Simulating permeabilities based on 3D image data of a layered nano-porous membrane

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Abstract

The macroscopic properties of materials are strongly influenced by their microstructure. Essential micro-structural information can be obtained from high-resolution 3D images, usually obtained by computed tomography. Here, a layered nano-porous ZrO\textsubscript{2} Al\textsubscript{2}O\textsubscript{3} membrane for fluid filtration is investigated, whose two finer layers can not be resolved properly by computed tomography. 3D image data of these structures are therefore obtained by the combination of focused ion beam sectioning and scanning electron microscopy. Reconstruction of porous structures from this type of image data and subsequent numerical simulation of flow properties within the reconstructed structure bear several sources of error. Using stochastic geometry models for the solid component, we examine more closely the error due to the typical anisotropic voxel setting as well as the error due to the also typical uneven sample shape.

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1. Introduction

The microstructure of materials influences decisively their macroscopic properties. Essential micro-structural information can be obtained from high-resolution 3D images using appropriate methods for quantitative image analysis. Computed tomography nowadays can yield 3D images at lateral resolutions down to the 100 nm range. For finer structures, the combination of sequential slicing by a focused ion beam (FIB) and imaging by scanning electron microscopy (SEM) – FIB-SEM – is the method of choice. Ceramics have been imaged by FIB-SEM as early as 2004, see [1]. In [2, 3], 3D imaging techniques as well as microstructure characterization and simulation for porous ceramics is reviewed.

For highly porous structures, FIB-SEM imaging poses a particular segmentation problem: Reconstruction (or segmentation) of the 3D solid structure from the stack of SEM images is hampered by structural information from lower slices shining through the pores in the current one. Dedicated segmentation algorithms have been developed [4, 5] for FIB-SEM images of highly porous carbon based structures, validated using synthetic image data [6, 7], and successfully applied for numerical simulation of macroscopic materials properties, even in multi-scale problems [8, 9].

Conductivity and diffusivity in multiscale structures for battery/fuel cell applications are simulated e.g. in [10, 11, 12]. Recent work identifies geodesic tortuosity and constrictivity [13, 14] to yield geometric information that is
highly correlated with effective conductivity and permeability. However, caution is needed w. r. t. quality of the reconstructed 3D structure, see e. g. [15] who show that insufficient resolution causes strong deviations of the diffusivity and conductivity.

The subject of the current investigation is a layered nano-porous ZrO$_2$ Al$_2$O$_3$ membrane for fluid filtration, see Figure 1 for a cross-section. 3D image data of its two finer layers is obtained by FIB-SEM. The coarsest third layer is imaged by micro-computed tomography based on synchrotron radiation. Reconstruction is quite different from the one for the carbon based materials as non-conductive ceramics are investigated. Moreover, the finest, top layer has to be resolved extremely highly and features a much higher porosity than the materials considered e. g. in [2]. These two facts cause additional sectioning and imaging artifacts. On the other hand, signals from both secondary and back-scattered electrons are available, allowing to reconstruct the finest, upper most layer of ZrO$_2$ by combining the secondary electron (SE) and the energy-selective back-scatter electron (EsB) signals via morphological reconstruction by dilation.

The flow properties within the membrane can be simulated numerically based on the segmented solid components. However, deshearing and alignment result in non-cubic voxels even if the original slice thickness for the FIB is chosen exactly as the resolution of the SEM. Moreover, typically, the field of view imaged is much shorter in slicing direction than in the other two coordinate directions. We use Boolean models of spheres in order to study the error caused by these two sources.
2. Material and methods

2.1. Nano-porous membrane

We study a nano-porous membrane with flat setup and asymmetric structure (see Figure 1) as typically used as insert for fluid filtration.

Figure 1: SEM image of the polished membrane cross section showing all layers. Here, the back-scattered electron signal is used.

The sample is supported by a 950 µm thick layer of highly porous Al₂O₃ with particle sizes in the range of 5 – 10 µm. This substrate is prepared by tape casting and sintered at 1650°C.

