Assessment of structural and transport properties in fibrous C/C composite preforms as digitized by X-ray CMT. Part II: Heat and gas transport

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Assessment of geometrical and transport properties of a fibrous C/C composite preform as digitized by x-ray computerized micro-tomography. Part II: Heat and gas transport properties

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LIST OF SYMBOLS

LATIN

Symbol	Signification	Unit
a	Sub-sample edge length	m ; μm
$\mathbf{b}$	Closure variable	m
$\underline{\underline{B}}$	Permeability	m^2 ; μm^2
d_p	Pore diameter	m ; μm
$\underline{\underline{D}}$	Diffusion coefficient	$m^2.s^{-1}$
$\mathbf{f}$,$\underline{\underline{F}}$	Closure variables	m, m^2
G	Image greyscale value	-
$\underline{\underline{Id}}$	Identity tensor	-
M	Corrective constant in eq. (12)	-
N	Distribution number	-
$\mathbf{n}$	Normal vector	
P	Pressure	Pa
Kn	Knudsen number	-
Q	Auxiliary variable, see eq. (16)	
$\mathbf{v}$	Velocity	$m.s^{-1}$

GREEK

Symbol	Signification	Unit
α	Characteristic exponent for tortuosity-porosity correlations	-
ε	Porosity (volume fraction)	-
$\underline{\underline{\eta}}$	Tortuosity factor	-
$\underline{\underline{\lambda}}$	Thermal conductivity	$W.m^{-1}.K^{-1}$
μ	Viscosity	$Pa.s$
ρ	Density	$kg.m^{-3}$
ϕ_{fi}	Fibre volume fraction	-
Ψ	Any material property	
Ω	Space domain	$[m^3]$

SUPERSCRIPTS AND UNDERSCRIPTS

Symbol	Signification
$<\bullet>$	Intermediate-scale average
$<\bullet>^{\varepsilon}$	Porosity-distribution-based average
$<\bullet>^{f}$	Intrinsic fluid average
$\{\bullet\}$	Large-scale average
$\bullet^{\lim}$	Limiting value
$\bullet^{\mathrm{ref}}$	Reference
$\bullet^{\mathrm{eff}}$	Effective value
$\bullet_{b}$	Relative to binary diffusion
$\bullet_{f}$	Relative to fluid phase
$\bullet_{fi}$	Relative to fibre
$\bullet_{fs}$	Relative to fluid-solid interphase
$\bullet_{gas}$	Relative to gas phase
$\bullet_{p}$	Relative to percolation threshold
$\bullet_{pyC}$	Relative to pyrocabon
$\bullet_{s}$	Relative to solid phase
$\bullet_{v}$	Relative to viscous flow
$\bullet_{x,\,y \text{ or } z}$	Relative to *x*, *y* or *z* direction
$\bullet_{0}$	Initial state
$\bullet_{//}$	Relative to parallel direction
$\bullet_{\perp}$	Relative to perpendicular (transverse) direction

Abstract

Raw and partially infiltrated carbon-carbon composite preforms have been scanned by high-resolution synchrotron radiation x-ray computerized micro-tomography (CMT). Three-dimensional (3D) high-quality images of the pore space have been produced at two distinct resolutions and have been used for the computation of transport properties: heat conductivity, binary gas diffusivities, Knudsen diffusivities, and viscous flow permeabilities. The computation procedures are based on a double change-of-scale strategy suited to the bimodal nature of pore space, and on the local determination of transport anisotropy. Good agreement has been found between all calculated quantities and experimental data.

Key words

Thermal conductivity, diffusion, X-ray tomography

1 Introduction

Thermostructural composites are characterized by their ability of operating under high mechanical stresses and high temperature (above 1000 °C), such as in spatial propulsion systems or aircraft brake disks. They are made of carbon or ceramic fibres (SiC, Al_2O_3…) linked together by a carbon or ceramic matrix. The association of these two brittle components leads to a material with pseudo-plastic mechanical behaviour. They are manufactured, among other processes, by chemical vapour infiltration (CVI): a heated fibrous preform is infiltrated by the chemical cracking of a vapour precursor of the matrix material inside its pore space [1, 2]. The final quality of materials fabricated by CVI relies on processing conditions

(such as vapour precursor concentration, temperature and pressure), as well as on geometrical, gas and heat transport properties of the preform. This rather expensive process has a strong need for model-based optimization, either in isothermal or in thermal-gradient process modifications. Numerous previous works have shown the crucial importance of adequate structural models in the quality of the modelling procedures [3-9]. In addition to these process-related issues, thermal characteristics of these composites are also of great interest: in some cases (braking, re-entry into the atmosphere…) they are as important as their mechanical or chemical properties.

The aim of the present work is to provide for fibrous carbon/carbon (C/C) composites estimates of such properties based on accurate 3D representations obtained by x-ray computerized micro-tomography (CMT). In a previous study, it was demonstrated that geometrical properties of the composite preform could be determined from such images [10]. In this companion paper, the same 3D images are used to assess heat and gas transport properties.

