Joachim Ohser[a], Claudio Ferrero[b], Oliver Wirjadi[c],

Alina Kuznetsova[c], Jochen Düll[a], Alexander Rack[b]

[a] University of Applied Sciences, Dept. Math. & Natural Sci., Darmstadt, Germany

[b] European Synchrotron Radiation Facility, Grenoble, France

[c] Fraunhofer Institute of Industrial Mathematics, Kaiserslautern, Germany

Estimation of the probability of finite percolation in porous microstructures from tomographic images

Percolation is an important property of porous media, as it describes the connectivity of pores. We propose a novel, direction-dependent percolation probability which can efficiently be estimated from three-dimensional images obtained by microtomography. Furthermore, in order to describe the penetrability of the pore space by particles of a given diameter or a fluid of a given surface tension, we introduce a percolation probability depending on the width of the pores, from which we may also derive a measure of the mean pore channel width. As application examples, we consider the penetrability of porous berillium pebbles, the connectivity of pores in arctic firn, the percolation of the pore space of aluminium foams and the mean width of the percolating space between the fibers in a laminate's percolating pore space.

Keywords: Connectivity of pore space; Helium cooled pebble bed; Arctic firn; Aluminium foam; Fibre reinforced materials

1 Introduction

The topic of percolation theory is the investigation of the connectivity behavior of random systems such as random graphs, random particle systems, random sets, etc. A representative question in materials science is that for the continuum percolation of a constituent of a macroscopically homogeneous material. Let us consider for example a porous material with a (ductile) solid constituent. Obviously, the solid matter must be totally connected, i. e. all volume elements of the solid are connected by paths inside the solid and, thus, the percolation probability is one. In contrast to this, the geometric properties of pore spaces can vary from low percolation (closed porosity, isolated pores) to high percolation (open porosity, where all pores are interconnected).

Assume a thick section of the pore space of a material which can be seen as the pore space between two parallel planes of fixed distance s. Then the problem of percolating pores can be formulated as follows: The probability P_0 that a randomly chosen point of the pore space in the first plane is connected with a point of the pore space belonging to the opposite plane depends on the the distribution of the pore space, on the distance s as well as on the normal direction θ of the planes. This directed percolation probability P_0 describes the behavior of a wall consisting of a porous material. Will a gas or a liquid be able to penetrate this wall? What is the volume fraction of pores connected with the top as well as the bottom face of the slice? Notice that the percolation probability P_0 is exactly what is meant e. g. by the in-depth fuel migration into the pore space of the carbon reinforced carbon fiber (CFC) materials considered in [24].

Clearly, in physics the capability of being penetrable is usually described by the permeability of the material with respect to a specific matter (gas or liquid). In contrast to this, the percolation probability is a pure geometric characteristic independent of the matter properties. It involves information on the existence of closed porosity not explicitly contained in the permeability. A porous material with a high permeability can have a low percolation probability, and vice verse. Consider for example a 'totally' percolating pore space, where the pores are connected with their neighbors by small necks. Then the permeability decreases with decreasing widths of the necks. Thus, percolation probability and permeability supplement each other in the characterization of porous materials, see also [1]. On the other hand, the computation of percolation probability from micro-tomographic (μCT) data is much faster than that of permeability even if one takes into consideration that there are approaches for rapid estimation of permeability using e. g. Brownian motion paths [8] or a modified random trajectory approach [21, 22]. Therefore, estimating the percolation probability from μCT images appears as an efficient experimental tool for pore space characterization.

In the literature one can find various approaches to continuous percolation. One of the most popular characteristic describing aspects of percolation is Torquato's two-point cluster function C_2, see [25, Section 9.22] and references therein. The function $C_2(x, y)$ is the probability that two points x and y belong to the same connected component. Notice that in case of macroscopic homogeneity, the function C_2 depends only on the Euclidean distance $s = \|y - x\|$ of the points x and y and the direction $\theta = (y - x)/\|y - x\|$, i. e. $C_2(x, y) = C_2(0, y - x)$ with $y - x = (s, \theta)$. Clearly, in case of total percolation, C_2 is the equivalent to the covariance function of the constituent itself. In general, there is no way to compute C_2 from P_0 and vice verse and, thus, both quantities carry different information on percolation. For a sound introduction into percolation theory and its application on flow in porous media see also [7].

We also refer to Hilfer's approach which is originally based on the concept of local porosity, see e. g. [6, 4]. In this approach a cube of edge length a is used as a test window, where the window is chosen as the smallest cube hitting the pore space as well as the solid matter with probability 1. Hilfer considers the percolation of points belonging to opposite cube faces, and he determines the mean local porosity given that there exist points in opposite cube faces which are connected by a path inside the sample observed through this cube. For fixed a the local percolation probability is seen as a function of the local porosity. Hilfer's local percolation was successfully applied to the characterization of porous sandstone.

