

Experimental and numerical analysis of the complex permittivity of open-cell ceramic foams

Hernandez, J. N. C.; Link, G.; Soldatov, S.; Füssel, A.; Schubert, M.; Hampel, U.;

Originally published:

July 2020

Ceramics International 46(2020)17, 26829-26840

DOI: https://doi.org/10.1016/j.ceramint.2020.07.159

Perma-Link to Publication Repository of HZDR:

https://www.hzdr.de/publications/Publ-31444

Release of the secondary publication on the basis of the German Copyright Law § 38 Section 4.

- 1 Experimental and numerical analysis of the complex permittivity of open-cell
- 2 ceramic foams
- 3 Jesus Nain Camacho Hernandez^{a,1}, Guido Link^b, Sodatov Sergey^b, Alexander Füssel^c,
- 4 Markus Schubert^a, Uwe Hampel^{a,d}
- ^a Helmholtz-Zentrum Dresden-Rossendorf, Institute of Fluid Dynamics, Bautzner Landstr.
- 6 400, 01328 Dresden, Germany
- 7 b Karlsruhe Institute of Technology, Institute for Pulsed Power and Microwave Technology
- 8 IHM, Eggenstein Leopoldshafen, 76344, Germany
- 9 ^c Fraunhofer Institute for Ceramic Technologies and Systems IKTS, Winterbergstraße 28,
- 10 01277 Dresden, Germany
- 11 d Chair of Imaging Techniques in Energy and Process Engineering, Technische Universität
- 12 Dresden, 01062 Dresden, Germany
- ¹Corresponding author. Tel.: +49 351 260 3316 E-mail address: j.hernandez@hzdr.de (J.
- 14 Hernandez)

Abstract

- 17 Open-cell ceramic foams are promising materials in the field of microwave heating. They 18 can be manufactured from susceptor materials and can, therefore, be used as selective 19 heating elements. In this study, the complex permittivities of ceramic foam materials, 20 including silicon-infiltrated silicon carbide (SiSiC), pressureless sintered silicon carbide 21 (SSiC), silicate bonded silicon carbide (SBSiC), and cordierite were determined. The 22 dielectric properties of the foams were determined by the cavity perturbation technique 23 using a TE₁₀₄ WR340 waveguide resonator at 2.45 GHz. Samples were preheated in a 24 tubular furnace, enabling temperature-dependent permittivity measurements up to 200 °C. 25 The effective dielectric constant and effective loss factor were found to depend on the 26 porosity and material composition of the foam. The SiSiC material had a higher effective 27 dielectric constant than the SSiC and SBSiC ceramics. The effective dielectric constant of 28 the foams showed a trend of gradual increase with increasing temperature. Some selected 29 dielectric mixing relations were then applied to describe the effective permittivity of the 30 foams and compare them with predictions from finite element simulations performed using 31 the CST Studio Suite. The foam morphologies and simple block inclusions were used in the 32 simulations.
- 33 **Keywords**: A. microwave processing, C. dielectric properties, D. silicon carbide, D.
- 34 cordierite.

1 Introduction

- 36 Open-cell solid foams are interconnected solid networks (also referred to as skeletons),
- through which fluids can pass (see Figure 1A). They exhibit characteristics that make them
- 38 extremely attractive for various thermal applications, such as heat exchangers, porous
- burners, electric heaters [1], and catalyst supports [2,3].
- 40 **Figure 1**. (A) Image of the SiSiC foam morphology obtained by SEM; micro X-ray
- 41 tomography (μCT) cross-sectional images of the SiSiC foams (B) 30 ppi and (C) 45 ppi.
- 42 Microwave heating has been increasingly applied to many industrial processes and can also
- be considered as a promising approach for the selective heating of foams. The microwave
- 44 absorbance and the heating characteristics of a material are commonly described by its
- 45 complex permittivity, which is constituted of real and imaginary parts ($\varepsilon = \varepsilon' j\varepsilon''$), i.e.,
- 46 the dielectric constant (ε') and dielectric loss factor (ε'') , respectively. The effective
- 47 permittivity (ε_{eff}) is the dielectric permittivity of a system or object that is made of two or
- 48 more materials. In the case of open-cell foams, which can be manufactured from any
- sinterable material (e.g., ceramics), $\varepsilon_{\rm eff}$ is a function of the permittivities of the fluid ($\varepsilon_{\rm air}$)
- 50 and solid ($\varepsilon_{\text{ceramic}}$) phases.
- 51 Silicon carbide (SiC) is a ceramic base material that is suitable for the manufacturing of
- 52 open-cell solid foams. Its high-temperature stability, hardness, and corrosion resistance
- make it an outstanding candidate for many engineering applications. Due to its excellent

54 microwave susceptibility (i.e., the degree of absorption of the electromagnetic energy and 55 its conversion to heat), SiC is used to improve the heating efficiency in microwave 56 applications, either by blending low-loss materials with SiC particles or by placing those 57 materials close to or in contact with the bulky susceptor portions [4]. The temperature-58 dependent complex permittivity of SiC powder (bulk density of 1.89 ± 0.06 g/cm³) was 59 measured at 2.45 GHz by Fernandez et al. [5], as shown in Table 1. However, the 60 permittivity can be significantly influenced by doping or impurities. For example, boron-61 doping increases both the dielectric constant and loss factor, from $\varepsilon = 6.70 - j0.02$ to $\varepsilon =$ 62 65.44 - j32.40 (at 2.45 GHz) [6].

Table 1. Complex permittivity of SiC powder and cordierite.

63

64 The porosity (P) of solid foams is defined as the ratio of the volume of voids to the total 65 volume of the foam. As P increases, the effective permittivity decreases. For example, an 66 SiC foam with $P = 83.7 \pm 1.0$ % and a linear pore density of 50 pores per inch (ppi), has an $\varepsilon_{\rm eff} = 4.7 - j0.33$ at 2.45 GHz (at room temperature of approximately 20 °C) [6], which is 67 lower than the $\varepsilon_{\rm eff}$ for P = 41.1 ± 0.18 % (SiC powder) [5], as reported in Table 1. 68 69 The objective of this study was the analysis of the relationship between the morphology and 70 effective permittivities (at 2.45 GHz) of SiC-based and cordierite-based open-cell solid 71 foams. A low-loss magnesium-aluminum-cyclosilicate (Mg2Al3(Si5AlO18)) ceramic 72 material known as cordierite was compared with the SiC-based foams; the permittivity of 73 cordierite bulk as indicated in the literature is provided in Table 1. Thus, a systematic study 74 of the dielectric parameters was performed for ceramic-based solid foams with different

porosities, pore densities, and compositions. Furthermore, the effective permittivities that
 were predicted by some mixing relations were compared against those derived from
 performed electromagnetic simulations.

2 Experimental methods and materials

2.1 Experimental dielectric measurements

Different techniques can be used to measure the dielectric properties of solid materials [9].

Among them, the cavity perturbation technique compares the resonance frequency (f_1) and
quality factor (Q_1) of a cavity without perturbation against the resonance frequency (f_2) and
quality factor (Q_2) of the same cavity after perturbation by a small material sample. The
complex permittivity of the perturbation sample is then calculated according to

$$\varepsilon' = \frac{V_c}{V_s} \frac{(f_1 - f_2)}{A * f_2} + 1$$
, and (1)

$$\varepsilon^{\prime\prime} = \frac{V_c}{B * V_s} \left(\frac{1}{Q_1} - \frac{1}{Q_2} \right),\tag{2}$$

where V_c is the cavity volume and V_s is the sample volume [10]. The coefficients A and B can be determined by calibration, either using samples of an identical size and well-known permittivities that are similar to the material of interest or by employing a numerical calibration procedure. The latter is based on full-wave 3D electromagnetic simulations of the forward problem. By variation of the sample permittivity within the anticipated interval,

these simulations allow the correlation of the calculated frequency shift $(f_1 - f_2)$ and the changes in the quality factor $({}^1/Q_1 - {}^1/Q_2)$ with a given permittivity. The complex permittivity is then calculated from the measured scattering parameters (*S*-parameters) using this correlation [10]. The transmission factor S_{21} , which is the ratio of the transmitted power (at output port 2) to the incident power (at input port 1) characterizes the resonance behavior.