Experimental determination of heat [11-13] and gas [14-15] transport properties of thermostructural composites can be performed. However, this characterization has to be undertaken each time a new material is produced. Moreover, measuring the effective diffusivity of porous media is still a challenging task [15]. To complete these approaches, numerous works have been performed to compute thermal conductivity [16-19], diffusivity [20-22] and permeability [23-25] of ideal media such as regular or random arrays of cylinders. However, real composites exhibit a much more complex structure. In this paper, 3D CMT images are directly used to compute the transport properties of a preform at different stages of densification with the aim of performing direct comparisons either with experimental data, or with known model results.

In high-resolution images, carbon fibres can be seen in details whereas low-resolution images enable us to view components of the fibrous reinforcement (yarns and needlings). Our computational method, described in the next paragraph, makes use of low-resolution images to compute thermal conductivity, gas diffusivity and permeability of the material. High-resolution images are also used to determine the local arrangement of fibres inside yarns, and to compute diffusivity-porosity relations. Results related to each property are presented and discussed in the last section.

2 Experimental study

A thorough description of the material studied here can be found in our previous paper [10]. It is a C/C composite, provided by Snecma Propulsion Solide, Le Haillan, France. The preform weave layers are stacked horizontally ($x - y$ plane). Then, harpoon-shaped needles are used to punch these cloth stacks: as a consequence, fibres are broken and partially transferred in the z direction. The stacks are now held together by needlings. The volume fraction of fibres is about 30%, and 4% lie in z direction. The diameter of carbon fibres is about 8 μm. The yarn, less than 1 *mm* in diameter, is made up of the gathering of a large number of fibres. In this study, pores inside a yarn will be referred to as micro-pores while pores between yarns will be called macro-pores. Thus, there are two scales of heterogeneity to take into account in order to assess correctly the properties of the preform.

Three samples were extracted from the preform at different stages of densification. The first one, **CC0**, was taken from the raw preform. Its bulk density, obtained by weighing and measuring the sample, is 470 $kg.m^{-3}$, and the

corresponding porosity is 73%. Two other samples were taken after the preform was partially infiltrated. Owing to the fabrication process (isothermal isobaric CVI), the core of the preform is more porous than its borders.

The samples, extracted at different depths, have consequently distinct characteristics. **CC1** has a bulk density of 770 $kg.m^{-3}$ (corresponding to 58% porosity) and **CC2** a bulk density of 1520 $kg.m^{-3}$ (corresponding to 20% porosity). The samples were embedded in organic resin and manufactured so as to image them at two different resolutions (high and low), and view the two scales of porosity. In addition to these samples, another sample with an infiltration level intermediate between **CC1** and **CC2** (giving a 35% porosity) has been imaged only at low resolution. It will be referred to as **CC1b**.

The interested reader is referred to the preceding paper [10] for a detailed description of the experimental procedure featuring sample preparation and synchrotron x-ray CMT acquisition at European Synchrotron Radiation Facility (ESRF, Grenoble). Images used in this study are high and low-resolution images (effective voxel size equal respectively to 0.7 and 7.46 μm). Figure 1 is an example of the results obtained for sample **CC1**.

3 Computational methods

3.1 Double change of scale strategy

Due to the fabrication process (gathering of a large number of fibres in yarn and then arrangement by weaving and needling of these yarns), the preform exhibits two scales of heterogeneity. High-resolution images enable us to view small-scale details [at the scale of the yarn, see Fig. 1(b)] whereas large-scale patterns can be fully captured in low-resolution images [see Fig. 1(a)]. Geometrical assessments

[10] strongly support such an evidence, at least for partially infiltrated samples. Since low-resolution images only provide information on large-scale properties (i.e., at Representative Elementary Volume (REV) size), small-scale information has to be integrated into it. This is the first change of scale: its procedure, schematised in Fig. 2, consists in determining the local properties of the high-resolution image by dividing it into cubic sub-samples of edge length a (variable parameter). Porosity, fibre orientation and then transport properties inside each sub-sample are determined. The results are cast into expressions relating the transverse and parallel properties to the local porosity. The second change of scale consists in computing the effective properties (thermal conductivity, gas diffusivity and permeability) of the low-resolution image from a field of small-scale properties by the method of volume averaging (see Fig. 3). The procedure starts from the subdivision of the low-resolution images into sub-images; then, inside every sub-image, an evaluation of porosity and local fibre orientation is performed. Then, local property tensors may be affected to every sub-image; they may come either from the results of the first change of scale, or from analytical models.

This strategy is suggested by the following argument: in a macro-scale image, there exists a distribution of porosity $N(\varepsilon)\mathrm{d}\varepsilon$, that may be assessed by means of image analysis. If some property Ψ is known to be correlated to porosity by a relation $\Psi = \Psi(\varepsilon)$, a porosity-average of the property is given by:

$$\langle \psi \rangle^{\varepsilon} = \int_0^1 \psi(\varepsilon) N(\varepsilon) \mathrm{d}\varepsilon \tag{1}$$

Then, the proposed procedure is a mere extension of this simple averaging by taking into account the true space distribution of porosity, as sampled at macro-scale. The

hypothesis that is taken here is that the porosity-property correlation is correctly sampled at micro-scale and directly transposable for macro-scale computations.