The following layers adjusting the porosity are generated by spin coating. First, two Al₂O₃ layers (layers 2 and 3) with a thickness of approximately 10 – 12 µm and finely graduated particles are deposited. Finally an 8 – 10 µm thick nano-porous ZrO₂ layer (layer 1) is added. After every spin coating step, the membrane is sintered at temperatures between 1200 and 1400°C. Layer 1, the active membrane layer, is formed by very fine ZrO₂ powders
with particle sizes in the range of 10 – 50 nm (Figure 2). For details on the ceramic processing methods tape casting and spin coating see e.g. [16]. In

![Figure 2: SEM images of the ZrO$_2$ powder used for generating layer 1 and the resulting micro-structure. Gray values represent secondary electron intensities as determined by an in-lens detector.](image)

(a) Nano-crystalline ZrO$_2$ powder (b) Layer 1

the sintered sample, grain sizes are in the range of 50 – 100 nm and pore sizes in the range of 10 – 100 nm with a clear fine to coarse transition from top layer 1 towards the support.

2.2. FIB-SEM imaging

A Crossbeam NVision 40 Field Emission Scanning Electron Microscope (FE-SEM) by Carl Zeiss is used for the FIB-SEM measurements. The advanced tomography package ZEISS Atlas 5 3D Tomography that enables recording of high resolution 3D data sets with selected voxel sizes and simultaneous imaging using two different sets of SEM conditions is an integral part of this equipment. The process control by SEM AutoTune and 3D Tracking Marks enables automated measurements.
In order to avoid charging processes during SEM imaging of the poorly conductive ceramic membrane sample, the whole sample surface is coated by an approximately 500 nm to 1 µm thick gold layer. An additional 1 – 2 µm platinum layer is applied locally by FIB deposition to smooth the rough porous surface. Finally, the Atlas 5 SEM AutoTune and 3D Tracking Marks are deposited. See Figure 3 for a thus prepared cross-section.

Figure 3: Setup for automated FIB-SEM measurements on the membrane, here layer 1.

Both layers 1 and 2 are sliced in membrane thickness direction. That means, for the FIB-SEM image data, the SEM images are the $xy$-slices with $y$ being the membrane thickness direction while $z$ is the direction in which
the 2D images are stacked. Thus, given the production methods described above, the structure can be expected to be invariant w. r. t. rotations about the $y$-axis.

Layer 1 is imaged at SEM pixel size 3 nm and FIB slice thickness 6 nm, while layer 2 is imaged with SEM pixel size and slice thickness 20 nm. The raw FIB-SEM stack for layer 1 consists of 204 slices of $3400 \times 2700$ pixels, the one for layer 2 of 300 slices of $1500 \times 600$ pixels.

For both, first and second layer, secondary electron (SE) as well as energy-selective back-scatter electron (EsB) signals are detected, see Figure 4. The SE images feature strong gray value contrast while suffering from pronounced curtaining and charging artifacts, too. In particular for layer 1, where pixel sizes of 3 nm are reached, the FIB cutting clearly induces a surface roughness in approximately the same size range as the structure itself, see Figure 5(b). The EsB images are free of these artifacts but more blurred. For layer 2, a higher acceleration voltage is used – 3.0 kV instead of 1.5 kV for layer 1. This results in much stronger charging in the SE images, but at the same time much sharper edges and overall better contrast in the EsB images, see Figure 6(b).

2.3. Reconstruction of the FIB-SEM image data

The FIB-SEM stack is desheared using a bilinear interpolation that scales the voxel size by $\sin(52^\circ)$ in $y$-direction. In theory, the angle applied in this deshearing should be the one formed by FIB and electron beam. However, here, it is estimated from the image data and found to differ from the former slightly by $2^\circ$. Then, the images are aligned using the StackReg plugin from ImageJ [17]. StackReg aligns by propagating the image information from
one slice to the subsequent one. More precisely, for each slice, a pyramid from fine to coarse is created by low pass filtering and sub-sampling [18]. Within each pyramidal level, the mean squared differences of voxels’ gray values in consecutive slices are minimized recursively to determine the best aligning transformation. For porous structures, the alignment is to some extent misguided by the shine-through artifacts causing a distortion most prominently visible as a parallelogram distortion in the $yz$-plane, see e. g. [19]. This distortion is subsequently corrected by TransformJ from ImageJ [20]. Finally, the whole stack is rotated back by $52^\circ$ about the $x$-axis. Due to the interpolation needed for the described affine transformation, the voxels of the resulting 3D images are not necessarily cubic after processing, even if slicing distance and pixel size for the SEM imaging coincide. The voxels in layer 1 have edge lengths $3\text{ nm} \times 2.4\text{ nm} \times 6\text{ nm}$, those in layer 2 $20\text{ nm} \times 20\text{ nm} \times 16\text{ nm}$. The corresponding 3D images have sizes $2.97\text{ \mu m} \times 2.97\text{ \mu m} \times 1.14\text{ \mu m}$ for layer 1 and $4.1\text{ \mu m} \times 4.1\text{ \mu m} \times 3.3\text{ \mu m}$ for layer 2.