Our aim is to define continuous percolation in porous media in such a way that it fits a specific question of interest. We consider the in-depth migration of a gas or a liquid into the pores and we ask for the probability that the length of the percolation with respect to a direction is larger than a given threshold. Furthermore, on the one hand, percolation of multiply connected pores is a long-range property and, on the other hand, percolation probability must be estimated from finite samples. Thus, careful handling of edge effects is of huge importance, in particular when most of the pores intersect the frame of the image. Since the data obtained by μCT are given on point lattices, connectedness of pores depend on the assumed adjacency of lattice points. The relationship between continuous percolation in the pore space and discrete percolation of corresponding image data is studied in detail. Finally, in many applications we are interested in the width of the paths through the pore space. Thus, we study the percolation probability depending on the erosion of the pore space with balls of varying radii.

The percolation probability is investigated for the pores in a beryllium pebble, the pore space of an arctic firn, the pores in closed aluminium foams, and the space between the fiber system in a fiber reinforced material.

2 Experimental

As an application we consider a porous beryllium pebble. Beryllium is used as neutron multiplier in the Helium Cooled Pebble Bed (HCPB) blankets investigated in the frame of the European Fusion Technology Programme. A key issue is the behavior of beryllium under the irradiation of neutrons coming from the plasma chamber. The knowledge of long-term tritium and helium accumulation in HCPB blankets is crucial for the reliable and safe operation of fusion reactors. A combination of microstructural analysis and gas release measurements can support the evidence that at high temperatures the tritium inventory is concentrated either in helium bubbles or trapped strain fields in the bubbles' vicinity. In order to investigate the microstructure of the HCPB blankets, μCT was applied which has been proven useful for non-destructive testing of the packing factor of beryllium beds resp. porosity networks characterization, see [11, 14].

The sample is a 2 mm diameter beryllium pebble with grain sizes comprised between 40 and 150 μm which had been neutron irradiated at 770 K, thus producing through a $(n, 2n)$-reaction 480 appm Helium and 12 appm Tritium in the beryllium matrix, and subsequently annealed at 1 500 K, see [11]. This gas concentration is responsible for the presence of the pore network visible in Figure 1a). The microstructure after cooling down the material remains essentially unchanged, since the formation and growth of gas bubbles and percolation pathways during the heating is irreversible.

The μCT measurements were performed at the European Synchrotron Radiation Facility (ESRF) in Grenoble using a monochromatic X-ray beam of 7 keV. The experimental setup is schematically depicted in Figure 2. It consists of a 2D detector system with magnifying lenses and a 2048×2048 pixel CCD camera. The spatial resolution of the field of view was adjusted by means of the detector's optics to be 1.4 μm. The vertical sample rotation axis was aligned with the central axis of the detector with a precision comprised between 0.1 and 0.5 pixels. During the scan, 900 images were acquired covering a total rotation angle of 180°. The pebble was glued on the bottom of a plexiglas cylindrical sample-holder to avoid possible movement artifacts during the rotation scans (original size of the image $512 \times 512 \times 512$ pixels).

The firn sample in Figure 1b comes from the Vostok region in the Antarctis where there is one of the world's longest ice cores. The core samples extracted close to the top of the underlying subglacial lake (lake Vostok) have been assessed to be as old as 420 000 years, thus providing very important paleoclimatic information about our planet. In particular, it is desirable to measure the amount of air trapped in the ice pores and thereby the evolution of the closed porosity as a function of depth [3].

The firn sample shown here is a cylinder of about 10 mm diameter and 10 mm height and was also probed at the ESRF ID19 beamline by μCT. During the rotational scans it was kept at about $-50\,^\circ$C in a plexiglas cryogenic cell. The experimental setup was the same as for the beryllium pebble but the X-ray energy was $\approx$19 keV and the CCD chip was 1024×1024 pixel, with a spatial resolution of $10\,\mu$m (pixel size).

As outlined in the introduction, highly porous media are of natural interest to apply a percolation analysis. We have chosen metallic foams produced by the powder-manufacturing route as example for this kind of materials: the specimens were prepared as part of a larger study aiming to understand the pore formation in the early stages of the evolution of aluminium foams [16, 17]. For the powder-manufacturing route, blowing agents inside a solid precursor are employed to trigger the foaming process when the specimen is heated up to a temperature above the melting point of the foaming material: gas is set free which forms the pores. In order to control the achieved pore structure in a later stage of the development it is desirable to understand if gas exchange paths between the pores do exist [2]. The tomographic images were acquired at the BAM*line* of the BESSY-II synchrotron light source (Helmholtz-Zentrum Berlin, Germany) [18]. The experimental setup is similar as described above for the beamline ID19 of the ESRF. A slightly lower spatial resolution of $12\,\mu$m was used ($3.6\,\mu$m effective pixel) to investigate larger samples. Depending on the individual sample size, X-ray photon energies between 20 keV and 25 keV were employed, 900 projection images were acquired during a 180° scan.

As a further application we investigate the highly percolating space between a strongly anisotropic system of fibers in fiber-reinforced material, where the fiber system appears as a woven textile, see Figure 1c. Such fiber-reinforced materials are an important class of light-weight construction materials used e. g. in automotive applications. One way to produce such materials is by injection of resin into woven textiles. The injection process is rather complex and the resin can spread anisotropically, depending on the geometry of the textile's pore space. One application of the percolation probability proposed in the present paper is to characterize the pore spaces with the ultimate goal of a gaining deeper insight into the infiltration processes during production of fiber laminates. In particular, we consider an anisotropic glass fiber twill weave textile (390 g/m2) injected by epoxy resin (K 506). The sample was injected by epoxy resin in an resin transfer mold to a final fiber volume density of about 40 %. A specimen was scanned using at 100 kV using a Phoenix μCT system. Further details regarding the samples can be found in [20]. The spacing was 2.5 μm, but the data were subsampled to an effictive resolution of 5 μm prior to analysis. An overview on the three-dimensional (3D) images is given in Table 1.