The double change of scale procedure is suggested for the computation of effective properties of the fibrous preform at the beginning of the densification process (sample **CC0** and **CC1** with respective porosities 73% and 58%). In the latter stages of densification, a simple change of scale procedure for the computation of gas transport properties is proposed (samples **CC1b** and **CC2** with respective porosities 35% and 20%). Indeed, in such a case, the micro-pores are quite sealed off by the deposition reaction, so that they may be safely disregarded.

3.2 Determination of local porosity

Six sub-samples, 210^3 voxels (i.e., 1.6^3 mm^3) in size, have been selected, three of them in the tomographic low-resolution image of **CC0** and the three other ones in the low-resolution image of **CC1**. Each sub-sample has been subdivided into cubic sub-sub-samples of 6 voxels edge size (i.e., 0.045 mm^3), the porosity of which has to be determined. The tomographic image is a 3D reconstruction of the absorption coefficient of the material traversed by the x-ray beam. Therefore, the grey level of each voxel is a linear combination of the average grey level of each individual component of the material. Let G_1 be the grey level associated to void space, and G_2 the grey level of the carbon components (fibre or deposit). Then, the average grey level $\langle G \rangle_i$ of the i^{th} sub-sample of porosity ε_i is:

$$\langle G \rangle_i = \varepsilon_i \, G_1 + (1 - \varepsilon_i) \, G_2 \tag{2}$$

Finally, a proportionality relationship is given between the average grey level of the sub-sub-sample and its porosity:

$$\varepsilon_i = \frac{1}{G_1 - G_2} \times \langle G \rangle_i - \frac{G_2}{G_1 - G_2} \tag{3}$$

However, there is a phase contrast phenomenon [26] which makes this law not exactly true. Moreover, we have assumed that the grey level was the same for all carbon-containing components, which is not strictly the case, as their densities are indeed somewhat different [27]. Instead of using Eq. (3), we have determined, in an image of 500 voxels edge length, the threshold greyscale value G_t that leads to a binary image whose porosity matches the experimentally determined value. Then, the porosity ε_i of each sub-image of 6 voxels edge length has been determined by a thresholding operation at greyscale level G_t. A linear correlation has finally been established between ε_i and the average value of the greyscale level $\langle G \rangle_i$ of the sub-image.

Using these correlations, the porosity of each sub-sub-sample inside low-resolution images of **CC0** and **CC1** has been calculated. The mean pore volume fraction of the whole sub-samples (210 voxels edge length) are 72.7, 73.6, and 73.4% for **CC0** and 60.6, 58.7, and 55.3% for **CC1**, in agreement with measurements (73 ± 1.5% for **CC0** and 58 ± 2% for **CC1**). The scatter of pore fraction values indicates that the sub-samples are not large enough to be representative, particularly for **CC1**. Consequently, the effective property of the composite will be assessed by averaging the values determined in the three sub-samples.

3.3 Determination of local fibre orientation

The second geometrical property that is crucial in the computational procedures is the local orientation of the fibres. This issue has been addressed with an original method based on a random-walk algorithm sensitive to the local anisotropy. Details are given in the first part of this article [10]. Each cubic region of 6 voxels edge length was arbitrarily thresholded to 50% pore fraction and random

walkers were allowed to travel in the “fluid” phase until the covariance matrix of the centred displacements, divided by twice the walk time, converges to a pseudo-diffusion tensor. The eigenvector associated to the largest eigenvalue indicates the direction of preferred diffusion, which is assimilated to the local fibre orientation. The validity of the method has been checked by quantifying the proportion of fibres lying in the *x*, *y* and *z* orientation. The proportion detected by our method is in good agreement with values established otherwise [28].

3.4 First change of scale by direct computation

Once the porosity and the fibre orientation inside each sub-sub-sample of 6 voxels edge length have been computed, the next step consists in determining effective transport properties from each sub-sample by homogenisation.

3.4.1. Determination of local gas transport properties

Computations make use of a random-walk algorithm described by Vignoles [29]. It is indeed a homogenisation procedure since the effective property is computed over walks the size of which is much larger than one single image. When a walker crosses an image border, it is reintroduced in the opposite border. Another way to describe the method is the following: consider that the space is paved by repetition of the elementary image. At $t = 0$, all walkers are placed randomly in the fluid phase of an elementary image located at the space origin: thus the initial average concentration field is (up to some noise due to the finite number of walkers) a hat-function. The random walk provides a numerical Lagrangian solver for the classical unsteady diffusion problem represented by Fick’s second law, subject to the above mentioned initial condition and to boundary conditions of null concentration at infinite distances. The diffusion coefficient that arises from the convergence of the covariance matrix is also an inverse identification of the

analytical Gaussian solution in the limit of large spreading (i.e., such that the initial hat-function is considered as a Dirac impulse distribution).

There are three diffusion regimes depending on the Knudsen number *Kn*, which is the ratio between the mean free path of the molecules and the pore diameter: the ordinary (continuum) regime (*Kn* << 1), the transition regime (*Kn* ~1) and the Knudsen regime (*Kn* >> 1). The random walk performed by the molecules introduced in the void space of the porous medium is directly linked to the Knudsen number and allows one to determine the effective diffusivity tensor $\underline{\underline{D}}$ at any value of *Kn*.

