25)

where $d_{pi}$ is the nanoparticle dust size, $k_B$ is the Boltzmann constant, $T$ is the absolute temperature, $\mu$ is the air viscosity, and $F_s$ is the slip correction factor which is given by Allen and Raabe as\(^{50}\)

\[ F_s = 1 + \frac{\lambda}{d_{pi}} \left[ 2.34 + 1.05 \exp \left( -0.39 \frac{d_{pi}}{\lambda} \right) \right] \]  

(26)

where $\lambda$ is the mean free path of air. The single-collector inertia efficiency is given by\(^{31}\)

\[ \eta_i = 0.2589 N_{St,eff \epsilon} N_{Re} \]  

(27)

The interception parameter ($N_{Re}$) and the effective Stokes’ number ($N_{St,eff}$) are

\[ N_{Re} = \frac{d_{pi}}{d_c} \]  

(28)

\[ N_{St,eff} = \frac{[A_S + 1.14 N_{Re}^{-1.5} N_{St}^2]}{2} \]  

(29)

The Reynolds number ($N_{Re}$) and the Stokes’ number ($N_{St}$) are given by

\[ N_{Re} = \frac{\rho v_{d} d_{c}}{\mu} \]  

(30)

\[ N_{St} = \frac{\rho v_{d} F d_{pi}^2}{9 \mu d_{pi}} \]  

(31)

The expression for estimation of the collection efficiency due to direct interception is given by\(^{52}\)

\[ \eta_{int} = 6.3 e^{-2.4 N_{Re}^2} \]  

(32)

The collection efficiency due to gravitational effects is given by\(^{52,53}\)

\[ \eta_G = 3.75 \times 10^{-10} \left( \frac{\eta_i}{\nu_t} \right)^{0.5} \]  

(33)

where the terminal settling velocity ($v_t$) of the dust particle is obtained within the Stokes free-fall regime ($N_{Re} < 1$) and $g$ is the acceleration due to gravity.

\[ v_t = \frac{\rho g d_{pi}^2}{18 \mu} \]  

(34)

The single-collector electrophoretic efficiency ($\eta_e$) was not considered in the present case as the NaCl dust particles were neutralized (by Krypton-85 Neutralizer) before they entered the filter.

Figure 9 presents the typical contribution of each mechanism to the total single-collector efficiency ($\eta^*$) in the size range of

![Figure 9 contributions of different dust collection mechanisms to the single-collector efficiency for a filter with porosity of 68.42% (where, for example, 1E-3 represents 1 $\times 10^{-3}$).](image-url)
The overall filter efficiency was estimated by eq 35. The accuracy of $E_{\text{overall}}$ depends on the proper fitting of the single-collector size in eq 19.

The dust collection efficiency test was conducted for samples with three levels of porosity ($\varepsilon = 62.2$, 67.1, and 68.4%). Table 2 shows that the model and the experimental overall efficiencies tally very well if the values of the single-collector size are selected from a range of 19.6–23.7 $\mu$m—a range of particle diameter with which the single-collector size agrees very well in the present study. Figure 11 shows a very good agreement

![Figure 10. Comparison between modeling and experimental curves of fractional collection efficiency versus particle size at different levels of porosity of SiC filters.](image1)

![Figure 11. Effect of sample porosity on the overall particle collection efficiency.](image2)

### Table 2. Comparison of the Predicted (Model) and Experimental Overall Efficiency, $E_{\text{overall}}$

<table>
<thead>
<tr>
<th>porosity (%)</th>
<th>experiment</th>
<th>model</th>
<th>fitting value of collector size, $d_{\text{i}} (\mu\text{m})$</th>
</tr>
</thead>
<tbody>
<tr>
<td>68.42</td>
<td>96.7</td>
<td>96.9</td>
<td>23.7</td>
</tr>
<tr>
<td>67.13</td>
<td>98.6</td>
<td>98.5</td>
<td>22.0</td>
</tr>
<tr>
<td>62.17</td>
<td>99.9</td>
<td>99.9</td>
<td>19.6</td>
</tr>
</tbody>
</table>