3 Connected components

In order to underline the broad application potential of our approach, different porous materials were studied based on μCT images. This section briefly introduces the parameters of the data acquisition and the scientific context related to the specimen. In this section, we consider the relationship between continuous percolation of pore space and discrete percolation of the image data. First, we have a look on the continuous case and define connectedness of a set in Euclidean space $\mathbb{R}^3$. A path in $\mathbb{R}^3$ is a continuous mapping $f : [0, 1] \mapsto \mathbb{R}^3$. If $f(0) = x$ and $f(1) = y$ then f is called a path from x to y for $x, y \in \mathbb{R}^3$. Then a non-empty set $X \subset \mathbb{R}^3$ is called path-connected if for every $x, y \in X$

there exists a path $f : [0,1] \mapsto \mathbb{R}^3$ from x to y such that $f([0,1]) \subseteq X$. We write $x \sim y$ for path-connected points $x, y \in \mathbb{R}^3$. The equivalence classes $X_1, X_2, \ldots$ of X under $\sim$ are called the path-connected components of X.

Let $\mathbb{L}^3$ be a homogeneous point lattice in $\mathbb{R}^3$, i. a. $\mathbb{L}^3$ is invariant with respect to lattice translations, $\mathbb{L}^3 = \mathbb{L}^3 + x$ for all $x \in \mathbb{L}^3$. Discrete percolation of a subset of the lattice points $\mathbb{L}^3$ is closely related to adjacency of lattice points. We follow the approach of [12, 13] and define adjacency on $\mathbb{L}^3$ as follows: Let be chosen a lattice cell C of $\mathbb{L}^3$ hitting the origin (called the unit cell). We consider collections (i. e. subsets) of vertices of C and their convex hulls. Then a local adjacency system $\mathbb{F}_{\mathrm{loc}}$ is defined as a subset of the set of the convex hulls of all collections of vertices of C. Such a local adjacency system must fulfill certain properties addressed in [12]. Then a (global) adjacency system $\mathbb{F}$ is definded as the union of all lattice translations of $\mathbb{F}_{\mathrm{loc}}$, that is $\mathbb{F} = \bigcup_{x \in \mathbb{L}^3} (\mathbb{F}_{\mathrm{loc}} + x)$. The system of edges and vertices contained in $\mathbb{F}$ forms the neighborhood graph and the order of the nodes of this graph is said to be the adjacency of $\mathbb{F}$. Of practical importance are the adjacency systems $\mathbb{F}_6$, $\mathbb{F}_{14.1}$, $\mathbb{F}_{14.2}$ and $\mathbb{F}_{26}$ defining the 6-adjacency (where each pixel is connected with 6 neighbors), two 14-adjacencies and the 26-adjacency, respectively. It is well-known that the 6-adjacency is complementary to the 26-adjaceny, i. e. if one chooses the 6-adjacency for the foreground pixels of a binary image, then the background is connected with respect to a 26-adjacency, and vice verse. The two 14-adjacency systems are self-complementary [12].

A discrete path connecting the lattice points $x, y \in \mathbb{L}^3$ with respect to the adjacency system $\mathbb{F}$ – in the following called an $\mathbb{F}$-path – is a sequence of lattice points $(x_i)_{i=0}^m \subset \mathbb{L}^3$,

$m \in \mathbb{N}$, with $x_0 = x$, $x_m = y$ and the edges $[x_{i-1}, x_i] \in \mathbb{F}$, $i = 1, \ldots, m$. A nonempty discrete set $Y \subseteq \mathbb{L}^3$ is called $\mathbb{F}$-connected if Y consists of only one lattice point or for all pairs $(x, y) \in Y^2$ with $x \neq y$ there exists an $\mathbb{F}$-path from x to y. The equivalence classes $Y_1, Y_2, \ldots \subseteq Y$ defined through the $\mathbb{F}$-connectedness are called the connected components of Y under $\mathbb{F}$. Clearly, a finite set Y can consist of only a finite number of equivalence classes. We use the notation $Y_{\mathbb{F}} = \{Y_1, \ldots, Y_m\}$ for the set of equivalence classes of a finite set Y under $\mathbb{F}$.

4 Percolation probability

Let γ be a random closed set modeling the pore space of a porous medium. We assume that the porous medium is macroscopically homogeneous. That is, in the language of stochastic geometry, γ forms a stationary random set in $\mathbb{R}^3$. Furthermore, we assume that the realizations of γ almost surely belong to the extended convex ring of $\mathbb{R}^3$ and γ fulfills the condition that the expectation of the random number $2^{\#(\gamma \cap K)}$ is finite for all compact and convex sets $K \subset \mathbb{R}^3$, where $\#X$ is the minimal number m such that the set X has a representation $X = K_1 \cup \ldots \cup K_m$ with compact and convex sets $K_1, \ldots, K_m$.