The numerical procedure, applied with 10,000 walkers over 20,000 voxel-long walks (the image edge size was 141 voxels) has been validated in comparison with analytical results [16] in bulk diffusion regime for square and hexagonal regular arrays of parallel cylinders, and by comparing with numerical results in Knudsen regime for a square array of parallel cylinders [22] with differences inferior to 5% in all cases, the worst ones lying close to the percolation threshold.

Results are shown in Figs. 4 and 5 in terms of tortuosities for Knudsen and ordinary transport in transverse direction. Tortuosity $\underline{\underline{\eta}}$ is defined by:

$$\underline{\underline{\eta}} = \varepsilon D^{\mathrm{ref}} \underline{\underline{D}}^{-1} \tag{4}$$

where D^{ref} is a reference diffusivity, corresponding to the longitudinal diffusivity in a capillary the diameter of which is equal to the mean pore diameter. This reference value is computed using the Bosanquet formula [30]. When *Kn* has appreciable values, D^{ref} depends on an assessment of the pore diameter, which is available in the images as shown previously [10].

The results are then collected together in a porosity-transport property correlation [31]. To do so, gas diffusivity has been evaluated in a large number of

sub-images within the high-resolution 3D images of **CC0** and **CC1** (respectively 384 for sample **CC0** and 1176 for sample **CC1**). These high-resolution sub-samples have nearly the same physical size as the sub-samples in the low-resolution images (i.e., ~0.05 mm^3) so that correspondence between the microscopic correlation established from these extracts and the property of the sub-samples is valid.

The simulated longitudinal and transverse tortuosity of each sub-sample can be modelled in the form suggested by Tomadakis and Sotirchos [32], which is an extension of Archie's law:

$$\eta_i = \eta_i^{\lim} \left(\frac{\varepsilon_0 - \varepsilon_p}{\varepsilon - \varepsilon_p} \right)^{\alpha_i} \tag{5}$$

Using a unit initial porosity, the identified coefficients for continuum and rarefied limits in parallel and transverse direction are shown in Table I. In all cases the parameter fitting has been satisfactory, as indicated by the values of the R^2 correlation coefficient on Table I. It is worth to compare them with predictions for ideal media like 1D, 2D, and 3D random overlapping fibre arrangements [32,21], and regular arrays in the bulk diffusion regime [16] (Note that estimates in Knudsen regime cannot be cast into the form of Eq. (5) because of the "infinite horizon effect" that would yield an infinite value for D^{ref}). Table I shows that the percolation porosity matches well the situation for 3D random fibre arrangements. The tortuosity limits in Knudsen regime lie between values for 1D and 3D random arrangements, and the characteristic exponent is close to the value for a 1D random arrangement. In the bulk diffusion regime, the limit tortuosity differs from all ideal predictions by only 9%; on the other hand, the characteristic exponent is neatly lower than all values for random media and higher than values for regular 1D arrays.

However the estimates for the regular arrays using Eq. (5) are not very accurate with respect to the original formulae.

From Figs. 4 and 5 and Table I, it is seen that there is not a large difference between samples **CC0** and **CC1** in Knudsen transport. On the other hand, for binary diffusion, an apparently more marked difference appears between samples **CC0** (very similar to a 1D random arrangement) and **CC1** (much closer to a 3D random arrangement). However, the ordinate scale is linear in Fig. 5, while it is logarithmic for Knudsen diffusion (Fig. 4), denoting a much larger sensitivity to fibre arrangement in the latter regime.

3.4.2. Verification of the locally orthotropic character

The computational results yield three values of tortuosity, the lowest being close to 1 and defining the direction parallel to fibres, and the other two related to the perpendicular direction. These two values should be equal if the medium were locally orthotropic, but it is generally not the case. The error made when one considers the medium as orthotropic is quantified by the relative difference of these two values :

$$\text{Error} = \frac{2\left|\eta_x - \eta_y\right|}{\eta_x + \eta_y} \tag{6}$$

Figure 6 is a histogram plot of the distribution of this error. It appears that sample **CC0** is very close to obey orthotropy, especially for continuum diffusion (the error average is 5%), and to a lesser extent for Knudsen diffusion (average error 22%); on the other hand, the discrepancy is stronger for **CC1**: 16% average error for binary transport, and 72% average error for Knudsen diffusion. However, this has not a strong influence on further computations, since the anisotropy ratio is much larger than this relative difference.