At each porosity level the theoretical efficiencies also show a decreasing trend for larger particles up to 300 nm. A comparison between the modeling (theoretical) and experimental curves shows satisfactory agreement between the proposed correlation of fractional collection efficiency with particle size and the experimental results. The small differences that still exist between the modeling (theoretical) and the experimental curves may be due to the fact that calculations of fractional collection efficiencies are done on the basis of granular filter medium whereas the experiments were done using filters made of sintered porous ceramics. Figure 9 also reveals that the diffusional mechanism plays the most dominant role on the capture of nanoparticles by the filtering media. Other authors also indicated that Brownian diffusion is the most dominant dust collection mechanism for the capture of submicrometer size particles by fibrous filters. The overall efficiency was found to decrease with porosity (Figure 10). The overall efficiency was also estimated using the single-collector model:

$$E_{\text{overall}} = \sum_{i=7}^{300} w_{i} E_{\text{frac}}$$

(35)

where $E_{\text{frac}}$ is the fractional efficiency for the $i$th particle (eq 19) and $w_{i}$ is the mass fraction:

$$w_{i} = \frac{C_{\text{inlet},i}}{C_{\text{inlet}}}$$

(36)

IV. CONCLUSION

(1) Cordierite bonded porous SiC ceramics for removal of airborne nanoparticles was successfully prepared by oxide bonding of powder compacts of SiC, Al$_2$O$_3$, and MgO at 1350 °C in air. Samples of varied porosity in a range of 30.1–71.7%
could be synthesized by the variation of petroleum coke pore former volume fraction from 0.23 to 0.76. Stoichiometric formation of cordierite was possible by maintaining the $\text{Al}_2\text{O}_3$ to $\text{MgO}$ molar ratio of 2:1; the desired amount of silica was obtained by the oxidation of $\text{SiC}$. SEM/EDX test supported formation of cordierite in the bond phase bridging the gap between the contacting $\text{SiC}$ particles. Microstructure examination also indicated formation of interconnected pore structure. That the porous ceramics behave like true cellular solids is also confirmed by mechanical property data.

(2) A parabolic relationship between pressure drop/superficial velocity was confirmed through high-quality fitting of experimental data to the Forchheimer equation. Permeability coefficient values could increase several orders of magnitude depending on the addition of the sacrificial pore former used to create porosity in the ceramic material, indicating that it is possible to tailor this parameter according to the need of a specific application.

(3) The permeability coefficients of the porous $\text{SiC}$ ceramic samples with porosity greater than $~48\%$ indicated that the values are well within the typical range of high aerosol filters.

(4) The Darcian permeability coefficient, $k_d$, but not the non-Darcian permeability coefficient, $k_f$, fitted well to the Ergun-like equations for granular media. Based on the relationship between the permeability coefficients obtained by fitting a large number of reported $k_d$ and $k_f$ data in a permeability map, equations can be proposed for more reliable prediction for $k_d$ compared to the prediction made by the Ergun-like equations:

$$k_d = \exp\left(-\frac{2.57394(1 - \epsilon)^{0.16186}}{\epsilon^{0.24279} d_p^{0.16186}}\right), \text{ based on particle size}$$

$$k_f = \exp\left(-\frac{2.41044}{\epsilon^{0.08093} d_p^{0.16186}}\right), \text{ based on pore size}$$

(5) Permeation curves obtained at high temperatures (up to $~750 \, ^\circ\text{C}$) and high superficial velocities (up to $0.90 \text{ m s}^{-1}$) indicated that resistance to fluid flow increased with temperature mainly because of the increase of the fluid viscosity with temperature. Preliminary analysis of the permeability data for air flow at superficial velocities up to $0.05 \text{ m s}^{-1}$ and temperatures up to $~750 \, ^\circ\text{C}$ revealed that pressure drop is mostly governed by viscous effects and Darcy’s law is valid for the test conditions, suggesting the possibility of application of porous $\text{SiC}$ ceramics as filtering media for typical aerosol filtration processes.