Then the percolation probability $P_0(s, \theta)$ of γ can be defined as probability that for a point x randomly chosen in γ there is a point y in γ connected with x such that the orthogonal projection of the difference $y - x$ onto the direction θ is larger than s, $(y - x)\theta > s$. Formally, we consider a set $X \subset \mathbb{R}^3$ being a finite union of compact and convex sets and a function f given by

$$f(X,x,\theta,\varrho) = \begin{cases} \sup\limits_{y\in X\cap(B_\varrho+x)} \big\{(y-x)\theta \,:\, x \overset{X}{\sim} y\big\}, & x \in X \\ 0, & x \notin X \end{cases}$$

where $x \overset{X}{\sim} y$ means that there is a path from x to y in X and $B_\varrho + x$ is the ball of radius ϱ centered in x. Now the percolation probability P_0 of a random set γ can be defined as the limit

$$P_0(s,\theta) = \lim_{\varrho\to\infty} \mathbb{P}(f(\gamma,0,\theta,\varrho) > s \,|\, 0 \in \gamma), \qquad s \geq 0. \tag{1}$$

Obviously, $P_0(s_1,\theta) \geq P_0(s_2,\theta)$ for all $\theta \in S^2$ and $0 \leq s_1 \leq s_2$. Furthermore, the percolation probability P_0 is larger than Torquato's (normalized) two-point cluster function, $P_0(s,\theta) \geq C_2\big(0,(s,\theta)\big)/V_V^2$ for all $s \geq 0$ and for all $\theta \in S^2$, where V_V is the volume fraction of the pores.

Assume now that the realizations of γ are almost surely morphologically regular, i. e. there is an $\epsilon > 0$ such that γ is invariant under morphological closure and morphologial opening with a ball B_ϵ. If there is an $s_0 > 0$ such that $P_0(s_0,\theta) = 0$ for all θ, the pores are not infinitely percolating (closed porosity), otherwise the pore space is infinitely percolating (e. g. in the sense of Torquato [25]). The limit $p = \lim_{s\to\infty} P_0(s,\theta)$ is the volume fraction of infinitely percolating pores with respect to θ. For $p = 1$ the pore space is said to be totally percolating (open porosity).

5 Estimating the percolation probability

The core of estimating P_0 from 3D image data of porous media (i. e. the observation of a realization of γ on a lattice $\mathbb{L}^3$ and through a cuboidal window W) is a labeling algorithm. Very efficient labeling algorithms ate published in [9, 23], see also [13, Section 4.3.2.3] for further details.

Estimates of the percolation probability P_0 obtained from image data depend on the edge of the window W through which the specimens are observed. Let $\gamma_0, \gamma_1, \ldots$ be the equivalence classes of γ with respect to $\sim$. In general the set $\{\gamma_0 \cap W, \gamma_1 \cap W, \ldots\}$ of intersections with a window $W \subset \mathbb{R}^3$ differs from the set of the path-connected components of $\gamma \cap W$. This means that tessellation into path-connected components is influenced by the windowing. With increasing percolation lengths s there is an increasing number of points in $\gamma \cap W$ (more or less close to the edge of W) for which it is not be decidable whether or not the percolation length is larger s. These points must be excluded from the estimation procedure by reducing the window.

In the following, edge correction is described for the special case of a cuboidal window with edges parallel to the coordinate axes. The direction θ may be parallel to one of the outer normals of the cuboid faces. Then a reduced window $W' \subset W$ is a cuboid with edges parallel to those of W. Throughout this paper the volume of window W' was set to 75 % of the volume of W (i. e. 75 % of the image volume), where W' touches the face of W with the outer mormal θ. This empirical choice is due to our experience based on experiments with various image data.

Let Y be the set of the foreground pixels of a binary image, i. e. Y is the intersection of a realization of γ with $\mathbb{L}^3 \cap W$. Then the algorithm of estimating P_0 is as follows:

1. Computing a label image, i.e. finding the equivalence classes Y_k of Y with respect to an adjacency system $\mathbb{F}$.

2. A distance image is computed where each pixel x is assigned the distance

$$d = \max_{y \in Y} \{(x - y)\theta \, : x \sim y \text{ under } \mathbb{F}\},$$

   for all foreground pixels $x \in Y$, and $d = 0$ otherwise, where $\sim$ denotes the equivalence relation on Y under a given adjacency system $\mathbb{F}$. This is based on determining the maximum point $y_k = \max\{y\theta : y \in Y_k\}$ for each equivalence class Y_k of Y and assigning each pixel x of Y_k to the distance $(x - y_k)\theta$.

3. For a given distance s we consider the eroded window $W'_s = W' \ominus [0, -s\theta]$ where $\ominus$ is the Minkowski subtraction and $[0, -s\theta]$ is the segment between the origin 0 and the point $-s\theta$. Now $P_0(s, \theta)$ is estimated as the number of pixels in W' with percolation lengths larger s over the total number of foreground pixels in W'_s.

Clearly, $P_0(s, \theta)$ can be estimated only for those s for which the number of foreground pixels in W'_s is not zero.