Additionally, we use versions of the images interpolated onto a cubic lattice by MatLab’s interp3 with the option “cubic” [21]. The voxels in the isotropic versions have edge lengths $3\text{ nm}$ for layer 1 and $20\text{ nm}$ for layer 2.

The SE images are morphologically opened by a cuboidal structuring element of $3 \times 2 \times 2$ voxels to reduce the curtaining artifacts. The structuring element is chosen to be larger in $x$-direction as these artifacts appear in the $xy$-plane as thin stripes along the $y$-direction. Both, the SE and EsB images are denoised by a $3 \times 3 \times 3$ median filter. To these filtered images, manually chosen global gray value thresholds are applied.

In order to reconstruct the spatial solid structure for layer 1, morpho-
logical reconstruction by dilation \cite{22} is applied to the pore space. That is, both thresholded images - SE and EsB - are inverted yielding the pores as foreground. The SE signal is particularly topography sensitive (see e.g.\cite{23}) and thus captures the pores very well. However, due to the curtaining and waviness caused by the FIB, pores appear locally too large. In particular, they are artificially elongated in \textit{y}-direction. In the thresholded EsB image, the solid component is too strong, e.g. pores are filled due to shine through artifacts. The captured pores are however surely belonging to that phase. Thus, the EsB pore image is chosen as the marker image.

The mask image for the morphological reconstruction is the voxel-wise maximum of the two pore images. That way, voxels for which both thresholded images agree are assigned to the respective component in the reconstruction result, too. The actual reconstruction is an iterative process, starting with morphologically dilating the EsB pore image by the elementary isotropic structuring element. Then the voxel-wise minimum of the resulting dilated pore image and the mask image is taken. This basic procedure is iterated until the result does not change anymore. Thus, the resulting image is forced to remain below the mask image.

The reconstruction of the solid structure of layer 1 is illustrated in Figure 5. Figure 7 shows volume renderings of sub-volumes of the reconstructed samples.

For layer 2, the SE image is not used due to the strong charging and curtaining. Instead, the EsB image, that features a much better gray value contrast than the EsB image for layer 1, is binarized by a manually chosen global gray value threshold, see the close-ups in Figure 6.
2.4. Microtomography using synchrotron radiation

Layer 3 is too coarse for FIB-SEM imaging. It is therefore scanned at beamline ID19 of the European Synchrotron Radiation Facility (ESRF) in Grenoble, France [24], with a resulting voxel edge length of 160 nm and a propagation distance of 6 mm between sample and detector. Leaving a drift space between the sample and the detector emphasizes interfaces within the sample due to X-ray refraction (propagation-based or inline phase-contrast). Two tomographic reconstructions are computed – one just exploits the edge-enhanced contrast due to the propagation of the partial coherent wave fronts after transmitting the sample. Standard filtered back projection results in an edge-enhanced volume image [25]. The second volume image is achieved by performing phase-retrieval on the projection images prior to reconstruction using a modified single-distance approach by Paganin et al. [26]. Example slices are shown in Figure 8(a) and (b). The edge-enhanced image is excellently suited to mark borders between material phases while the phase-retrieved image offers volume information. During tomography, the sample was rotated about an axis in membrane thickness direction. This is defined to be the $y$-axis from now on, in order to keep consistency with the coordinate system used for FIB-SEM. For geometric analysis, a volume of $1600 \times 1600 \times 1700$ voxels corresponding to $256 \mu m \times 256 \mu m \times 272 \mu m$ is used, while flow simulations are performed on a $650^3$ voxel cubic sub-volume of edge length $104 \mu m$.