Figure 2. Schematic of the dielectric measurement system based on the cavity perturbation technique. $S_{21}(f)$ is the frequency-dependent S-parameter response at port 2 to the signal at port 1.

99

100

101

102

103

104

105

106

107

108

109

A schematic of the experimental system used to acquire the complex permittivity of the samples is shown in Figure 2. A more detailed description of the dielectric measurement system developed by Soldatov et al. can be found in [11]. The resonator was based on a standard rectangular WR-340 waveguide cavity, and the length of the cavity was chosen to be resonant at a frequency of 2.45 GHz in the TE_{104} mode. Thus, the electric field standing wave pattern had four maxima along the longest side of the cavity. The microwave signals were coupled using inductive coupling ports with a 10 mm width and 43.18 mm height on both ends. The cavity was connected to a vector network analyzer (VNA Agilent E5071C) with coaxial-to-waveguide adaptors and coaxial cables to measure the S_{21} parameter. Two small opposing holes were made in the walls of the cavity at the position of one of the four electric field maxima to enable the positioning of the sample inside the cavity (within a

quartz tube sample holder). A pyrometer (CellaTemp-PA) at the opposite end measured the sample surface temperature inside the resonator. A resistive tubular furnace (Carbolite MFT) was placed in front of the waveguide, allowing the sample to be heated to $1200 \,^{\circ}$ C before entering the cavity. In this study, the temperature range for the characterization was $30 \,^{\circ}$ C- $200 \,^{\circ}$ C. The parameters S_{21} and the temperature were recorded by a computer, which provided automatic control of the entire data acquisition process as well as predefined heating scenarios. The acquired data (i.e., resonance frequency shifts and Q-factors) were then post-processed to obtain the real and imaginary parts of the effective material permittivity (at different temperatures), via Equations (1) and (2).

2.2 Ceramic foams

- Very high temperatures or pressures (or both) are required to densify pure SiC powder by sintering due to its covalent bond characteristics [12]. Therefore, the use of additives to make the sintering process easier is preferred. In this study, the following types of SiC ceramics were investigated [13]:
- Silica-bonded silicon carbide (SBSiC) is an oxide-bonded SiC ceramic manufactured
 from coarse and medium-grained SiC powders by sintering at standard pressure, during
 which amorphous silica or cristobalite (or both) bond the SiC particles.
- Pressureless sintered silicon carbide (SSiC) is produced using very fine SiC powder that contains sintering additives.

- Silicon-infiltrated silicon carbide (SiSiC) is produced by the reaction of molten silicon with compact SiC powder, containing a high amount of free carbon. Initially, the carbon reduces the native silica layer on the SiC particles. The infiltrating melted silicon then reacts with the remaining free carbon to produce secondary SiC, leading to SiC bonding between the SiC grains. The remaining melted silicon fills the inter-particle voids creating a somewhat dense microstructure.
- The compositions and structures of SBSiC, SSiC, and SiSiC are shown in Table 2 and Figure 3, respectively.
- Figure 3. Schematic structure of (A) SBSiC, (B) SSiC, and (C) SiSiC.
- **Table 2.** Material compositions.

Solid foams have a macro-porous open-cell structure composed of stiff vertices interconnected with struts, which form a continuous grid. Images example of an SiSiC solid foam and its pore structure, obtained by scattering electron microscopy (SEM) and X-ray micro-computed tomography (μ CT), is presented in Figure 1. The struts have small cavities inside them, which are isolated from the exterior. As the pore density increases, the pore size decreases. The foam structure results from the replication technique, wherein the polymeric template defines the cell size and shape. The open-cell polymer foams were impregnated with a ceramic slurry. The excess slurry was then removed by squeezing and centrifugation, resulting in the uniform coating of the polymer foam struts. During the

subsequent heat treatment, the polymeric template was decomposed (by pyrolysis), and the ceramic material became sintered. The same SiC powders were used to manufacture the SSiC and SiSiC test samples, which were used to identify the influences of the various compositional materials and binding types (see Table 2). These powders were fine and reactive, with a median grain diameter $d_{50} \approx 2 \mu m$. The SBSiC also included a second, coarser SiC powder ($d_{50} \approx 20 \,\mu\text{m}$) to achieve a fine to coarse ratio of 2:3. Cylindrical samples of the bulk material and the open-cell foams having nominal pore densities of 30 ppi, 45 ppi, and 60 ppi and porosities in the range of 86.9 % to 96.5 % were produced (IKTS Fraunhofer, Germany) for dielectric characterization based on the cavity perturbation technique. The average cell diameters, densities, porosities, and dimensions of bulk and open-cell ceramic foam samples are summarized in Table 3. The average cell diameters of the foams were determined by optical 2D-image analysis of light micrographs using a Pore!Scan-System from Goldlücke GmbH [40]. The density of the foam strut material was lower than the maximum feasible density because no subsequent compaction, excepting the eventual sintering shrinkage, can be applied to open-cell ceramic foams. The same suspension was used for both sample types to identify the structure-related differences between the foam and bulk samples.

Table 3. Bulk and foam sample properties.

148

149

150

151

152

153

154

155

156

157

158

159

160

161

162

163

164

3 Calculations

166

167

168

169

170

171

172

173

174

175

176

177

178

179

180

181

182

183

184

3.1 Mesh construction for the numerical simulations

Two morphology classes, i.e., foams and blocks, were analyzed to compare their influences on the effective complex permittivity. The foam morphology chosen for the simulation was obtained from numerous 2D digital cross-sectional images of a rectangular 45 ppi SiSiC foam sample (size: $40 \text{ mm} \times 40 \text{ mm} \times 25 \text{ mm}$, porosity: 85 %) recorded by μ CT. The backprojection algorithm [14] was used for the 3D sample reconstruction. The spatial resolution of the reconstructed tomographs was 56 µm per voxel. The foam stack was first cut to a smaller representative cubic volume with a 15 mm edge length to reduce the computational burden of the simulations. A representative cube is a rectangular cuboidal representation that utilizes a particular volume of the foam that has the same porosity as the sample. Additional meshes with lower porosities were produced using a 3D dilatation filter to study the porosity over a wide range (see Section 4.3) [15]. This filter dilates the cross-sectional structure of the foam using a spherical kernel diameter of 3 voxels, which increases the skeleton volume, and, consequently, decreases the porosity. The mesh reconstruction (see Figure 4) was then achieved by applying the marching cubes algorithm [16], following which the small non-connected structures were eliminated. Further, mesh simplification (i.e., the transformation of a given polynomial mesh into another with fewer faces, edges, and vertices), mesh smoothing (i.e., the removal of noise from the mesh surface), and

uniform mesh resampling (to optimize the point distribution) were performed [39]. A detailed description of the mesh construction can be found in [17].

Figure 4. Foam morphology reconstruction and meshing.

Finally, the meshes were imported into the CST Microwave Studio Suite and incorporated as inclusions in a box. The outer box represented the air and the inclusions for the ceramic foams. The resulting boxes with the inclusions had porosities of 11 %, 30 %, 50 %, 70 %, and 85 %, based on the ceramic and air domains.

For the block morphology inclusions, a series of blocks were used instead. The blocks were generated in the directions of the box corners, resulting in an arrangement of eight blocks. The block size was selected to obtain porosities ranging from 0 % to 100 % at 10 % increments. Additionally, the porosities were adjusted to facilitate direct comparison with the porosities of the foam morphologies mentioned previously. Two approaches were used for the block model generation, wherein the properties of the blocks and matrix were interchanged. The first approach involved block-shaped ceramic inclusions in an air matrix, while the second used block-shaped air inclusions in a ceramic matrix. These arrangements facilitated the comparison of the effects of the non-connected and connected ceramic structures to those of real foam structures. Examples of the foam and block inclusions are shown in Figure 5.

Figure 5. Examples of the foam morphology inclusions (upper row), ceramic block inclusions (middle row), and air block inclusions (lower row), within the boxes. Ceramic and air materials are color-coded in red and blue, respectively.