3.4.3. Computation of the viscous flow permeabilities

The values of the permeabilities are obtained from micro-scale images by an averaging technique, performing a Stokes-Darcy change of scale. At low Reynolds numbers, it is sufficient to describe the fluid movement by Stokes equations:

$$\begin{cases} -\mu\nabla^2\mathbf{v} + \nabla P = 0 \ \text{ in } \Omega_f \\ \nabla\cdot(\rho\mathbf{v}) = 0 \ \text{ in } \Omega_f \\ \mathbf{v} = 0 \ \text{ on } \partial\Omega_{fs} \end{cases} \tag{7}$$

Either by homogenization [33-34], or volume averaging [35], the macroscopic behaviour is proved to have the following form [36]:

$$\begin{cases} \nabla\cdot\langle\mathbf{v}\rangle = 0 \\ \langle\mathbf{v}\rangle = -\mu^{-1}\underline{\underline{B}}^{\text{eff}}\nabla\cdot\langle P\rangle^f \end{cases} \tag{8}$$

where $\underline{\underline{B}}^{\text{eff}}$ is the mesoscopic permeability tensor (m^2). Eq. (8) is the well-known Darcy law [37]. The numeric value of the permeability tensor may be attained as an average of a closure tensor $\underline{\underline{F}}$:

$$\underline{\underline{B}}^{\text{eff}} = -\varepsilon\langle\underline{\underline{F}}\rangle^f \tag{9}$$

where $\underline{\underline{F}}$, together with another vector closure variable **f**, satisfies the following problem [38]:

$$\begin{cases} -\nabla\mathbf{f} + \nabla^2\underline{\underline{F}} = \underline{\underline{Id}} & \text{in } \Omega_f \\ \nabla.\underline{\underline{F}} = 0 & \text{in } \Omega_f \\ \underline{\underline{F}} = 0 & \text{on } \partial\Omega_{fs} \\ \langle\mathbf{f}\rangle^f = 0 & \\ + \text{periodic B.C.'s for } \mathbf{f} \text{ and } \underline{\underline{F}} & \end{cases} \tag{10}$$

The resolution has been implemented in a finite-volume solver [39-40], using an artificial compressibility transient scheme.

It has not been possible to perform as many computations as for diffusion, because the large size of the sub-samples required too much computer memory;

rather, it has been chosen to select some sub-samples and correlate permeabilities to existing expressions. Of particular interest is the correlation with effective binary diffusivities suggested by Johnson et al. [41] and Tomadakis & Robertson [42]. If one defines viscous flow tortuosity $\underline{\underline{\eta}}_v$ as:

$$\underline{\underline{B}}^{\text{eff}} = \varepsilon \frac{d_p^{\,2}}{32} \underline{\underline{\eta}}_v^{\,-1} \tag{11}$$

Then, summarizing the theory of Johnson [41], the permeability-diffusivity correlation is given by the simple formula:

$$\eta_{v,i} = \frac{\eta_{b,i}}{M}\left(1 - \frac{\partial \ln \eta_{b,i}}{\partial \ln \varepsilon}\right)^2 \tag{12}$$

where M is a constant close to 1. Recalling the extended Archie's law result [Eq. (5)], one has the following prediction:

$$\eta_{v,i} = \frac{\eta_{b,i}}{M}\left(1 + \frac{\alpha_{b,i}\varepsilon}{\varepsilon - \varepsilon_p}\right)^2 \tag{13}$$

The best found value for M in perpendicular direction is 0.95. Figure 7 is a plot of the tortuosities in perpendicular direction predicted with Eq. (13) against the value computed directly. Even though some scatter is present, the agreement is satisfactory on a broad enough tortuosity range. On the other hand, results in parallel direction are somewhat underestimated through this method; they match more satisfactorily the correlations given by van der Westhuizen and Du Plessis [25] and by Tomadakis and Robertson [42] for random arrays of nonoverlapping parallel cylinders, as illustrated in Fig. 8. Also reported is the Kozeny relationship identified by Gutowski et al. [43], with a less satisfactory agreement.

3.5 First change of scale from ideal media: local thermal conductivity

Heat conduction in carbon/carbon composites is strongly related to the precise nature of the carbon-based components (carbon fibre and pyrocarbon deposit), as well as to the gas. The thermal conductivity of carbon fibres is not easy to measure and is a subject of research [44-45]. In lack of precise measurements, we will refer to studies conducted by Sauder [46], who deduced from electrical resistivity measurements the longitudinal conductivity of carbon fibres. According to Jumel et al. [47], the thermal conductivity of carbon fibres lies between 4 and 10.5 $W.m^{-1}.K^{-1}$. A value of 5 $W.m^{-1}.K^{-1}$ has been retained for this study. The values determined by Jumel *et al.* [47,48] for the pyrocarbon deposit have been used. However, it is worth mentioning that there is a great variety of pyrocarbons, with different nanotextures and consequently various thermal properties. The values taken for this work are summarized in Table II. Of course, the gas thermal conductivity is markedly lower than all other values, and its possible variation with pressure and temperature may be safely neglected.

It appears immediately that the heat conductivity of the solid phases is strongly anisotropic; moreover, the pyrocarbon deposit displays a cylindrical symmetry: this renders extremely difficult a direct simulation from CMT images, since the central axis of every fibre should be determined before assigning a local value of the heat conductivity to any voxel. So, it has been decided to provide a somewhat less satisfactory but readily feasible estimation based on a square array of parallel cylinders. The periodic unit cell is described in Fig. 9.