2.5. Segmentation of the SRµCT image data

The two tomographic reconstructions are combined by simple voxel-wise summation and denoised by a $3 \times 3 \times 3$ median filter. The final segmentation
is then achieved by a global gray value threshold – 101% of the one suggested by Otsu’s method [27], see Figure 9. A rendered sub-volume of the segmented solid component is shown in Figure 10. Note that the gray value histogram does not feature a clear local minimum separating solid structure and pores. Due to the partial volume effect, all surface voxels (approximately 10%) are at risk to be mis-classified. However, varying the threshold from 99% to 102% of Otsu’s threshold – that is from 30 412 to 31 333 – changes the porosity only from 42.6% to 47.2%. Here, 101% of Otsu’s threshold was chosen by visual impression.

2.6. Stochastic geometry models

Realizations of stochastic geometry models are a tool for validating numerical simulations of macroscopic properties, see e. g. [28]. In particular, they allow to investigate various error sources in the final permeability simulations separately that occur inextricably combined in real data. Here, we therefore exploit stochastic geometry models but do not aim at modeling the membrane layers in the strict sense of e. g. [29]. We use a simple model – a stationary Boolean model whose typical grain is a ball of constant radius $R$. This model is defined as the union set of a system of balls with radius $R$ whose centers are simulated according to a stationary Poisson point process with intensity (mean number of points per unit volume) $\lambda$. To ensure some resemblance with the membrane layers, the values of $R$ and $\lambda$ are estimated using Miles’ formulas [30, 31, 32] relating the densities of the intrinsic volumes, in particular the solid volume fraction $V_{fr}$ and the specific surface area
$S_V$ to the model parameters:

\[ V_V = 1 - e^{-\lambda_V} \]
\[ S_V = e^{-\lambda_V} \lambda S \]

where $V = 4/3\pi R^3$ and $S = 4\pi R^2$ denote the volume and the surface area, respectively, of a ball of radius $R$.

2.7. Flow Simulation

To simulate the flow behavior of the membrane, we use a multiscale flow model characterized by the membrane and the pore scale. On the membrane scale, there are three homogeneous layers, where the flow can be described by Darcy’s law,

\[ \mathbf{v}_D = -\frac{\kappa}{\mu} \cdot \nabla p_D, \]

where $\nabla p_D$ is a macroscopic pressure drop in the layer, $\mu$ is the dynamic viscosity of the fluid, $v_D$ is the Darcy velocity and $\kappa$ is the effective permeability of the respective layer.

To compute the effective permeability $\kappa$ for each of the layers, we simulate the flow of an incompressible fluid through the layer on the pore scale. It is assumed that this model is correct, provided that the Reynolds number is small, the pore space of the medium is fully saturated and the typical pore diameter is much smaller than the thickness of the respective layer. The computational domain is given by the segmented image data or synthetically generated model realizations as described in Sections 2.3, 2.5, and 2.6, respectively. The computational domain $\Omega$ is divided into a solid phase $S$ and the pore space $P$. Then we solve the Stokes equations in the pore space of
the layers, i.e. we solve the equations

\[ \mu \nabla^2 v - \nabla p + f = 0, \quad x \in P \]

\[ \nabla \cdot v = 0 \]

for \( v(x), p(x) \) in the pore-space, with the no-slip interface condition

\[ v(x) = 0, \quad x \in \partial S. \]

Also, in the solid phase of the domain, we set:

\[ v(x) = 0, \quad x \in S. \]

For the computation of the effective permeabilities in the stochastic model realizations, periodic boundary conditions in the velocity and the pressure are assumed in all directions. These are:

\[ v(x + e_i L_i) = v(x), \quad x \in \partial \Omega, \]

\[ p^*(x + e_i L_i) = p^*(x), \quad x \in \partial \Omega, \]

where \( p^* \) is the periodic fluctuation of the pressure around its macroscopic gradient across the domain [33]. The macroscopic pressure drop is enforced by applying a volume force \( f_i = -\langle \partial_i p \rangle \) [34].

For the estimation of the permeability from the FIB-SEM and SR \( \mu \)CT images, Equations (1) are solved with symmetric boundary conditions, that is:

\[ v \cdot n = 0 \]

\[ \nabla p \cdot n = 0 \]
for the boundaries in transversal direction. The symmetric boundary conditions on the boundaries normal to the flow direction are enforced by mirroring the domain across this boundary and then applying periodic boundary conditions to the extended domain.