3.2 Numerical modeling and complex permittivity calculation

Electromagnetic wave propagation was simulated at 2.45 GHz using the CST Microwave Studio Suite and the model shown in Figure 6 to estimate the effective permittivities of the different foams. Simulations of the hexagonal meshes representing the foam morphologies (minimum 1 000 000 cells) and block morphologies (minimum 300 000 cells) were performed using a transient domain solver. The boundary conditions were defined as follows: the pairs of normal faces in the x, y, and z directions were taken as the electric walls, magnetic walls, and free space boundaries with no reflections (i.e., open walls), respectively. The input and output microwave ports were defined at reference planes shifted to the normal faces in the z-direction of the models. The bulk permittivity from the effective-medium approximation of the SiSiC was used for the ceramic inclusions (foams and blocks). Air properties were set for the waveguide domain. Finally, the scattering parameters were acquired by numerical simulation. The simulation setup and sequential procedure are visually summarized in Figure 6.

Figure 6. Sequential procedure for the foam morphology (upper row) and block morphology (lower row) inclusions used for computing the scattering parameters using numerical simulations.

- In the next step, the effective permittivities were determined using the S-parameters [18].
- 224 The S-parameter retrieval method [19,20] was used to calculate the permittivity from the
- reflected (S_{11}) and transmitted scattering signals (S_{21}). Initially, the impedance (z) was
- obtained as

$$z = \pm \sqrt{\frac{(1 + S_{11})^2 - S_{21}^2}{(1 - S_{11})^2 - S_{21}^2}}.$$
(3)

- 227 The refractive index (n) was then calculated by applying the obtained impedance and
- 228 Equation (4):

$$n = \frac{1}{k_0 L} \left[lm \left\{ ln \left(\frac{S_{21}}{1 - \frac{(z-1)S_{11}}{(z+1)}} \right) \right\} + 2m\pi - i \cdot Re \left\{ ln \left(\frac{S_{21}}{1 - \frac{(z-1)S_{11}}{(z+1)}} \right) \right\} \right], \quad (4)$$

- where k_0 is the wavenumber, L is the sample length in the wave propagation direction
- between ports 1 and 2, m is the fundamental branch of the sinusoidal function periodicity
- (in this work, m = 0 because $L < \lambda_{foam}/4$), and λ_{foam} is the wavelength in the foam at
- 232 2.45 GHz. The effective permittivity was then calculated as

$$\varepsilon_{eff} = \frac{n}{z}.\tag{5}$$

4 Results and discussion

234

235

236

237

238

239

240

241

242

243

244

245

246

247

248

249

250

251

252

253

4.1 Determination of the complex permittivity

Figure 7 provides the effective dielectric constants (ε'_{eff}) and the effective loss factors (ε''_{eff}) of the cordierite and SiC-based samples obtained from the temperature dependence measurements using the cavity perturbation method. Notably, the bulk did not necessarily refer to samples having a 100 % theoretical density (as listed in Table 3) because of the previously described preparation technique. In contrast to the SiSiC, the other bulk materials had significant residual porosities (SBSiC: 29.1 %, SSiC: 8.7 %, cordierite: 29.3 %). The permittivity of the bulk SiSiC sample is not shown because it was detected uncertainty regarding its composition and porosity. Further, the high losses of the bulk SiSiC, SSiC, and SBSiC samples resulting from excessive perturbation caused the solution of the inverse problem associated with the cavity perturbation technique to provide unreliable results. Thus, the results for these bulk materials are also not shown. For all materials, the samples had a range of complex permittivity values (represented by the error bars in Figure 7), which was primarily caused by the spread in the sample geometry (see Table 3). No clear trend could be observed for the relationship of the permittivity to the skeleton structure or pore size (in terms of the pore density). Notably, the pore density was provided as the mean ppi-value and is a measure of the interface density [21], which corresponded to the inverse of the mean chord length. The unit of ppi is not recommended for use in

scientific papers because the inch is not a unit recognized in the international system of units. However, it is used in various industrial standards for foam production [22] (as was the case in this study). Further, it can be observed that, in most of the samples, the permittivity decreased with increasing porosity because the contribution of the air to the effective permittivity increased ($\varepsilon_{\rm air} < \varepsilon_{\rm ceramic}$).

Figure 7. Effective dielectric constants (left) and effective loss factors (right) of cordierite (first row), SiSiC (second row), SBSiC (third row), and SSiC (fourth row) at a frequency of 2.45 GHz.

As seen in Figure 7, the permittivity of the SiSiC differed significantly from that reported for the SSiC and SBSiC, highlighting the impact of the free silicon infiltration. The dielectric properties of SiC ceramic foams depend strongly on the characteristics of their doping, additives, or impurities.

From Figure 7, it is evident that the SBSiC and SSiC had similar effective dielectric constants. In comparison, the SiSiC exhibited a significantly higher effective dielectric constant, presumably related to the free silicon that infiltrated the SiSiC foam structure (Figure 3). Given that silicon is a semiconductor with a dielectric constant of approximately 11.9 [23] at 2.45 GHz, the resulting effective dielectric constant was higher as compared to the SiC foams that did not contain free silicon.

The effect of the temperature on the loss factor was considerable for the SSiC but not the other foams, presumably related to the comparably higher fraction of silicon carbide and the excess carbon that was not completely consumed by the native silica layer of the SiC particles during the sintering process (see Table 2). When adding carbon to SiC (C-SiC) [24], its conductivity changes from *n*-type to *p*-type, so a greater amount of carbon would increase the conductivity and, therefore, its loss factor. Furthermore, increasing temperatures result in increased carbon-based conductivity and permittivity [25]. Moreover, after the manufacturing process is complete, some additive materials used in the manufacturing process may remain as reacted materials or binders. In this study, boron was employed as the sintering aid. Boron is a trivalent element that creates p-type electron vacancies and, thus, further increases the electrical conductivity in the SSiC. Highly porous materials always contain some water in their pores due to the ambient atmosphere. Being exposed to elevated temperature causes water to leave the material, thus leading to a decrease in its effective permittivity. This effect, however, coincides with the change of permittivity in the skeleton material of the foam. The effect of water depletion on the effective permittivity competes in foams made from typical dielectrics like oxides, nitrides, or carbides, which have increasing trends in permittivity versus temperature. Semiconducting additives, such as silicon and carbon (in this case conducting), in SiSiC and SSiC foams, respectively, could cause increased effective loss factors, which correlates with the SiC skeleton temperature behavior and inversely with water depletion. The interplay of these three factors that define the resulting temperature behavior of the permittivity of the foams in this study is clearly different between SiSiC and SSiC.

272

273

274

275

276

277

278

279

280

281

282

283

284

285

286

287

288

289

290

291

292

Compared to SSiC, SBSiC has no carbon or boron source added, and includes alumina (Al_2O_3) and silica (SiO_2) . Fused amorphous silica (SiO_2) has a dielectric constant of approximately $\varepsilon'_{SiO_2} \sim 3.8$ throughout the frequency range of 1×10^{-5} to 40 GHz [26], and is known for its very low loss factor of $\varepsilon''_{fused \, silica} < 1 \times 10^{-3}$ [27]. Thus, the competition of only two substances, i.e., SiC and water, could be assumed. Interestingly, the water depletion resulted in a declining trend of the loss factor with temperature, but an increasing trend with the dielectric constant. This is likely a consequence of the relatively higher contribution of the water loss factor than its dielectric constant.

The notable increase of the cordierite permittivity for P = 29 % is a result of its domination of the skeleton material composition. In contrast, at $P \sim 85$ %, the water depletion almost fully compensated for the trend of increasing permittivity. As seen in Figure 7, only the loss factor of SSiC increased significantly with temperature. Thus, the microwave absorbance of SiSiC and cordierite foams could be considered to be constant within the studied temperature range.

The assessment of whether the foams behaved as homogeneous effective media is also important. Foam can be considered to behave as an effective medium when the wavelength of the incident electromagnetic radiation λ is much larger than the mean cell size. Therefore, the criterion used to determine whether the system behaves as an effective medium is based on its inclusion size parameter, which is calculated as $x = 2\pi r/\lambda$, where r represents the inclusion or particle size. Mishchenko et al. proposed a threshold value of x < 0.15 [37].