6 Percolation depending on pore width

Now we ask for possibility that particles can penetrate through the pore space. This question is important for designing filters such as dust or soot particle filters for cars. For simplicity we consider spherical particles of radii less than a given parameter r. Then the percolation of those particles through the pore space γ can be characterized by the percolation probability of the reduces pores space $\gamma \ominus B_r$, where $\ominus$ denotes the Minkowski subtraction and B_r is a ball of radius r. Clearly, because of the symmetry of the structuring element B_r, the Minkowski subtraction is equivalent to erosion.

Notice that the percolation probability depending on the pore width r can also used for modeling the penetration of the pore space γ by a glutinous liquid of given surface tension where the critical surface tension is proportional to the curvature $1/r$. The last aspect is of importance e. g. for the exploration of oil included in the pore space of Savonnier-oolithic sandstone, see [6, 4]. Let Δp be capillary pressure difference sustained across the interface between two static fluids, such as oil and water, due to the phenomenon of surface tension. The Young-Laplace equation relates the pressure difference to the mean curvatures $M(x)$ at a surface point x, $\Delta p \sim M(x)$. Taking into consideration the pore width and the liquid-solid surface tension of oil and sand stone, one can estimate the outer pressure difference necessary for penetration of the oil through the pore space of the sand stone.

By $P_r(s, \theta)$ we denote the percolation probability of the reduced pore space γ_r, $r \geq 0$. It can be shown that for fixed $s \geq 0$ and $\theta \in S^2$, the product $P_r(s, \theta) \cdot \mathbb{P}(0 \in \gamma_r)$ is decreasing in r and, thus,

$$F_{s,\theta}(r) = 1 - \frac{P_r(s, \theta)\, \mathbb{P}(0 \in \gamma_r)}{P_0(s, \theta)\, \mathbb{P}(0 \in \gamma)}, \qquad r \geq 0$$

is a probability distribution function (for those s and θ with $P_0(s, \theta)\, \mathbb{P}(0 \in \gamma) > 0$). Obviously, $F_{0,\theta}(r)$ is the spherical contact distribution function of γ. The mean width $\bar{d}_{s,\theta}$

of percolating pore space is twice the expectation,

$$\bar{d}_{s,\theta} = 2\int_0^\infty r dF_{s,\theta}(r), \qquad s \geq 0,\ \theta \in S^2.$$

Differentiation by parts yields

$$\bar{d}_{s,\theta} = 2\int_0^\infty \big(1 - F_{s,\theta}(r)\big)\, dr = \frac{2}{P_0(s,\theta)\, \mathbb{P}(0 \in \gamma)} \int_0^\infty P_r(s,\theta)\, \mathbb{P}(0 \in \gamma_r)\, dr.$$

Given a binary image of γ observed through a window, then the reduced pore space $\gamma \ominus B_r$ can be computed by the Euclidean distance transform (EDT) which maps to each foreground pixel the minimum Euclidean distance to the background. The result is a (real valued) distance image. Then $\gamma \ominus B_r$ can be obtained by a simple thresholding of the distance image, where the parameter r serves as the threshold level. More precisely, the pixels in the distance image with values less than r are forming the foreground pixels of $\gamma \ominus B_r$, where $\gamma \ominus B_r$ is free of edge effects in the reduced window $W \ominus B_r$. An algorithm of the EDT with computation time depending linear on the pixel number is given in [10], see [12, Section 4.2.7.2] for an overview on further algorithmic approaches.

It should be taken in mind that for fixed s and θ the function $F_{s,\theta}$ is the distribution function of a spherical contact distribution. As a consequence, the mean width $d_{s,\theta}$ is twice the expectation with respect to the corresponding spherical contact distribution. Consider for example a random system of infinite cirular cylinders of direction ϕ and fixed diameter ϱ. Then the mean width $d_{s,\theta}$ is smaller than ϱ; one gets $d_{s,\theta} = \frac{2}{3}\varrho$ for all $s \geq 0$ and θ not orthogonal to ϕ. This seems to be in conflict with the interpretation of $d_{s,\theta}$ as the mean width of percolating pores, but one should notice that $d_{s,\theta}$ is the mean diameter of maximum balls not hitting the solid matter where the balls centers are uniformly distributed in the pore space.

7 Results

The percolation probability $P_r(s,\theta)$ and the mean width $\bar{d}_{s,\theta}$ of the percolating pore spaces were computed basing on the `C++` library `MAVIlib` created at [5] designed for the analysis of μCT images of materials structures. The pore spaces were labeled with respect to the self-complementary 14.1-adjacency.

For the beryllium pebble the percolation probability P_0 decreases rapidly, see Figure. 3. Nevertheless, there is an considerable fraction of the pore space (about 10 %) with percolation length larger than 1 mm. Furthermore, the percolation is anisotropic; the volume fractions of the percolating pore space with $s \geq 1\,\text{mm}$ are 12.8 % (for the x-direction), 7.0 % (for the y-direction) and 2.5 % (for the z-direction), respectively. For example, this means that only 12.8 % of pore space at distance 1 mm from the pebble's surface element (with outer normal direction parallel to the x-axis) can accumulate tritium and helium. Under certain conditions (isotropic percolation, equilibrium state of the gas absorption process, s much smaller than the mean width of pebble), the concentration profiles of tritium and helium in a pebble are proportional to $P_0(s,\theta)$ where s is the distance from the surface.