The effective permeability $\kappa$ can be computed from the solution of Equation (1) by relating the average pressure gradient through the domain to the Darcy velocity $v_D$, i.e.

$$v_{D,i} = \langle v_i(x) \rangle = -\frac{\kappa_{ij}}{\mu} \langle \partial_j p \rangle.$$

In all flow simulations the pressure drop in direction $i$ was fixed to $\partial_i p L_i = 0.02$ Pa and the fluid viscosity was fixed to $\mu = 0.0089$ Pa s, corresponding to water at 25°C, though any other choice of the parameters would yield the same result.

For all boundary conditions, the flow equations were solved using the SimpleFFT solver implemented in the software GeoDict [35]. The software uses a finite volume discretization and a SIMPLE projection scheme to solve the flow equations. The Fast Fourier Transform is used to accelerate the solution of the pressure correction equation.

Several sources of error have to be taken account of when comparing the permeabilities computed in realizations of the stochastic models with the ones computed in the reconstructed image data. Aside from error due to discretization of the equations and numerical accuracy of the solver, errors due to imaging artifacts and image segmentation, and boundary effects due to the finite image volume have to be considered. In order to quantify these errors, the same effects are reproduced in the synthetic data.

First, the permeability of the stochastic model is computed based on a
representative volume with periodic boundary conditions. Thus the resulting values are not biased by imaging errors and boundary effects and can serve as an accurate estimate of the model’s effective permeability. Second, permeabilities are computed after cropping the model realizations to the same dimensions as the image data and then solving equations with symmetric boundary conditions. Finally, model realizations are discretized on non-isotropic grids corresponding to those observed in the non-interpolated FIB-SEM images.

3. Results and Discussion

3.1. Image analysis

The following Table 1 contains the median pore and grain sizes from spherical granulometry as well as volume fractions and specific surface areas based on the reconstructed 3D FIB-SEM images for layers 1 and 2 and the segmented SRµCT image for layer 3, respectively. Moreover, as a measure for anisotropy, we report the proportions of the mean chord lengths in Table 2. All geometric characteristics are measured using MAVI [36] applying the simultaneous calculation of the densities of the intrinsic volumes from the frequencies of $2 \times 2 \times 2$ voxel configurations in the 3D binary image as described in [32]. The mean chord lengths are obtained that way, too, from the solid volume fraction and the area of the generalized projections in the respective direction, see [37, 5.3.3]. The spherical granulometry distribution of a random closed set $\Xi$ is a generalized volume weighted size distribution defined by successive openings with spherical structuring elements of growing size mimicking a sieving procedure. More precisely, the distribution is
given by the cumulative distribution function $G(r) = 1 - \frac{V_r(\Xi \circ B_r)}{V(\Xi)}$, where $\Xi \circ B_r$ denotes the morphological opening of $\Xi$ by a ball of radius $r$. As naive implementation in 3D is too time consuming, MAVI uses a more efficient algorithm based on the medial axis of the Euclidean distance image [38].

Anisotropy due to the FIB sectioning should be revealed by deviations of the mean chord length $\bar{\ell}_z$ in stack direction ($z$) from the mean of the mean chord lengths in the other two coordinate directions (anisotropy slicing). On the other hand, by design, there is a certain anisotropy to be expected in $y$-direction as this is the membrane thickness direction (anisotropy construction). For layer 1 however, $\bar{\ell}_x$ is significantly shorter than the other two due to the wavy FIB slicing artifacts while $\bar{\ell}_y$ and $\bar{\ell}_z$ coincide. That means, the FIB cut is not ideally planar as visible in Figure 5(b). Structure extending in $x$-direction is thus artificially cut-off occasionally causing an underestimation of $\bar{\ell}_x$. Layer 2 features the expected stretch in slicing direction with $\bar{\ell}_z$ being clearly larger than $\bar{\ell}_x$. The benefit of interpolation onto a grid with isotropic voxels is not obvious. For layer 1, interpolation reduces the differences in $x$- and $z$-directions considerably. For layer 2, however, interpolation seems to worsen the artificial anisotropy.