The permittivity of a medium affects the wavelength according to $\lambda = \lambda_0/\sqrt{\epsilon_{eff}}$, where λ_0 is the wavelength in a vacuum. In this study, the cells are considered as spheres, and their diameters ranged between ~ 1.46 mm (30 ppi) and < 1 mm (60 ppi). As shown in Table 4, inclusion size parameters x < 0.15 were obtained for all foam types; i.e., they can be treated as effective homogeneous media.

Table 4. Maximum inclusion size parameter (*x*) of foams; 30 ppi foams were used for the calculations because they have the largest cell diameter.

4.2 Permittivity mixing relations

As in the effective-medium approximations and empirical mixing relations (see Table 5), two-phase mixing relations were used to describe the effective permittivity of the ceramic foams in terms of their porosities and bulk permittivities. The bulk permittivities of the SiSiC, SBSiC, and SSiC were not determined and, accordingly, were considered as parameters in the mixing relations that were fitted to the experimental data using iterative least-squares estimation.

When the porosity reaches 100 %, the effective permittivity should approach the permittivity of the material occupying the pores, in this case, air; $\varepsilon'_{\rm eff}(P=1)=\varepsilon'_{\rm air}\approx 1.00$, which corresponds to $\varepsilon'_{\rm air}$ at 25 °C and is almost constant within the studied temperature range. In contrast, if the porosity is 0 %, $\varepsilon'_{\rm eff}(P=0)=\varepsilon'_{\rm cer}$ represents the

dielectric constant of the ceramic material of the skeleton or the fully dense ceramic samples.

Table 5. Mixing relations for the description of effective permittivities. (¹ The exponential relation proposed by Pabst and Gregorová [29] is not identical to the simple exponential relation promoted by Rice [36]).

4.2.1 Effective-medium approximations

The most popular effective-medium approximations [28–30,35] were used for simultaneous data fitting: the Maxwell-Garnett approximation (Maxwell type relation), Bruggeman-Landauer approximation (self-consistent relation), Differential Effective Medium (DEM, differential relation), the exponential relation (from Rice [35]), and the Pabst-Gregorová exponential relation [29] (see Table 5), as shown in Figure 8. The dimensional parameter (d) for the spheres was set as d=3. The adjustable parameter (B) of the Pabst-Gregorová exponential relation was set to B=2 for the spherical pores [29], and B in the exponential relation (from Rice [35]) was estimated from experimental data fitting.

Figure 8. Comparison of the measured and predicted effective dielectric constants using various effective medium approximation relation for SiSiC (upper-row, left), SBSiC (upper-row, right), SSiC (lower-row, left) and cordierite (lower-row, right) at 2.45 GHz and 100 °C. The SSiC permittivities from power extrapolated to 100 °C powder* [5] and foam** [6] were used for comparison.

352 The estimated effective dielectric constants of the fully dense materials were: $\varepsilon'_{SiSiC} =$

353 95.05 \pm 0.31, ε'_{SBSiC} = 19.34 \pm 0.10, ε'_{SSiC} = 20.26 \pm 0.40, and ε'_{cord} = 5.35 \pm 0.21.

Among the compared relations, the best agreement was obtained using the Maxwell-

Garnett relation for the SiC-based foams. The Maxwell-Garnett approach is a non-

symmetric relation wherein the sphere inclusions are well-separated from each other.

Because $\varepsilon'_{air} < \varepsilon'_{cer}$, the Maxwell-Garnett approach corresponds to the Hashin-Shtrikman

upper bound, as shown in Figure 8.

354

355

356

357

358

360

361

362

363

364

365

367

369

Additionally, there was some agreement between the extrapolated permittivities (at 100 °C)

of the SiC powder [5] and foam [6] with boron impurities and the Maxwell-Garnett

approach relation for the SSiC, despite these materials having a slightly different

composition. Further, the dielectric constant of fully dense SSiC ($\varepsilon'_{SSiC} = 20.26 \pm 0.40$)

agrees with the extrapolated value of $\varepsilon'_{SSiC} = 20.3 \pm 0.3$ (at 100 °C) from a fully dense tile

sample [38] of SiC with boron impurities.

4.2.2 Fixed-parameter weighted mean relations

Fixed-parameter weighted mean relations [28], such as those based on the arithmetic,

harmonic, and geometric means (see Table 5) were also used for simultaneous data fitting.

The Hashin-Shtrikman (HS) bounds were used along the fixed-parameter weighted mean

relations because the foams were considered to be anisotropic. The results are shown in

370 Figure 9.

- 371 **Figure 9**. Comparison of the measured and predicted effective dielectric constants using
- the weighted mean relations with Hashin-Shtrikman bounds for SiSiC (upper-row, left),
- 373 SBSiC (upper-row, right), SSiC (lower-row, left), and cordierite (lower-row, right) at 2.45
- 374 GHz and 100 °C.
- 375 The estimated effective dielectric constants of the fully dense materials for the fixed-
- parameter weighted mean relations were: $\varepsilon'_{\text{SiSiC}} = 91.07 \pm 0.31$, $\varepsilon'_{\text{SRSiC}} = 19.73 \pm 0.10$,
- 377 $\varepsilon'_{SSiC} = 20.63 \pm 0.40$, and $\varepsilon'_{cord} = 5.69 \pm 0.21$, which agreed exceptionally well with the
- values estimated by the effective-medium approximations.
- For the SiC-based foams, the weighted mean relations were practically reduced to the fitted
- 380 upper bound because the data were primarily concentrated at the right side of the graph
- 381 (P > 0.8). Thus, the lines for the upper Wiener or HS bounds ($\Psi_{\rm arithm} \approx \Psi_{\rm harm} \approx$
- 382 $\Psi_{\text{geom}} \approx 0$) nearly overlapped. Conversely, the measured data for the cordierite was
- between the HS bounds. All weighted mean relations agreed well with the measured data.
- The weight parameters estimated for cordierite using the HS bounds were
- 385 $\Psi_{\text{geom/arithm/harm}} \approx 0.25 \text{ and } \varepsilon'_{\text{cord}} = 5.69.$

4.2.3 General sigmoidal mean relations

- The N-parameter (N) in the general sigmoidal mean relation [28] denotes the type of
- volume-weighting (which is subharmonic for N < -1, harmonic for N = -1, geometric
- for N = 0, arithmetic for N = 1, and superarithmetic for N > 1). Figure 10 shows the

comparison of the measured and predicted effective dielectric constants for different Nparameters; one parameter was fitted to the experimental data with the condition of $N \neq 0$,
and the other was fitted by setting N = 0.

Figure 10. Comparison of the measured and predicted effective dielectric constants using the general sigmoidal mean relations for different *N*-parameters and bounds for SiSiC (upper-row, left), SBSiC (upper-row, right), SSiC (lower-row, left), and cordierite (lower-row, right) at 2.45 GHz and 100 °C.

The estimated effective dielectric constants of the fully dense materials for the general sigmoidal mean relation were essentially equal to those obtained from the weighted mean relation, which is probably related to both relations using the HS bounds. Good agreement was obtained for the non-zero N-parameter ($N \neq 0$) for all materials $N \geq 60.83$, which corresponds with the volume-weighted superarithmetic mean relations coinciding with the Hashin-Shtrikman upper bound.

4.2.4 General power mean relations

Similar to the previous relation, the n-parameter in the general power mean relation [28] denotes the type of volume-weighting, which is harmonic for n = -1, geometric for n = 0, and arithmetic for n = 1. The comparison of the predicted and measured effective dielectric constants is shown in Figure 11.

Figure 11. Comparison of predicted and measured effective dielectric constants using the general power mean relations (solid line: n = 0, dashed line: $n \neq 0$); and bounds for the SiSiC (upper-row, left), SBSiC (upper-row, right), SSiC (lower-row, left), and cordierite (lower-row, right) at 2.45 GHz and 100 °C.