The results confirm the qualitative findings of [15]. Beryllium is an anisotropic material both as a single crystal and as polycrystal, the latter being the case of the sample used for the present investigation which was produced by fluoride reduction process. Although one of the main aims of beryllium powder metallurgy is achieving as nearly as possible an isotropic orientation of beryllium grains, some texture, i. e. alignment of different grains is typically produced during manufacturing (extrusion or machining) and this leads to larger percolation paths where the basal plane layers are. This is the case of the axial planes (i. e. the panes perpendicular to the z-direction) of the reconstructed pebble volume, as shown by Fig. 1a. Since the powder particles, which are generally platelet shaped, are produced by basal plane cleavage, they tend to align in the pressing dies in a preferred slip direction which is transversal to the compression axis.

The firn sample is isotropic, and in particular the percolation probability $P_r(s,\theta)$ of its pore space is independent of the direction θ. As a consequence we can take the average over the x-, y- and z-direction. Obviously, the pore space is totally percolating (open porosity) with lengths at least 6 mm, i. e. $P_0(s,\theta) = 1$ for $0 \leq r \leq 6$ mm, see Fig. 4. Moreover, also the reduced pore space is totally percolating, $P_r(s,\theta) = 1$ for $0 \leq r \leq 6$ mm and $r \leq 0.1$ mm. (When reducing the pores space by erosion with balls of radii at least 0.2 mm, the pore space gets closed.) This means that the pores are interconnected by channals of widths at least 0.2 mm. As a consequence, extremely long-range gas exchange can happen which leads to difficulties in precise dating of the paleoclimatic information by local probing. The paleoclimatic information involved in the air trapped in the firn measured at a depth is a convolution of the true air composition (as a function of the depth assigned to a paleonthologic period) and the percolation probability P_0.

The results of the analysis on the tomographic images of metallic foams in the early stages of the expansion process are depicted in Fig. 5. Different characteristics can be identified: the two plots on the right side (spec. no. 4 and 6) belong to foams consisting mainly of small and isolated pores with a rather isotropic shape. Corresponding to that, the percolation probability drops fast due to the lack of interconnection between the individual voids. The plot in the upper left (spec. no. 3) belongs to a foam with larger pores but still isotropic shape. The percolation probability is significantly enlarged with respect to the samples belonging to the plots on the right side. Finally, the lower left (spec. no. 5) shows the result for a foam with anisotropic pores (crack-like, cf. Fig. 1b).

From the plots, one can derive that the foaming process in the studied aluminium foams starts with small isolated pores without any gas exchange. Once the pore space grows due to the continued gas release by the blowing agent, pore coalescence starts, i. e. individual pores start to be connected with each other, gas exchange paths establish, leading to a merger of pore space as larger pores attend to include smaller ones. In case of an early gas release before the precursor material has reached its melting temperature, cracks are known to appear perpendicular to the direction of compression applied for manufacturing. The two-dimensional shape of the cracks leads to an enlarged percolation probability in the corresponding two direction in space.

The mean width $\bar{d}_{d,\theta}$ of the pore space in the glass fibre laminate data is shown in Fig. 6. Clearly, the percolation probability is lower in x-direction, which corresponds to the injection direction during infiltration of the dry textile with resin. The mean pore width in this direction tends to about $48\,\mu$m with increasing percolation length s. In the y- and z-directions (the so-called in-plane directions) the mean pore widths are larger than in x-direction, but not equal, and no convergence can be observed at the investigated lengths s.

Firstly, we observe that these results are in line with earlier, experimental measurements of the same material reported in [19], where the flow front during injection was observed to be anisotropic (in the plane orthogonal to the injection direction, i.e., in the yz-plane). This could be a consequence of difference in mean pore width of about $3\,\mu$m between the y- and z-directions, cf. Fig. 6.

Secondly, phenomenological laws such as Darcy's laws and the Hagen-Poiseuille equation predict that permeability is proportional to pore diameter. Thus, we would expect a low permeability for this material in x-direction (injection direction) and a higher – but not equal – permeability for the two remaining axis directions. This has indeed also been experimentally shown for this material, see [20].

8 Discussion

As the percolation probability P_0, Torquato's two-point cluster function C_2 can be estimated from a label image. The algorithm works as follows: Compute the the volumes and the auto-correlation functions of all connected components Y_k. Then C_2 can be estimated by the volume-weighted mean of the auto-correlation functions, see Fig. 7 for an example. In order to speed-up the algorithm, the auto-correlation functions can be computed via frequency space. Nevertheless, in case of a large number of connected components, the computation time for C_2 is much higher than for P_0. Notice that characterization of percolation based on C_2 can pretend closed porosity. Assume for example that the pore space is formed by a system of infinitely long and straight channals (cylinders) with directions neither parallel nor orthogonal to θ. Then $C_2(s,\theta)$ is zero just for small s, but the pore space is totally percolating with respect to the θ. The pore space of the pebble is an impressive practical example where C_2/V_V^2 is much smaller than P_0, see Fig. 7.