3.2. Geometric modeling

We use stationary Boolean models whose typical grain is a ball of constant radius $R$ as simple models for layers 1-3. The parameters $R$ and $\lambda$ are estimated using Miles’ formulas as given in Subsection 2.6. The results can be found in Table 3. The model realizations thus coincide with the real structure w. r. t. solid volume fraction and specific surface area. However, as clearly visible when comparing Figures 5(d), 6(d), 8(b) with 11, neither grain
Table 1: Selected geometric characteristics determined based on the segmented 3D images. “iso” refers to cubic voxels of edge lengths 3 nm for layer 1 and 20 nm for layer 2, “aniso” to cuboidal ones (3 nm \times 2.4 nm \times 6 nm for layer 1 and 20 nm \times 20 nm \times 16 nm for layer 2).

<table>
<thead>
<tr>
<th>layer</th>
<th>material</th>
<th>(V_{V}) [%]</th>
<th>(S_{V}) [(\mu m^{-1})]</th>
<th>median pore diameter</th>
<th>median grain diameter</th>
</tr>
</thead>
<tbody>
<tr>
<td>1, iso</td>
<td>ZrO(_2)</td>
<td>49.31</td>
<td>28.19</td>
<td>52.7 nm</td>
<td>49.2 nm</td>
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<tr>
<td>1, aniso</td>
<td>ZrO(_2)</td>
<td>49.89</td>
<td>26.59</td>
<td>–</td>
<td>–</td>
</tr>
<tr>
<td>2, iso</td>
<td>Al(_2)O(_3)</td>
<td>70.57</td>
<td>7.96</td>
<td>128.2 nm</td>
<td>216.9 nm</td>
</tr>
<tr>
<td>2, aniso</td>
<td>Al(_2)O(_3)</td>
<td>70.16</td>
<td>8.28</td>
<td>–</td>
<td>–</td>
</tr>
<tr>
<td>3</td>
<td>Al(_2)O(_3)</td>
<td>54.92</td>
<td>0.59</td>
<td>2.21 (\mu m)</td>
<td>2.58 (\mu m)</td>
</tr>
</tbody>
</table>

3.3. Flow simulation

3.3.1. Simulation on Image Data

The effective permeabilities computed on the image data for the three layers are shown in Table 4. For the reconstructed/segmented image data, symmetric boundary conditions are applied in all directions.

It is clear from the production process, that the permeabilities should be at least transversely isotropic, i.e. \(\kappa_{xx}\) and \(\kappa_{zz}\) should coincide. This means, that the anisotropy in the permeabilities must be caused by errors either
in imaging or processing. Computing on anisotropic voxels alters the result only slightly with the exception of $\kappa_{xx}$ for layer 1.

### 3.3.2. Simulation on Synthetic Model Data

In order to reproduce the distortions observed in the image data, the aforementioned sources of error are investigated by evaluating the permeabilities on realizations of the stochastic model with different boundary conditions, sample sizes and computational grids.

**Grid Error and Numerical Solver Error.** In order to estimate the error coming from the discretization of the domain, we conducted a small grid study on a cubic domain of edge length $a$ with a single sphere inclusion of diameter $a/2$ discretized with voxel sizes of $a/100$, $a/50$ and $a/25$. From this, we estimate the grid error as being below 1% in the permeability. In order to eliminate errors due to lack of numerical accuracy, the error threshold of the

<table>
<thead>
<tr>
<th>layer</th>
<th>$\bar{\ell}_x$ [nm]</th>
<th>$\bar{\ell}_y$ [nm]</th>
<th>$\bar{\ell}_z$ [nm]</th>
<th>anisotropy slicing $2\bar{\ell}_z/(\bar{\ell}_x + \bar{\ell}_y)$</th>
<th>anisotropy construction $2\bar{\ell}_y/(\bar{\ell}_x + \bar{\ell}_z)$</th>
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<tr>
<td>1, iso</td>
<td>63.84</td>
<td>72.59</td>
<td>72.31</td>
<td>1.06</td>
<td>1.07</td>
</tr>
<tr>
<td>1, aniso</td>
<td>64.29</td>
<td>74.64</td>
<td>86.59</td>
<td>1.25</td>
<td>0.99</td>
</tr>
<tr>
<td>2, iso</td>
<td>346.96</td>
<td>336.47</td>
<td>367.16</td>
<td>1.07</td>
<td>0.94</td>
</tr>
<tr>
<td>2, aniso</td>
<td>342.04</td>
<td>331.62</td>
<td>334.00</td>
<td>0.99</td>
<td>0.98</td>
</tr>
<tr>
<td>3</td>
<td>3520.20</td>
<td>3739.53</td>
<td>3795.00</td>
<td>$-$</td>
<td>1.02</td>
</tr>
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</table>