Sigmoidal relations did not well represent the effective dielectric constant of the SiC-based materials because the data were mainly located at the right side of the graphs, which caused unrealistically high values of the fully dense effective dielectric constant ($\varepsilon'_{\rm SiSiC} \gg 1000$) and the n-parameters to be almost zero. In contrast, n=0.327 was determined for the cordierite to ensure satisfactory agreement between the experimental and predicted results. This value was close to the n-parameter value (n = 1/3) predicted by Landau-Lifshitz and Looyenga [33–34] for the formula ($\varepsilon'_{\rm eff}$)^{1/3} = $(1-P)(\varepsilon'_{\rm cer})^{1/3} + P(\varepsilon'_{air})^{1/3}$. Interestingly, this formula was developed for the finely dispersed mixing of isotropic particles, which would seem a reasonable assumption for cordierite foams as well. The dielectric constant estimated for the fully dense cordierite was $\varepsilon'_{\rm cord} = 6.038$.

4.3 Complex permittivity calculated by the numerical simulations

The effective permittivities were also calculated from the scattering parameters obtained via electromagnetic simulations for the SiSiC foam inclusions (porosities of 11 %, 30 %, 50 %, 70 %, and 85 %) and their corresponding block morphologies at 2.45 GHz. The effective permittivities of the fully dense SiSiC that were obtained from the effective medium approximations were used. Predictions from the simulations in comparison with

the effective medium approximation relations are shown in Figure 12. The simulated effective dielectric constants of the foam morphologies demonstrated a tendency similar to the Bruggeman-Landauer relation. Further, the effective dielectric constants of the block morphologies with ceramic and air inclusions behaved similarly to the DEM-complement relation [35] and Maxwell-Garnett relations, respectively; the DEM-complement relation refers to the mixing wherein the phases and their volume fractions are interchanged, as is the case for ceramic inclusions: $\frac{\varepsilon'_{\text{eff}} - \varepsilon'_{\text{cer}}}{\varepsilon'_{\text{air}} - \varepsilon'_{\text{cer}}} \left(\frac{\varepsilon'_{\text{air}}}{\varepsilon'_{\text{eff}}}\right)^{1/d} = P.$

Figure 12. Comparison of the effective dielectric constants for the SiSiC open-cell foams and blocks at 2.45 GHz and 100 °C obtained via simulations with the experimental data and effective medium approximation relations.

The Bruggeman-Landauer relation handles each phase symmetrically, making it invariant to the simultaneous interchanges of the phase permittivities and porosities. The host medium or inclusions are not required to be defined as ceramic or air for the entire porosity range. Therefore, phase-inversion symmetry of the material is allowed, as is expected from the foam morphology models. However, the relation predicts a percolation threshold at P = 2/3 (because air is an insulator), which results in an inflection at this point. This does not hold completely in the case of the simulated foam with $P \ge 0.70$ (see Table 9), indicating that the struts of the ceramic material were still connected at even high porosity values. The permittivity of the simulated foam further explains the apparent agreement in the trends

with P = 0.50, which matches the bi-continuous [30] segment (1/3 < P < 2/3) of the relation.

449

450

451

452

453

454

455

456

457

458

459

460

461

462

463

464

465

466

The Maxwell-Garnett relation is derived from the effective-medium approximation homogenization theory for approximately spherical dielectric particles that are uniformly distributed on a background host medium [26] and provides better prediction accuracy when large numbers of inclusions are present. Further, the inclusions are not required to be the same size as long as they are much smaller than the wavelength of the operating field. Additionally, the effective permittivity depends on the degree of polydispersivity of the spherical particles [30], which leads to a percolation threshold of P = 1, i.e., the ceramic phase is no longer connected when everything is entirely composed of air. This holds for the air block inclusions because the ceramic remains interconnected throughout the porosity range and thus, agrees well with the Maxwell-Garnett relation, which approaches the upper HS bound because $\varepsilon'_{cord} > \varepsilon'_{air}$. In comparison, the cells of the simulated foams tend to not be as well-represented by spherical particles isolated in a host medium because they are interconnected through open pores, resulting in less agreement with the Maxwell-Garnett relation (see Table 9). Moreover, the measured data were better represented by the air block inclusions, possibly due to the semiconductive silicon effectively connecting the entire skeleton.

The DEM and DEM-complement relations are non-symmetric. These relations always address the spherical inclusion phase as a dilute concentration with respect to the effective

medium, which is the best approximation of the ceramic inclusions embedded in a continuous medium, i.e., the ceramic block inclusions.

For the block inclusions, non-equal contributions generally arise due to the significant difference between their dielectric properties. When the host medium was air $(\varepsilon'_{air}/\varepsilon'_{SiSiC} = 0.058)$, charge carrier accumulation near the interphase boundaries was promoted, which caused the mixing to be a non-conducting network. Conversely, when the host medium was the SiSiC $(\varepsilon'_{SiSiC}/\varepsilon'_{air} = 17.20)$, the mixing is a conducting network [34], which increases the leak current and results in increased dielectric loss. Consequently, the dielectric loss factor of the air block inclusions (SiSiC host medium) was higher than the loss factor of the ceramic block inclusions (air host medium), as shown in Table 6.

Table 6. Comparison of the effective dielectric permittivities of the SiSiC, as predicted by the Bruggeman-Landauer, Maxwell-Garnett, and DEM-complement relations, with the results of the block and foam simulations.

5 Conclusions and outlook

In this study, the dielectric properties of SiC-based solid foams, namely SBSiC, SSiC, and SiSiC, as well as cordierite were analyzed. The complex permittivity was of particular interest because it provides the imaginary part or loss factor, which is a key parameter for predicting and designing systems heated by microwaves. Both the effective dielectric constant and the effective loss factor of the SiSiC and SBSiC foams had remarkably

different values as compared to the pure SiC, which could be attributed to the additives or dopants used during the sintering of the foams. The primary conclusions from this study are:

- The dielectric properties of open-cell solid foams based on SiC were strongly correlated with the foam porosity. In contrast, no correlation was observed for the pore density.
 - Satisfactory agreement was obtained for cordierite foams by applying the mixing relations. Notably, the general power mean relation (fitted to the experimental data) returned an exponent $n \approx 1/3$, which agrees with that of the well-known Landau mixing model prediction exponential value of n = 1/3.
 - The effective permittivities predicted by the Bruggeman-Landauer relation reasonably agreed with the numerical simulations that were based on the real foam morphology because this model accounted for the phase-inversion symmetry of the foam skeleton as its porosity increased. By comparison, the Maxwell-Garnett and DEM-complement relations were suggested to predict the numerical simulation results based on simple block non-interconnected (air and ceramic) inclusions, respectively.

Future work will focus on numerical simulations and experimental predictions of the dielectric properties of other foam materials and the disclosure of SiSiC, SBSiC, and SSiC

fully dense permittivities. Furthermore, the effects of dopants and sintering additives on the dielectric properties of SiC foams will be studied systematically.

Acknowledgment

505

506

507

- This support of this by Consejo Nacional de Ciencia y Tecnología CONACYT

 (scholarship number: 91681100) in cooperation with the German Academic Exchange
- 510 Service DAAD (scholarship number: 470810) is gratefully acknowledged.

511 **Conflict of interest**

- The authors declare that they have no known competing financial interests or personal
- relationships that could have appeared to influence the work reported in this paper.

References

- 515 [1] S. Gianella, D. Gaia, A. Ortona, High temperature applications of Si-SiC cellular
- 516 ceramics, Adv. Eng. Mater. 14 (2012) 1074–1081. doi:10.1002/adem.201200012.
- 517 [2] X. Ou, S. Xu, J.M. Warnett, S.M. Holmes, A. Zaheer, A.A. Garforth, M.A. Williams,
- Y. Jiao, X. Fan, Creating hierarchies promptly: Microwave-accelerated synthesis of
- ZSM-5 zeolites on macrocellular silicon carbide (SiC) foams, Chem. Eng. J. 312
- 520 (2017) 1–9. doi:https://doi.org/10.1016/j.cej.2016.11.116.