The longitudinal conductivity $\lambda_{i,//}$ of the i^{th} cell is directly given in analytical form by a law of mixtures (for conductances in parallel), where $\phi_{fi,i}$ is the fibre volume fraction:

$$\lambda_{i,//} = \varepsilon_i \times \lambda_{gas} + \phi_{fi,i} \times \lambda_{fi,//} + (1-\varepsilon_i - \phi_{fi,i}) \times \lambda_{pyC,//} \tag{14}$$

At the raw state (sample **CC0**), the material is only made of fibres and pores so that $\phi_{fi,i}$ is simply $(1-\varepsilon_i)$. When the material is partially infiltrated, the local fibre fraction can be deduced from the local porosity by using the following proportionality relation:

$$\phi_{fi,i} = (1-\varepsilon_i)\frac{\langle \phi_{fi} \rangle}{\langle \phi_s \rangle} \tag{15}$$

where $\langle \phi_{fi} \rangle$ is the average fibre fraction in the material and $\langle \phi_s \rangle$ the average solid fraction. The ratio between these two values is about 0.643 for **CC1**.

For the transverse conductivity of **CC0**, the following analytic equation of Perrins et al. [16] is suitable:

$$\frac{\lambda_{i,\perp}}{\lambda_{gas}} = 1 - \frac{2\phi_{fi,i}}{Q + \phi_{fi,i} - \dfrac{0.305827\left(\phi_{fi,i}\right)^4 Q}{Q^2 - 1.402958\left(\phi_{fi,i}\right)^8} - \dfrac{0.013362\left(\phi_{fi,i}\right)^8}{Q}} \quad \text{with } Q = \frac{1+\lambda_{fi,\perp}/\lambda_{gas}}{1-\lambda_{fi,\perp}/\lambda_{gas}} \tag{16}$$

For **CC1**, constituted of three phases, the situation is more difficult. To partially circumvent this, the transverse conductivity of a square array of cylinders covered by an anisotropic deposit [see Fig. 9(a)] has been computed. The radius taken for the cylinders is 3.98 μm and the deposit thickness 2.6 μm (values assessed in [10] and confirmed by SEM micrographs). Computations have been performed for volume fraction of fibres lying between 0 and the percolation threshold for such an arrangement (i.e., 78.5%). The flux/force correlation method [49-50], solved by a finite element software package, has been used. The results established for **CC1** follow Eq. (16) if $\phi_{fi,i}$ lies below a given threshold $\phi_{fi,t}$, and in the converse case is:

$$\lambda_{i,\perp} = \frac{\text{-}0.0141}{\phi_{fi,i} - \phi_{fi,t}} - 143.1\left(\phi_{fi,i}\right)^2 + 137.9\phi_i^f \text{-}12.8 \tag{17}$$

Expressions (16) and (17) are plotted at Fig. 10(a). They are converted into a porosity-conductivity correlation for **CC1** as plotted at Fig. 10(b).

3.6 Second change of scale: Computation of "large scale" properties

3.6.1 Strategy

The second change of scale consists in determining the effective property in large-scale images, the size of which is broad enough to ensure adequate representation of the whole porous medium. Whatever the property, this is computed by the same method, since all integrated expressions have the same flux-force formalism.

Details of the application of the volume averaging method [35] to the process of heat conduction in two- and three-phase systems is described in [51]. In their work, focused on the first change of scale, spherical conductivity tensors are considered for each phase. However, the change of scale procedure for another diffusive process, namely the single phase flow in heterogeneous porous media (two-region media and heterogeneous porous media with continuously varying properties), leads to similar results in which the permeability tensors considered are not necessarily spherical [52]. The extension of their results to multi-region models is rather straightforward. In all the works cited above the averaging process and the closure is based upon an equilibrium assumption, assuming that a single large-scale equation, therefore a single large-scale effective property, is sufficient for a correct macroscopic description of the phenomena.

The averaging process [53] leads to the following closure problem:

$$\begin{cases} \underline{\underline{\lambda}}_i : \nabla\nabla \mathbf{b}_i = 0 \qquad \text{on } \Omega_i, \quad i = 1,...n \\ \mathbf{n}_{ij} \cdot \left[\underline{\underline{\lambda}}_i \cdot \nabla \mathbf{b}_i + \underline{\underline{\lambda}}_i \right] = \mathbf{n}_{ij} \cdot \left[\underline{\underline{\lambda}}_j \cdot \nabla \mathbf{b}_j + \underline{\underline{\lambda}}_j \right] \qquad \text{on } \partial\Omega_{ij} \\ \mathbf{b}_i = \mathbf{b}_j \qquad \text{on } \partial\Omega_{ij} \\ \{\mathbf{b}\} = \frac{1}{\Omega} \sum_i \int_{\Omega_i} \mathbf{b}_i \mathrm{d}V = 0 \\ + \quad \text{periodic B.C.'s} \end{cases} \tag{18}$$

where Ω is the large-scale averaging volume composed of n regions (Ω_i, i = 1,..,n). The tensor $\underline{\underline{\lambda}}_i$ is the conductivity tensor for the i^{th} region (Ω_i), $\mathbf{b}_i$ is the closure variable defined in Ω_i. The interface between regions Ω_i and Ω_j is denoted $\partial\Omega_{ij}$. The large-scale average of a quantity ψ is defined as

$$\{\psi\} = \frac{1}{\Omega} \int_{\Omega} \psi dv \tag{19}$$

The effective conductivity tensor is given explicitly as a function of the closure variable **b** and local conductivities resulting from the first change of scale:

$$\underline{\underline{\lambda}}^* = \left\{ \underline{\underline{\lambda}} \right\} + \left\{ \tilde{\underline{\underline{\lambda}}} : \nabla \mathbf{b} \right\} \tag{20}$$