Table 2: Mean chord lengths as well as anisotropy as indicated by proportions of them. All values reported here are measured on the solid component.
Table 3: Parameters of the Boolean models of balls with constant radius with volume fraction and specific surface area as reported in Table 1 for layers 1-3.

<table>
<thead>
<tr>
<th>layer</th>
<th>dimensions [voxels]</th>
<th>( \kappa_{xx} ) ([\text{m}^2])</th>
<th>( \kappa_{yy} ) ([\text{m}^2])</th>
<th>( \kappa_{zz} ) ([\text{m}^2])</th>
</tr>
</thead>
<tbody>
<tr>
<td>1, iso</td>
<td>990 \times 990 \times 382</td>
<td>1.68 \cdot 10^{-17}</td>
<td>2.28 \cdot 10^{-17}</td>
<td>3.13 \cdot 10^{-17}</td>
</tr>
<tr>
<td>1, aniso</td>
<td>990 \times 1,050 \times 191</td>
<td>1.91 \cdot 10^{-17}</td>
<td>2.30 \cdot 10^{-17}</td>
<td>3.03 \cdot 10^{-17}</td>
</tr>
<tr>
<td>2, iso</td>
<td>205 \times 205 \times 165</td>
<td>3.66 \cdot 10^{-17}</td>
<td>3.35 \cdot 10^{-17}</td>
<td>4.79 \cdot 10^{-17}</td>
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<td>205^3</td>
<td>3.71 \cdot 10^{-17}</td>
<td>3.39 \cdot 10^{-17}</td>
<td>4.69 \cdot 10^{-17}</td>
</tr>
<tr>
<td>3</td>
<td>650^3</td>
<td>4.02 \cdot 10^{-14}</td>
<td>3.80 \cdot 10^{-14}</td>
<td>4.05 \cdot 10^{-14}</td>
</tr>
</tbody>
</table>

Table 4: Computed effective permeabilities of the image data for the three individual layers. Symmetric boundary conditions were applied throughout.

The solver was set to \(10^{-4}\).

**Representative Volume Error.** The second source of error investigated here is the sample size of the reconstructed three dimensional structure. Clearly, effective permeabilities can not be deduced correctly, if the reconstructed field of view is too small to represent the structure. An extensive study of the representative volume element (RVE) of the Boolean model with spheres has been conducted in [28]. To estimate the RVE for our image data, a study was conducted on model realizations with dimension 650^3. For each model layer,
10 periodic realizations were generated and the permeability was computed using periodic boundary conditions. Then, the 95% confidence interval was estimated for each sample, by taking the sample mean $\pm 2 \frac{SD}{\sqrt{n}}$, where $SD$ is the standard deviation of the sample and $n$ is the number of realizations of the model. The first rows in Table 5 for each layer show that the 95% confidence intervals are smaller than $\pm 2\%$ of the estimated values, which is in agreement with [28]. Hence, these values can be used as reference points, i.e. as “true” values for the model permeabilities and deviations from this estimate can be considered to be a methodological error.

**Boundary Conditions.** To investigate the effect of the selected boundary conditions, the simulations from the previous paragraph were repeated on the periodic realizations, using symmetric boundary conditions. The resulting permeabilities are listed in Table 5 (2nd row for each layer) and show a small systematic error in layer 2. Clearly, the relative discrepancies to periodic boundary conditions are below 2%. This is expected, since boundary conditions should have negligible effect for representative volume elements.

The error analysis for the µCT data of layer 3 is thereby completed, as for this kind of data neither strongly anisotropic regions of interest nor anisotropic voxel sizes usually occur.