Estimates of the percolation probability P_r can highly depend on the adjacency system $\mathbb{F}$ chosen for the labeling of the pore space, see e. g. Fig. 7. (The same is true also for Torquato's two-point cluster function C_2 and Hilfer's local percolation probability.) Only in cases when the pore space is morphologically regular with respect to the edges, face diagonals and space diagonals of the unit cell C of the underlying lattice $\mathbb{L}^3$, the percolation probability P_r is independent of $\mathbb{F}$. Until now there is no responsible and practicable criterium for an appropriate choice of $\mathbb{F}$. As a consequence, one should specify the adjacency system $\mathbb{F}$ used for the estimation of P_r.

The crucial point is the joice of the reduced window $W' \subset W$. Let W and W' are cuboidal windows, where edges of W' parallel to those of W. The direction θ may the outer normal of a face F of W. In order to estimate P_0 free of edge effects, W' must be chosen such that it touches F and each connected component Y_k not hitting F but intersecting W' is completely inside W, i. e. Y_k must belong to the interior of W (denoted by $\mathrm{int}\, W$). This means, W' must have the property that

$$\text{if} \quad Y_k \cap F = \emptyset \quad \text{and} \quad Y_k \cap W' \neq \emptyset \quad \text{then} \quad Y_k \subset \mathrm{int}\, W, \quad k = 1, \ldots, m.$$

Clearly, the size of W' depends on the percolation length. For closed porosity as well as for mostly open porosity the size of W' is not much smaller than W. Nevertheless, for transition states between closed and open porosity, W' can be very small which leads to large statistical errors of estimates of P_0. In this case one must find a balance between a small statistical error (obtained for large W') and a small systematic error (for small

W'). Most estimates of P_r presented in this article are biased. Only in particular cases, e. g. in case of the firn sample for $r < 0.2$ mm (open porosity) and for $r > 0.6$ mm (closed porosity), the estimates are free of edge effects.

9 Conclusions

The percolation probability P_r has proven very useful for the characterization of various aspects of porous materials. It can be measured efficiently from μCT images using fast algorithms for the labeling and for the EDT. Edge effects resulting from the boundedness of the images can be reduced by a specific procedure.

In contrast to Hilfer's approach, the percolation probability P_r can be estimated also for large percolation length s, where s is in the range of the edge lengths of the image. The mean width $\bar{d}_{s,\theta}$ of percolating pores, seen as a function of the percolation length s and the direction θ, can be used to explain certain phenomena of injection processes in fibre laminates.

This work was supported by the German Ministry of Education and Research (BMBF) under grand 03MS603. The authors are grateful to R. A. Pieritz for providing the μCT data of beryllium pebble. We are grateful also to Prof. Mitschang's group at the Institute for Composites (IVW), Kaiserslautern, for providing the glass fiber laminate data, and for many fruitful discussions.

Correspondence address:

Prof. Joachim Ohser

University of Applied Sciences Darmstadt, Dept. of Mathematics and Natural Sciences, Schöffer-Straße 3, D-64295 Darmstadt, Germany

Tel.: ++49-(06151)168659

Fax.: ++49-(06151)168975

e-mail: jo@h-da.de

Table 1: Survey of the 3D images investigated in this article.

no	specimen	number of pixels	pixel size
1	beryllium pebble	$327 \times 554 \times 292$	$4.9\,\mu$m
2	arctic firn	$724 \times 724 \times 1\,024$	$10.0\,\mu$m
3	aluminium foams	$395 \times 480 \times 262$	$7.2\,\mu$m
4		$490 \times 495 \times 350$	$7.2\,\mu$m
5		$350 \times 400 \times 350$	$7.2\,\mu$m
6		$440 \times 500 \times 380$	$7.2\,\mu$m
7	fiber laminate	$400 \times 750 \times 750$	$5.0\,\mu$m

Figure 1: Visualizations of the image data; a) porous beryllium pebble where the pores are shown in dark blue, b) an aluminium foam in an early state of the foaming process, c) the fiber system of a fiber reinforced material, d) a firn sample from the Vostok region in the Antarctis.

Figure 2: Schematic view of the μCT setup at the ESRF ID19 beamline [26].

Figure 3: The percolation probability $P_0(s, \theta)$ for the pores of the beryllium pebble for the three space directions.

Figure 4: The percolation probability $P_r(s, \theta)$ for the pores of the arctic firn depending on s (in a logarithmic scale) and r, where the mean is taken over the three space directions.

Figure 5: The percolation probability $P_0(s, \theta)$ for the pores of the foam samples (in a logarithmic scale).

Figure 6: The mean beadth $\bar{d}_{s,\theta}$ of the percolating pores of the fiber reinforced material (in a logarithmic scale).

Figure 7: Comparison of the percolation probability P_0 and Torquato's two-point cluster function $C_2(s, \theta)$ for the pore space of the beryllium pebble, where θ is the x-direction. For comparison reasons the normalized version C_2/V_V^2 is plotted.

References

[1] Andersson, L., Jones, A. C., Knackstedt, M. A. and Bergström, L. (2011). Permeability, pore connectivity and critical pore throat control of expandable polymeric sphere templated macroporous alumina. *Acta Materialica* **59,** 1239–1248.

[2] Banhart, J. (2001). Manufacture, characterisation and application of cellular metals and metal foams. *Progress in Materials Science* **46,** 559–632.

[3] Barnola, J.-M., Pieritz, R. A., Goujon, C., P., D. and E., B. (2004). 3D reconstrucion of the Vostok firn structure by X-ray tomography. In *Proc. XIII Glaciological Symposium.* St.Petersburg, Russia.