Where $\tilde{\underline{\underline{\lambda}}}$ is the conductivity deviation resulting from the decomposition of the local conductivity as $\underline{\underline{\lambda}} = \left\{ \underline{\underline{\lambda}} \right\} + \tilde{\underline{\underline{\lambda}}}$

The closure problem has been solved numerically using a continuous-flux, locally conservative finite volume method with a 27-point scheme proposed by Edwards and Rogers [54]. Indeed, the typically employed seven-point stencil cannot be used in this particular case of full tensor local conductivity. A thorough description of the method can be found in Cherblanc et al. [55].

At advanced stages of densification, the pore space exhibits only one degree of heterogeneity with respect to gas transport, since the contribution of micro-pores vanishes because of size reduction and of connectivity loss. Thus, a simple change

of scale can be designed for the computation of gas transport properties of such a material. However, since heat transport is not ensured by the pore space, this assumption is not true for the computation of thermal conductivity. For the computation of gas transport properties, 3 extracts with edge size equal to 200 voxels were selected within low-resolution images of **CC1b** and **CC2.** Sub-images were segmented to separate the solid phase from the pore space. Effective diffusivities are then computed using the random-walk algorithm described previously. For the effective permeability tensor, the algorithm developed by Anguy and Bernard [39-40] presented above has been used again.

The results have been compared to experimental measurements as far as possible; data were available on thermal conductivity [56] and on pure gas transport [57-58]. The latter properties have been determined at LCTS on a simple traditional experimental setup, inspired from [59,15] using argon, with a calibrated gas flow meter, a pressure difference gauge, and a terminal low-pressure regulation unit (vacuum pump and vane). From steady-state flow versus pressure difference curves obtained at various average pressures, the permeability and Knudsen diffusion coefficient were recovered [60-61].

3.6.2 Results

Figure 11 is a plot of computed Knudsen diffusivities as compared to experimental determinations performed at LCTS; at macro-scale, the parallel and perpendicular directions now refer to the cloth stacking, and not to the fibre orientations. The calculated values are in qualitative agreement with the experiments, but there is a tendency to underestimate at high porosities. Many reasons may be put forward. First, the experimental determinations are of limited reliability, because of the small values of the flows involved. Also, the CMT-based

estimation relies on the assumption of linear relationship between local greyscale level and density, which is not exactly verified because of the presence of a certain amount of phase contrast – such an artefact is more pronounced for the most porous samples.

Even if there is no experimental data available on binary diffusion, it is of interest to investigate the effect of the Knudsen number on the effective tortuosity. Figure 12 is a plot of the estimated values for samples **CC0** and **CC1**, showing : (i) a very low value for all tortuosities, as can be expected in a very porous medium, and (ii) a monotonous evolution for **CC1** while there is a hump for **CC0**. The latter behaviour had been reported in [22] for regular unidirectional arrays of cylinders with large porosity. A direct interpretation for it is that the mean free path is drastically increased when going from the transitional to the rarefied regime, because a very open structure provides longer free paths; indeed, when it is too open, convergence of the effective transport law (i.e., flux/gradient relationship) to classical Knudsen diffusion is not even ensured.

Figure 13 is a plot of computed versus measured permeabilities, showing qualitative agreement, and a marked discrepancy for high-porosity samples. The aforementioned limitations on the image-based evaluation method appear again, more or less in the same way as for Knudsen diffusion.

Figure 14 is a plot of computed *vs.* measured heat conductivities, showing an excellent agreement in both directions.

Summarizing the results of Figs. 11, 13, and 14, it can be said that the porous medium family corresponding to the C fibre preform at various stages of infiltration has been correctly assessed on all transport (and geometrical) properties. Of these transport properties, Knudsen transport has been found to be the most sensitive to

infiltration, followed by viscous flow and finally by thermal conductivity. This is in agreement with the physicist's intuition: first, thermal conductivity contrasts are known to be low, *e. g.* compared to electrical conductivity or any other property; second, Knudsen transport displays in many media a stronger tortuosity increase with diminishing porosity than the other gas transport parameters, because of the rapid view-factor reduction for direct transport as soon as a conducting medium is not straight. This last fact is confirmed here again both numerically and experimentally.

4 Conclusion and outlook

Raw and partially infiltrated carbon-carbon composite preforms have been scanned by high-resolution synchrotron radiation X-ray Computerized Micro-Tomography (CMT). The quality of the images had been assessed on geometrical quantities in a previous work [10]. Here, the images of the pore space have been produced at two distinct resolutions and have been used for the computation of transport properties: heat conductivity, binary gas diffusivities, Knudsen diffusivities, and viscous flow permeabilities. It is the first attempt to perform such a simultaneous evaluation directly from tomographic scans on C/C composites, mostly because: (i) very high-scale 3D images with adequate phase identification were not previously available, and (ii) the bimodal nature of the samples implies to develop a double-change-of-scale strategy.