- 521 [3] M. Petasch, U., Füssel, A., Adler, J., & Graf, Microwave Heated open celled ceramic
- foams for catalyst applications in gas phase reactions., in: 93. DKG Jahrestagung Und
- 523 Symp. Hochleistungskeramik, München, 2018: pp. 10-13.4.2018.
- 524 [4] Microwave Heating of Metal-Based Materials, in: Microwaves Met., 2007: pp. 65–
- 525 157. doi:doi:10.1002/9780470822746.ch4.
- 526 [5] J. Fayos-Fernandez, I. Pérez-Conesa, J. Monzó-Cabrera, S. Del Pino-De León, J.
- 527 Carlos Albaladejo-González, temperature-dependent complex permittivity of several
- 628 electromagnetic susceptors at 2.45 GHz, AMPERE Newsl. 1 (2018).
- 529 [6] I. Zivkovic, A. Murk, Characterization of open cell SiC foam material, Prog.
- 530 Electromagn. Res. B. 38 (2012) 225–239. doi:10.2528/PIERB11121504.
- 531 [7] I. Trak Ceramics, dielectric materials, Magn. Gr. (2019) 7.
- https://magneticsgroup.com/wp-content/uploads/2019/09/tci-md-catalog-FOR-
- WEB.pdf (accessed December 17, 2019).
- 534 [8] A. Nakayama, A. Fukuura, M. Nishimura, Millimeter-wave measurement of complex
- permittivity using dielectric rod resonator excited by NRD-guide, IEEE Trans.
- 536 Microw. Theory Tech. 51 (2003) 170–177. doi:10.1109/TMTT.2002.806932.
- 537 [9] O. V Tereshchenko, F.J.K. Buesink, F.B.J. Leferink, An overview of the techniques
- for measuring the dielectric properties of materials, 2011 XXXth URSI Gen. Assem.
- 539 Sci. Symp. (2011) 1–4.

- 540 [10] L. F. Chen, C. K. Ong, C. P. Neo, V. V. Varadan, Vijay K. Varadan, Microwave
- 541 electronics measurement and materials characterization., Chichester, Engl. John Wiley
- 542 Sons Ltd. (2004).
- 543 [11] S. Soldatov, T. Kayser, G. Link, T. Seitz, S. Layer, J. Jelonnek, Microwave cavity
- perturbation technique for high-temperature dielectric measurements, IEEE MTTS.
- 545 Int. Microw. Symp. (2013) 1–4. doi:10.1109/MWSYM.2013.6697793.
- 546 [12] K. Raju, D.-H. Yoon, Sintering additives for SiC based on the reactivity: A review,
- 547 Ceram. Int. 42 (2016) 17947–17962.
- 548 doi:https://doi.org/10.1016/j.ceramint.2016.09.022.
- 549 [13] J.-H. Eom, Y.-W. Kim, S. Raju, Processing and properties of macroporous silicon
- carbide ceramics: A review, J. Asian Ceram. Soc. 1 (2013) 220–242.
- doi:https://doi.org/10.1016/j.jascer.2013.07.003.
- 552 [14] S. Grimnes, O.G. Martinsen, Chapter 6 Geometrical analysis (Third Ed.), Academic
- Press, Oxford, (2015): pp. 141–178. doi:https://doi.org/10.1016/B978-0-12-411470-
- 554 8.00006-4.
- 555 [15] J. Schindelin, I. Arganda-Carreras, E. Frise, V. Kaynig, M. Longair, T. Pietzsch, S.
- Preibisch, C. Rueden, S. Saalfeld, B. Schmid, J.-Y. Tinevez, D.J. White, V.
- Hartenstein, K. Eliceiri, P. Tomancak, A. Cardona, Fiji: an open-source platform for
- biological-image analysis, Nat. Methods. 9 (2012) 676–682. doi:10.1038/nmeth.2019.
- 559 [16] W. Lorensen, H. E. Cline, Marching Cubes: A high resolution 3D surface construction

- algorithm, ACM SIGGRAPH Comput. Graph. 21 (1987) 163.
- 561 doi:10.1145/37401.37422.
- 562 [17] J.N.C. Hernandez, G. Lecrivain, M. Schubert, U. Hampel, Droplet retention time and
- pressure drop in SiSiC open-cell foams used as droplet separation device A
- numerical approach, Ind. Eng. Chem. Res. (2019). doi:10.1021/acs.iecr.9b04247.
- 565 [18] Kuek Chee Yaw, Measurement of Dielectric: Material Properties: Application Note,
- 566 Rohde & Schwarz, 2012.
- 567 [19] A.B. Numan, M.S. Sharawi, Extraction of material parameters for metamaterials using
- a full-wave simulator [Education Column], IEEE Antennas Propag. Mag. 55 (2013)
- 569 202–211. doi:10.1109/MAP.2013.6735515.
- 570 [20] A.J. Pickles, M.B. Steer, Effective permittivity of 3-D periodic composites with
- 571 regular and irregular inclusions, IEEE Access. 1 (2013) 523–536.
- 572 doi:10.1109/ACCESS.2013.2279356.
- 573 [21] W. Pabst, E. Gregorová, T. Uhlířová, Microstructure characterization via stereological
- 574 relations A shortcut for beginners, Mater. Charact. 105 (2015) 1–12.
- 575 doi:https://doi.org/10.1016/j.matchar.2015.04.013.
- 576 [22] K.S. Joachim Ohser, 3D Images of Materials Structures: Processing and Analysis,
- 577 Wiley, 2009.
- 578 [23] S. Varadan, G. Pan, Z. Zhao and T. Alford, As-doped Si's complex permittivity and

- its effects on heating curve at 2.45 GHz frequency, Joint Conference of the IEEE
- International Frequency Control Symposium & the European Frequency and Time
- 581 Forum (2015), Denver, CO, 2015, pp. 111-116, doi: 10.1109/FCS.2015.7138802.
- 582 [24] Y. Taki, M. Kitiwan, H. Katsui, T. Goto, Electrical and thermal properties of off-
- stoichiometric SiC prepared by spark plasma sintering, J. Asian Ceram. Soc. 6 (2018)
- 584 95–101. doi:10.1080/21870764.2018.1446490.
- 585 [25] R.A. Buerschaper, Thermal and electrical conductivity of graphite and carbon at low
- 586 temperatures, J. Appl. Phys. 15 (1944) 452–454. doi:10.1063/1.1707454.
- 587 [26] P. Sarafis, A.G. Nassiopoulou, Dielectric properties of porous silicon for use as a
- substrate for the on-chip integration of millimeter-wave devices in the frequency range
- 589 140 to 210 GHz, Nanoscale Res. Lett. 9 (2014) 418. doi:10.1186/1556-276X-9-418.
- 590 [27] R.G. Geyer, Dielectric characterization and reference materials, 1st ed., U.S. Dept. of
- Commerce, National Institute of Standards and Technology, Washington, 1990.
- 592 [28] W. Pabst, S. Hříbalová, Describing the effective conductivity of two-phase and
- multiphase materials via weighted means of bounds and general power means, JOM.
- 71 (2019) 4005–4014. doi:10.1007/s11837-019-03693-4.
- 595 [29] W. Pabst, E. Gregorová, Critical Assessment 18: elastic and thermal properties of
- 596 porous materials rigorous bounds and cross-property relations, Mater. Sci. Technol.
- 597 31 (2015) 1801–1808. doi:10.1080/02670836.2015.1114697.