(6) The filtration of nanoparticles (of sizes from $7$ to $300 \, \text{nm}$) in the SiC filter ceramics had showed that fractional particle collection efficiency decreased with increasing particle size at fixed fluid superficial velocity ($0.10 \text{ m s}^{-1}$). The size-selective fractional collection efficiency of the filter ceramics could be described very well by the single-collector efficiency model for granular filters based on classical collection mechanisms. The comparison of experimental results with the calculated collector efficiency showed the predominance of diffusional mechanism in the range studied.

(7) The overall dust collection efficiency measured with the samples of porosity varying from $62$ to $68\%$ showed a decreasing trend with porosity, likely because of the deleterious effect of porosity on the diffusional mechanism. The overall efficiency could also be modeled on the basis of the single-collector size from a range of $19.6-23.4 \, \mu\text{m}$—a range of particle diameter used in the present study—and a very good agreement between the modeled and experimental overall collection efficiency–porosity relations was obtained. The overall efficiency of the investigated $\text{SiC}$ samples thus indicates their strong application possibilities in nanoparticle filtration processes.

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**Notes**

The authors declare no competing financial interest.

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**NOMENCLATURE**

\begin{itemize}
  \item $a$ fitting constant in eq 19
  \item $A$ filter face area exposed to fluid flow (m$^2$)
  \item $A_s$ Happler’s parameter
  \item $D$ diffusion coefficient (m$^2$ s$^{-1}$)
  \item $d_c$ average collector diameter (m)
  \item $d_{eq}$ mean spherical particle diameter (m)
  \item $d_{ps}$ dust particle size (m)
  \item $d_{ave}$ average pore size (m)
  \item $d_{sa}$ Sauter mean diameter of the particle/grain/fiber (m)
  \item $E$ Young’s modulus (GPa)
  \item $E_{frac}$ fractional collection efficiency
  \item $E_{overall}$ overall collection efficiency
  \item $F_n$ Forchheimer’s number
  \item $f_S$ slip correction factor
  \item $G$ acceleration due to gravity (m s$^{-2}$)
  \item $K$ dimensionless parameter defined in eq 20
  \item $k_d$ Darcian permeability coefficient (m$^2$)
  \item $k_f$ non-Darcian permeability coefficient (m$^2$)
  \item $k_B$ Boltzmann constant (m$^2$ kg s$^{-2}$ K$^{-1}$)
  \item $L$ sample thickness (m)
  \item $MM_{ad}$ average molar mass of dry air (kg mol$^{-1}$)
  \item $N_{Pe}$ Peclet number
  \item $N_E$ interception parameter
  \item $N_{Re}$ Reynolds’ number
  \item $N_{St}$ Stokes’ number
  \item $N_{St,e}$ effective Stokes’ number
  \item $P$ absolute pressure (kg m$^{-1}$ s$^{-2}$)
  \item $P_i$ absolute inlet pressure (kg m$^{-1}$ s$^{-2}$)
  \item $P_o$ absolute outlet pressure (kg m$^{-1}$ s$^{-2}$)
  \item $Q$ volumetric flow rate of the fluid (m$^3$ s$^{-1}$)
  \item $R^2$ correlation coefficient
  \item $T$ absolute temperature (K)
  \item $v_f$ fluid velocity (m s$^{-1}$)
  \item $v_t$ terminal setting velocity (m s$^{-1}$)
\end{itemize}

**Greek Letters**

\begin{itemize}
  \item $\alpha$ thermal expansion coefficient
\end{itemize}
apparent porosity (vol %)  
η\textsubscript{D} single collector efficiency due to diffusion  
η\textsubscript{DI} single collector efficiency due to direct interception  
η\textsubscript{E} single collector efficiency due to electrophoresis  
η\textsubscript{T} total single collector efficiency  
λ mean free path of gas molecules (m)  
μ fluid viscosity (kg m\textsuperscript{-1} s\textsuperscript{-1})  
\rho fluid density (kg m\textsuperscript{-3})  
\rho\textsubscript{p} dust particle density (kg m\textsuperscript{-3})  
σ flexural strength (MPa)

**REFERENCES**