[4] Biswal, B., Manwart, C. and Hilfer, R. (1998). Three-dimensional local porosity analysis of porous media. *Physika* **A 255,** 221–241.

[5] Fraunhofer ITWM, Department of Image Processing. MAVIlib – modular algorithms for volume images, C++ library. http://www.mavi-3d.de 2011.

[6] Hilfer, R., Rage, T. and Virgin, B. (1997). Local percolation probability for a natural sandstone. *Physica* **A 241,** 105--110.

[7] Hunt, A. (2005). *Percolation Theory for Flow in Porous Media.* Lecture Notes in Physics. Springer, New-York.

[8] Hwang, C.-O., James, A. G. and Mascagni, M. (2000). On the rapid estimation of permeability for porous media using Brownian motion paths. *Phys. Fluids* **12,** 1699--1709.

[9] Martín-Herrero, M. (2007). Hybrid object labelling in digital images. *Machine Vision Appl.* **18,** 1--15.

[10] Maurer, C. R. and Raghavan, R. (2003). A linear time algorithm for computing exact Euclidean distance transforms of binary images in arbitrary dimensions. *IEEE Trans. Pattern Analysis and Machine Intelligence* **25,** 265--270.

[11] Moeslang, A., Pieritz, R. A., Boller, B. and Ferrero, C. (2009). Gas bubble network formation in irradiated beryllium pebbles monitored by X-ray microtomography. *J. Nucl. Mat.* **386–388,** 1052--1055.

[12] Ohser, J., Nagel, W. and Schladitz, K. (2009). Miles formulae for Boolean models observed on lattices. *Image Anal. Stereol.* **28,** 77--92.

[13] Ohser, J. and Schladitz, K. (2009). *3D Images of Materials Structures -- Processing and Analysis*. Wiley VCH, Weinheim, Berlin.

[14] Pieritz, R. A., Reimann, J. and Ferrero, C. (2011). 3D tomography analysis of the inner structure of pebbles and pebble beds. *Adv. Eng.. Mat.* **13,** 145--155.

[15] Rabaglino, E., Baruchel, J., Boller, E., Elmoutaouakkil, A., Ferrero, C., Ronchi, C. and Wiss, T. (2003). Study by microtomography of 3D porosity networks in irradiated beryllium. *Nucl. Instr. Meth. in Phys. Res. B* **200,** 352--357.

[16] Rack, A., Helfen, L., Baumbach, T., Kirste, S., Banhart, J., Schladitz, K. and Ohser, J. (2008). Analysis of spatial cross-correlations in multi-constituent volume data. *J. Microscopy* **232,** 282--292.

[17] Rack, A., Helwig, H.-M., Bütow, A., Rueda, A., Matijašević-Lux, B., Helfen, L., Goebbels, J. and Banhart, J. (2009). Early pore formation in aluminium foams studied by synchrotron-based microtomography and 3d image analysis. *Acta Mater.* **57,** 4809--4821.

[18] Rack, A., Zabler, S., Müller, B. R., Riesemeier, H., Weidemann, G., Lange, A., Goebbels, J., Hentschel, M. and Görner, W. (2008). High resolution synchrotron-based radiography and tomography using hard X-rays at the BAMline (BESSY II). *Nucl. Instr. & Meth. in Phys. Res. A* **586,** 327--344.

[19] Rieber, G. and Mitschang, P. (2010). 2D permeability changes due to stitching seams. *Composites Part A: Applied Science and Manufacturing* **41,** 2 -- 7.

[20] Rieber, G., Wirjadi, W. and Mitschang, P. (2010). Correlation of permeability values with flow channel diameters determined by 3D-image analysis of a woven textile. In *Proc. 10th International Conference on Flow Processes in Composite Materials (FPCM10).* Monte Veritá, Ascona, CH. pp. 23--29.

[21] Romm, F. (2002). Evaluation of permeability of micropotous medua using the modified random-trajectory approach. *J. Colloid Interf. Sci.* **250,** 191--195.

[22] Romm, F. (2002). Theories and theoretical models for percolation and permeability in multiphase systems: comparative analysis. *Adv. Colloid Interf. Sci.* **99,** 1--11.

[23] Sandfort, K. and Ohser, J. (2009). Labeling of n-dimensional images with choosable adjacency of the pixels. *Image Anal. Stereol.* **28,** 45--61.

[24] Tiseanu, I., Tsitrone, E., Kreter, A., Craciunescu, T., Loarer, T., Pégourié, B. and Dittmar, T. (2011). X-ray micro-tomography studies on carbon based composite materials for porosity network characterization. *Fusion Eng. Design* submitted.

[25] Torquato, S. (2002). *Random Heterogeneous Materials: Microstructure and Macroscopic Properties.* Springer, New York.

[26] Weitkamp, T., Tafforeau, P., Boller, E., Cloetens, P., Valade, J.-P., Bernard, P., Peyrin, F., Ludwig, W., Helfen, L. and Baruchel, J. (2010). Status and evolution of the esrf beamline id19 ed. M. Denecke and C. T. Walker. vol. 1221 of *AIP Conf. Proc. (ICXOM20).* pp. 33--38.