- 598 [30] S. Torquato, Random heterogeneous materials, Microstructure and macroscopic
- properties, 1st ed., Springer-Verlag New York, New York, 2002. doi:10.1007/978-1-
- 600 4757-6355-3.
- 601 [31] M.A. Camerucci, G. Urretavizcaya, M.S. Castro, A.L. Cavalieri, Electrical properties
- and thermal expansion of cordierite and cordierite-mullite materials, J. Eur. Ceram.
- 603 Soc. 21 (2001) 2917–2923. doi:https://doi.org/10.1016/S0955-2219(01)00219-9.
- 604 [32] W. Westphal and A. Sils, Dielectric constant and loss data. MIT Technical Report
- 605 AFML TR 72 39, 1972.
- 606 [33] D.C. Dube, Study of Landau-Lifshitz-Looyenga's formula for dielectric correlation
- between powder and bulk. J. Phys. D: Appl. Phys. 3 (1970) 1648. doi:
- 608 https://doi.org/10.1088/0022-3727/3/11/313
- 609 [34] G. Bánhegyi, Comparison of electrical mixture rules for composites, Colloid Polym.
- 610 Sci. 264 (1986) 1030–1050. oi: https://doi.org/10.1007/BF01410321
- 611 [35] A .H. Shivola, Electromagnetic mixing formulas and applications, 1st ed., IEE
- electromagnetic waves series 47, Institution of Electrical Engineers, 1999. ISBN: 0-
- 613 85296-772-1,978-0-85296-772-0.
- 614 [36] R. W. Rice, Porosity of Ceramics, 1st ed., Materials Engineering 12, CRC Press, 1998.
- 615 ISBN: 0-8247-0151-8,0-8247-0151-8,9780824701512.
- 616 [37] M.I. Mishchenko, J.M. Dlugach, L. Liu, Applicability of the effective-medium

01/		approximation to neterogeneous aerosoi particles, J. Quant. Spectrosc. Radiat. Transf.
518		178 (2016) 284–294. doi:https://doi.org/10.1016/j.jqsrt.2015.12.028.
519	[38]	Y. Takeuchi, T. Abe, T. Kageyama and H. Sakai, "RF Dielectric Properties of SIC
520		Ceramics and their Application to Design of HOM Absorbers," Proceedings of the
521		2005 Particle Accelerator Conference, Knoxville, TN, USA, (2005), pp. 1195-1197,
522		doi: 10.1109/PAC.2005.1590705.
523	[39]	P. Cignoni, M. Callieri, M. Corsini, M. Dellepiane, F. Ganovelli, G. Ranzuglia
524		MeshLab: an Open-Source Mesh Processing Tool Sixth Eurographics Italian Chapter
525		Conference, (2008) 129-136.
526	[40]	Goldlücke GmbH. PORE!SCAN SYSTEMS Fürth, Germany.
527		https://www.giib.de/en/products-services/ (accessed June 22, 2020).
528		
529		
530		
531		
532		
533		
534		

Figure captions

635

636 Figure 1. (A) Image of the SiSiC foam morphology obtained by SEM; micro X-ray 637 tomography (µCT) cross-sectional images of the SiSiC foams (B) 30 ppi and (C) 45 ppi. 638 Figure 2. Schematic of the dielectric measurement system based on the cavity perturbation 639 technique. $S_{21}(f)$ is the frequency-dependent S-parameter response at port 2 to the signal at 640 port 1. 641 Figure 3. Schematic structure of (A) SBSiC, (B) SSiC and (C) SiSiC. 642 **Figure 4.** Foam morphology reconstruction and meshing. 643 **Figure 5.** Examples of the foam morphology inclusions (upper row), ceramic block 644 inclusions (middle row), and air block inclusions (lower row), within the boxes. Ceramic 645 and air materials are color-coded in red and blue, respectively. 646 **Figure 6.** Sequential procedure from foam morphology (upper row) and block morphology 647 (lower row) inclusions towards computational modeling for computing scattering 648 parameters using numerical simulations. 649 Figure 7. Effective dielectric constants (left) and effective loss factors (right) of cordierite 650 (first row), SiSiC (second row), SBSiC (third row) and SSiC (fourth row) at a frequency of 651 2.45 GHz. 652 **Figure 8.** Comparison of measured and predicted effective dielectric constants using 653 various effective medium approximation relations for SiSiC (upper-row, left), SBSiC 654 (upper-row, right), SSiC (lower-row, left) and cordierite (lower-row, right) at 2.45 GHz and 655 100 °C (SSiC permittivities from power extrapolated to 100°C powder* [5] and foam** [6] 656 were used for comparison). 657 **Figure 9.** Comparison of the measured and predicted effective dielectric constants using the 658 weighted mean relations with Hashin-Shtrikman bounds for SiSiC (upper-row, left), SBSiC 659 (upper-row, right), SSiC (lower-row, left), and cordierite (lower-row, right) at 2.45 GHz and 100 $^{\circ}$ C. 660 661 Figure 10. Comparison of the measured and predicted effective dielectric constants using 662 the general sigmoidal mean relations for different N-parameters and bounds for SiSiC 663 (upper-row, left), SBSiC (upper-row, right), SSiC (lower-row, left), and cordierite (lower-664 row, right) at 2.45 GHz and 100 °C. 665 Figure 11. Comparison of predicted and measured effective dielectric constants using the 666 general power mean relations (solid line: n = 0, dashed line: $n \neq 0$); and bounds for the 667 SiSiC (upper-row, left), SBSiC (upper-row, right), SSiC (lower-row, left), and cordierite 668 (lower-row, right) at 2.45 GHz and 100 °C. 669 Figure 12. Comparison of the effective dielectric constants for the SiSiC open-cell foams 670 and blocks at 2.45 GHz and 100 °C obtained via simulations with the experimental data and 671 effective medium approximation relations. 672 673 674

675 **Table captions** 676 **Table 2**. Complex permittivity of SiC powder and cordierite. 677 **Table 2.** Material compositions. 678 **Table 3.** Bulk and foam sample properties. 679 **Table 4.** Maximum inclusion size parameter (x) of foams; 30 ppi foams were used for the 680 calculations because they have the largest cell diameter. **Table 5.** Mixing relations for the description of effective permittivities. (1 The exponential 681 682 relation proposed by Pabst and Gregorová [29] is not identical to the simple exponential 683 relation promoted by Rice [36]). **Table 6.** Comparison of the effective dielectric permittivities of the SiSiC, as predicted by 684 685 the Bruggeman-Landauer, Maxwell-Garnett, and DEM-complement relations, with the 686 results of the block and foam simulations. 687

Table 3. Complex permittivity of SiC powder and cordierite.

Material	Temperature (°C)	Frequency (GHz)	Complex permittivity (ε)	Reference
	30		11.45 – <i>j</i> 2.00	
SiC powder	50	2.45	11.72 – <i>j</i> 1.65	[5]
	70		11.87 – <i>j</i> 1.36	
	~20	1x10 ⁻⁶	6 – <i>j</i> 0.06	[31]
Cordierite	~20	8.52	4.77 - j0.008	[32]
Cordicine	20	10	$4.30 - j8.6x10^{-4}$	[7]
	~20	18.3	$4.89 - j11.6x10^{-4}$	[8]

Table 2. Material compositions.

Material	Ingredients
SBSiC	Bi-modal SiC powder composition, silica sources and alumina powder as inorganic binder matrix.
SSiC	Fine SiC powder, organic temporary binder acting as carbon source after pyrolysis, and boron as a sintering aid.
SiSiC	Fine SiC powder, organic temporary binder acting as carbon source after pyrolysis to react with the infiltrated silicon.
Cordierite	Composition of cordierite chamotte, bentonite, alumina, clay, and soapstone.

Table 3. Bulk and foam sample properties.

Material Description		Averaged cell diameter (µm)	Density (g/cm³)	Porosity (%)	Diameter / height of samples (mm)
	Bulk	-	2.14	29.1	7.9 / 10.0
ana.a	30 ppi	≈ 1460	0.18 - 0.28	90.7 – 94.0	
SBSiC	45 ppi	≈ 890	0.16 - 0.22	92.6 - 94.7	$7.2 \pm 0.7 /$ 9.3 ± 0.7
	60 ppi	≈ 600	0.22	92.7	
	Bulk	-	2.88	8.7	7.9 / 10.0
aa.a	30 ppi	≈ 1460	0.19 - 0.21	93.3 - 94.1	
SSiC	45 ppi	≈ 890	0.11 - 0.15	95.2 - 96.4	6.9 ± 1.0 / 9.0 ± 1.0
	60 ppi	≈ 600	0.11 - 0.17	94.7 - 96.5	
	Bulk	-	2.69	0.0	7.9 / 10.0
a.a.c	30 ppi	≈ 1460	0.27 - 0.35	88.0 - 88.9	
SiSiC	45 ppi	≈ 890	0.20 - 0.22	90.9 - 91.8	$7.4 \pm 0.5 / 9.5 \pm 0.5$
	60 ppi	≈ 600	0.18 - 0.20	92.2 - 93.0	
	Bulk	-	1.80	29.3	7.9 / 10.0
Cordierite	30 ppi	≈ 1460	0.33 - 0.37	85.3 - 87.2	
	45 ppi	≈ 890	0.27 - 0.49	80.7 - 89.5	$7.4 \pm 0.5 / 9.5 \pm 0.5$
	60 ppi	≈ 600	0.27	89.5	

Table 4. Maximum inclusion size parameter (*x*) of foams; 30 ppi foams were used for the calculations because they have the largest cell diameter.