**Sample Dimensions.** FIB-SEM imaging often results in reconstructed observation windows of strongly anisotropic dimensions. We investigate the effect of these anisotropic regions on the estimation of effective permeabilities by cropping the representative volume elements anisotropically, as in the image data. Hence, the model representing layer 1 was cropped to a dimension of $650 \times 650 \times 382$ voxels and then the permeability was computed. As shown in
Table 5: Computed effective permeabilities of the model realizations with varying boundary conditions, dimensions, and voxel sizes as discussed in Section 3.3.2.
Table 5, the permeability is underestimated in the directions perpendicular to the FIB slicing and slightly overestimated in the remaining direction. This is unexpected, since in $x$ and $y$ dimensions the sample should be representative. However, the same trend is observed in the image data.

To test the effect of the cropping on the model for layer 2, we cropped 10 samples to the dimension of $205 \times 205 \times 165$ and then computed the permeability with periodic and symmetric boundary conditions. The resulting permeabilities are shown in Table 5. As can be seen, the statistical error increases by a factor between three and ten, due to the smaller sample size. Even though for periodic boundary conditions, all confidence intervals include the reference value, a trend towards underestimating permeabilities can be observed when applying symmetric boundary conditions as opposed to periodic ones.

*Computations on Anisotropic Grids.* Finally, the effect of the anisotropic sampling typical in FIB-SEM imaging is investigated based on realizations of the Boolean models discretized to anisotropic grids and computing the permeabilities in these anisotropic control volumes. However, in order to avoid numerical issues due to too strongly differing voxel edge lengths, the strong grid anisotropy observed in layer 1 was mimicked by doubling each slice.

More precisely, the model representing layer 1 was discretized on an image with dimension $650 \times 650 \times 382$, corresponding to a voxel size of $3\text{ nm} \times 2.4\text{ nm} \times 3\text{ nm}$ but with each second $xy$-slice being just a copy of the previous one. Thus the structure is effectively only discretized with a nominal resolution of $6\text{ nm}$ in $z$-direction, as in the real data. The model
representing layer 2 was discretized on a grid with dimension $205 \times 205 \times 205$ voxels, corresponding to a voxel size of $20 \text{ nm} \times 20 \text{ nm} \times 16 \text{ nm}$.

Although the results are qualitatively similar to the ones obtained on isotropic grids, the permeability of layer 2 seems to be overestimated, again with all confidence intervals overlapping reference values. For layer 1, the permeabilities are underestimated even more strongly than on isotropic grids.

4. Conclusions

Reconstruction of highly porous structures from FIB-SEM image stacks involves a number of steps that bear high danger of distorting the structure. Care has to be taken. In particular, visual validation should always involve views from the non-slicing planes.

If the final goal is simulation of macroscopic materials properties, then the additional experimental effort for imaging really representative volumes should be undergone.

Interpolation onto isotropic grids eases many processing and analysis methods or renders them properly applicable at all. A more detailed study, based on simulated data is needed in order to derive recommendations on when interpolation is beneficial and which interpolation method is preferable. Finally, this study could also be used to quantify which effect contributes how much to the overall error.

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Figure 4: Slices from the FIB-SEM images of layers 1 and 2, pixel edge lengths 3 nm and 20 nm, respectively.
Figure 5: Slices from the FIB-SEM images of layer 1 illustrating the reconstruction of the solid structure.
Figure 6: Slices from the FIB-SEM images of layer 2 illustrating the reconstruction of the solid structure.
Figure 7: Volume renderings of sub-volumes of the structures reconstructed from FIB-SEM images. Visualized are $360 \times 360 \times 190$ voxels corresponding to a volume of $2.97 \, \mu m \times 2.97 \, \mu m \times 1.14 \, \mu m$ for layer 1 and $205 \times 205 \times 165$ voxels corresponding to a volume of $4.1 \, \mu m \times 4.1 \, \mu m \times 3.3 \, \mu m$ for layer 2, respectively.
Figure 8: Slices from the SRµCT images of layer 3, voxel edge length 160 nm, thus the diameter of 2,560 voxels corresponds to 410 µm.

Figure 9: Slices from the combined SRµCT images of layer 3.
Figure 10: Rendered sub-volume of 92 $\mu m^3$ of layer 3.

Figure 11: Slices from realizations of the used Boolean models.