The numerical evaluation of fibre-scale gas diffusivities in continuum and rarefied regime in the studied media compare favourably with estimates on ideal media families like 1D, 2D and 3D random fibre arrays. Viscous flow permeabilities have also been assessed and compared to available correlations for 1D random fibre arrangements, showing excellent agreement with the van der Westhuizen-Du Plessis

[25] and Tomadakis [42] models in parallel direction, and with the Tomadakis model in perpendicular direction. Further work at this scale concerns primarily the direct computation of thermal conductivity or diffusivity: this requires a separate segmentation of fibres and matrix, with a local assignment of material principal axes of anisotropy; this work is currently under way.

Computations at a larger scale have been carried out for all transport properties; it has been possible to compare directly the results with experimental data acquired on the same materials for Knudsen diffusivity, viscous flow permeability, and thermal conductivity. Despite an excellent agreement on all properties, many improvements may be suggested. First, images of high-porosity samples do not totally verify the greyscale-porosity correlation because of phase contrast – tomographs taken at the same resolution with a classical x-ray source would be of better exploitation.

Second, concerning gas transport, there is a need for a unified solver, able to manage high- and low-porosity samples in the same way. This could be achieved for viscous transport by writing down a Stokes-Brinkman microscopic problem, followed by the traditional change-of-scale procedure. This is also under way.

Third, replacing the provided analytical estimates for fibre-scale thermal conductivity by estimates based on the results of a full numerical computation (as mentioned above), would increase greatly the confidence in the quality of this CMT-based approach.

Taking note of these improvement suggestions, it is claimed here that CMT-based simulations are a powerful tool for the assessment of all properties of interest in C/C composite preforms during infiltration, which is of great importance in CVI process modelling. By combining process-scale approaches and the correlations

obtained in the past [10] and present works, it is possible to carry out a multi-scale simulation. Repeating the procedure on several fibre arrangements would allow comparing them one to each other in the sense of “infiltrability”, that is, of the predisposition to receive as much matrix as possible in given conditions.

Finally, it is also possible to develop numerical methods for the detailed simulation of in-pore matrix deposition; utilizing them in a suitable double-change-of-scale strategy would also allow to assess quantitatively the notion of “infiltrability”. This is yet another work in progress.

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	Percolation porosity	***Rarefied (Knudsen) regime***		***Continuum (ordinary) regime***	
		Limit tortuosity	***Exponent***	***Limit tortuosity***	***Exponent***
Parallel					
Samples CC1 and CC0	**0.04**	**0.817**	**0.126**	**1.0**	**0.107**
Correlation coefficient R^2		***0.95***		***0.99***	
2D random	0.11	1.149	0.954	1.0	0.521
1D random	0	0.549	0	1.0	0.
Transverse					
Samples CC1 and CC0	**0.04**	**1.411**	**1.047**	**1.09**	**0.465**
Correlation coefficient R^2		***0.93***		***0.99***	
3D random	0.037	1.444	0.921	1.0	0.661
2D random	0.11	1.780	1.005	1.0	0.785
1D random	0.33	1.747	1.099	1.0	0.707
1D hexagonal	0.093			1.0	0.335
1D square	0.215			1.0	0.358

Table I : Comparison of parallel and transverse tortuosity-porosity correlation parameters [Eq. (12)] obtained from high-resolution tomographic images with estimations on ideal fibrous media [32]

Component	*Thermal conductivity* ($W.m^{-1}.K^{-1}$)	
Fibre	Longitudinal :	$\lambda_{fi,//} = 20$
	Transverse :	$\lambda_{fi,\perp} = 4$
Pyrocarbon	Longitudinal :	$\lambda_{pyC,//} = 114$
	Radial :	$\lambda_{pyC,\perp} = 44$
	Orthoradial :	$\lambda_{pyC,//} = 114$
Gas	Isotropic :	$\lambda_{gas} = 0.13$

Table II : Thermal conductivities of C/C composite components

Sample	*Method*	*Pore volume fraction* ε **(%)**	*In-plane heat conductivity* $\lambda_{//}$ **(*W.m*$^{-1}$.*K*$^{-1}$)**	*Transverse heat conductivity* $\lambda_{\perp}$ **(*W.m*$^{-1}$.*K*$^{-1}$)**
CC0	Computed in sub-samples	72.7	0.62	0.43
		73.6	0.61	0.37
		73.4	0.57	0.40
	Computed, average	**73.2**	**0.60**	**0.40**
	Measured	**73.0**	**0.60**	**0.38**
CC1	Computed in sub-samples	60.6	3.74	1.44
		58.7	3.74	2.05
		55.3	4.17	2.14
	Computed, average	**58.2**	**3.88**	**1.88**
	Measured	**56.5**	**5.00**	**1.75**

Table III: Simulated thermal conductivities obtained with the double change of scale procedure and measurements performed by Demange and Laizet [56]