Foams	SBSiC	SSiC	SiSiC	Cordierite
$\mathcal{E}_{eff,30ppi}^{\prime}$	1.85	1.67	7.10	1.45
λ (mm)	90.7	99.3	46.4	101.32
χ_{30ppi}	0.05	0.05	0.10	0.05

Table 5. Mixing relations for the description of effective permittivities. (¹ The exponential relation proposed by Pabst and Gregorová [29] is not identical to the simple exponential relation promoted by Rice [36]).

Effective-medium approximations				
Maxwell-Garnett approximation (Maxwell-	$\frac{\varepsilon'_{\text{eff}} - \varepsilon'_{\text{cer}}}{\varepsilon'_{\text{re}} + (d-1)\varepsilon'_{\text{re}}} = (P) \frac{\varepsilon'_{\text{air}} - \varepsilon'_{\text{cer}}}{\varepsilon'_{\text{re}} + (d-1)\varepsilon'_{\text{re}}}$			
type relation non-symmetric) [30]	$\varepsilon'_{\rm eff} + (d-1)\varepsilon'_{\rm cer} \qquad \varepsilon'_{\rm air} + (d-1)\varepsilon'_{\rm cer}$			
Bruggeman-Landauer approximation (self-consistent relation symmetric) [30, 35]	$(1-P)\frac{\varepsilon_{\text{cer}}' - \varepsilon_{\text{eff}}'}{\varepsilon_{\text{cer}}' + (d-1)\varepsilon_{\text{eff}}'} = -(P)\frac{\varepsilon_{\text{air}}' - \varepsilon_{\text{eff}}'}{\varepsilon_{\text{air}}' + (d-1)\varepsilon_{\text{eff}}'}$			
Differential Effective Medium (DEM) approximation (Bruggeman relation non-symmetric) [30, 35]	$rac{arepsilon_{ m air}^{\prime}-arepsilon_{ m eff}^{\prime}}{arepsilon_{ m air}^{\prime}-arepsilon_{ m cer}^{\prime}}{\left(rac{arepsilon_{ m cer}^{\prime}}{arepsilon_{ m eff}^{\prime}} ight)^{1/d}=1-P$			
Exponential relation [36]	$\frac{\varepsilon_{\rm eff}'}{\varepsilon_{\rm cer}'} = \exp(-B \cdot P)$			
Pabst-Gregorová ¹ exponential relation [29]	$\frac{\varepsilon'_{\rm eff}}{\varepsilon'_{\rm cer}} = \exp\left(-B\frac{P}{1-P}\right)$			
	Empirical mixing relations			
Weighted arithmetic mean of the upper and lower bounds [28]	$\varepsilon_{\mathrm{eff}}' = (1 - \Psi_{\mathrm{arithm}})\varepsilon_{\mathrm{B}}^{+} + \Psi_{\mathrm{arithm}}\varepsilon_{\mathrm{B}}^{-}$			
Weighted harmonic mean of the upper and lower bounds [28]	$arepsilon_{ ext{eff}}' = \left[rac{(1-arPsi_{ ext{harm}})}{arepsilon_{ ext{B}}^{+}} + rac{arPsi_{ ext{harm}}}{arepsilon_{ ext{B}}^{-}} ight]^{-1}$			
Weighted geometric mean of the upper and lower bounds [28]	$\varepsilon_{\rm eff}' = \exp \left[\left(1 - \Psi_{\rm geom} \right) \ln(\varepsilon_{\rm B}^+) + \Psi_{\rm geom} \ln(\varepsilon_{\rm B}^-) \right]$			
General sigmoidal mean of the upper and lower bounds [28]	$\varepsilon_{\mathrm{eff}}' = (1 - \delta_{0,N})[(1 - P)(\varepsilon_{\mathrm{B}}^+)^N + P(\varepsilon_{\mathrm{B}}^-)^N]^{\frac{1}{N}}$			
	$+ \delta_{0,N} \{ \exp[(1-P) \ln(\varepsilon_{\mathrm{B}}^+) + P \ln(\varepsilon_{\mathrm{B}}^-)] \}$			
	where $\delta_{0,N} = \left\{ egin{matrix} 1 & ext{if} & N=0 \\ 0 & ext{otherwise} \end{matrix} ight.$			
General power mean of the upper and lower dielectric constants [28]	$\varepsilon_{\mathrm{eff}}' = (1 - \delta_{0,n})[(1 - P)(\varepsilon_{\mathrm{cer}}')^n + P(\varepsilon_{air}')^n]^{\frac{1}{n}}$			
diciocute constants [20]	$+ \delta_{0,n} \{ \exp[(1-P) \ln(\varepsilon_{\mathrm{cer}}') + P \ln(\varepsilon_{\mathrm{air}}')] \}$			
	where $\delta_{0,n} = \left\{ egin{matrix} 1 & ext{if} & n=0 \\ 0 & ext{otherwise} \end{array} \right.$			
Wiener bounds ($\varepsilon_B = \varepsilon_W$)	Hashin–Shtrikman bounds ($\varepsilon_B = \varepsilon_{SH}$)			
Upper Wiener $\varepsilon_{W}^{+} = (1 - P)\varepsilon_{cer}' + P\varepsilon_{air}'$ bound [28]	Upper Hashin– Shtrikman bound [28] $\varepsilon_{HS}^{+} = (1 - P)\varepsilon_{cer}' + P\varepsilon_{air}' - \frac{(1 - P)P(\varepsilon_{cer}' - \varepsilon_{air}')^{2}}{3\varepsilon_{cer}' - (1 - P)(\varepsilon_{cer}' - \varepsilon_{air}')}$			
Lower Wiener bound [28] $\varepsilon_{\rm W}^{-} = \left[\frac{(1-P)}{\varepsilon_{\rm cer}'} + \frac{P}{\varepsilon_{\rm air}'}\right]^{-1}$	Lower Hashin– Shtrikman bound [28] $\varepsilon_{HS}^{-} = (1 - P)\varepsilon_{cer}' + P\varepsilon_{air}' - \frac{(1 - P)P(\varepsilon_{cer}' - \varepsilon_{air}')^2}{3\varepsilon_{air}' + P(\varepsilon_{cer}' - \varepsilon_{air}')}$			

Table 6. Comparison of the effective dielectric permittivities of the SiSiC, as predicted by the Bruggeman-Landauer, Maxwell-Garnett, and DEM-complement relations, with the results of the block and foam simulations.

Porosity	Effective dielectric permittivity $\varepsilon_{\mathrm{eff}}'$ (-)					
	Simulation with foam morphology	Simulated blocks – air inclusions	Simulated blocks -ceramic inclusions	Bruggeman- Landauer	Maxwell- Garnett	DEM- complement
0.11	79.83 – <i>j</i> 2.99	78.30 <i>– j</i> 3.94	20.74 - j0.25	79.62 – <i>j</i> 4.27	80.41 - j4.32	55.59 – <i>j</i> 2.40
0.30	51.03 - j1.43	56.80 – <i>j</i> 2.96	7.99 — <i>j</i> 0.04	53.12 – <i>j</i> 2.82	58.37 – <i>j</i> 3.12	19.35 – <i>j</i> 0.43
0.50	26.42 - j0.72	37.49 – <i>j</i> 1.97	4.29 – <i>j</i> 0.01	25.85 - j1.28	38.74 - j2.05	6.65 – <i>j</i> 0.05
0.70	8.96 - j0.39	21.62 – <i>j</i> 1.09	2.49 - j0.00	5.11 - j0.09	21.98 – <i>j</i> 1.13	2.75 - j0.01
0.85	5.40 - j0.01	0.00 - j0.00	1.64 - j0.00	1.75 - j0.00	10.95 – <i>j</i> 0.54	1.71 - j0.00