Topology of evolving pore networks

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Abstract. Morphological and topological quantification of complex pore networks is of great relevance for environmental engineering, earth science and industry. Recent developments of 3D imaging techniques such as X-ray microtomography or X-ray microscopy provide an opportunity to perform a comprehensive analysis of the pore network topology. Such an analysis is crucial to understand how transport or mechanical properties evolve during the growth and/or the aging of a pore network, especially near a percolation threshold. In the first part of this work, we present some properties related to the graph of retraction of a 3D pore network, a powerful way to characterize the topological evolution. In the second part, we analyze the topology of an evolving 3D pore network in the vicinity of a percolation transition. Two distinct *scenarii* of evolution are presented. The last part is dedicated to an experimental example of evolving pore network: the setting of an ordinary cement paste probed in its early age by synchrotron X-ray microcomputerized tomography.

1 Introduction

All around us, pore networks are ubiquitous and play a major role in fundamental processes occurring on the Earth's surface as well as in several industrial processes (civil engineering, oil industry, drilling fluids, food industry, cosmetic industry, etc.). Most of these porous media are characterized by a complex structure. In many instances, these materials evolve in time [1]. This is the case, for example, during grain consolidation and cementation processes of sandstones and limestones, pore network alteration by dissolution and reprecipitation, remodeling mechanism of biological tissues such as bone. The pore network geometry can also be modified by fluid invasion during partial imbibition and drainage [2]. All these evolutions strongly modified the long-range connection properties involved in transport processes and mechanical properties. An emblematic situation is the appearance of a percolation threshold [3] which is often associated to a strong hindrance of the transport and the mechanical ruin. There is an actual need to characterize the geometry of these evolving pore networks in order to provide reliable modelings able to assure the sustainability of these materials. Recent developments of 3D imaging techniques including transmission electron microscopy (TEM) [4], hard X-ray microtomography [5] and soft X-ray nanotomography [6] provide the opportunity

to investigate and image the 3D structure of numerous porous materials in evolution at different length-scales ranging from few nanometers to several millimeters. The characterization of these images can be performed at three different levels of complexity. At the first level, apparently the most simple, the aim is to obtain a few numbers, which characterize the global properties of the porous material. One can first ask how much space the pore network occupies and determine the porosity Φ , defined as the ratio of void (or pore) volume over total sample volume. A subtler question deals with the overall amount of interface per unit of volume S_v . However, Φ and S_v do not provide any clear information about the morphology of the pore network [7]. One has to reach a second level of analysis where questions are raised about average pore size, mean curvature, pore shape, surface roughness, structural correlation between points belonging to the solid, the interface or the pore network. The last level concerns a topological analysis of the matrix [8,9], which is closely related to the long-range connectivity of pore network. Several interesting properties should be analyzed at this level such as the number of available paths linking two distinct points of the pore network, the metric distance between two points compared to their shortest (geodesic) distance and the important role of pore throats. It is clear that several terms are ill defined or ambiguous. This is, for example, the case for the “average” pore shape of a disordered pore network. It becomes evident that porous media must be

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characterized in statistical terms [10]. In this work, we study the topology of some evolving pore networks especially close to a percolation threshold. The paper is organized as follows. In the first part, we present some properties related to the graph of retraction [11] of a 3D pore network, a powerful way to characterize the topological evolution. In the second part, we analyze the topology of an evolving 3D pore network in the vicinity of a percolation transition. Two distinct *scenarii* of evolution are presented. The last part is dedicated to an experimental example of evolving pore network: the setting of an ordinary cement paste in its early age.

2 Topology and skeleton graph of a pore network

The topological properties of a pore network quantify its connectivity. Remember that a geometrical set X is interconnected when every pair of points belonging to X may be joined by a curve included in X [12]. These topological properties are invariant under a continuous deformation of the matrix. The pore network is essentially defined by its internal interface; the topology, mainly the connectivity, is related to the genus G of this interface [8]. The genus of a surface is the maximum number of ways a surface may be cut by closed loops without losing its initial connectivity. $G = 0$ for a sphere and $G = 1$ for a torus. The genus is not additive and that is why a second parameter, the Euler-Poincaré constant N_3 , is generally used to the characterize the pore network topology [8,9,12]:

$$N_3 = \sum_{i=1}^{i=\max} (1 - G_i), \quad (1)$$

where $i \max$ is the number of disconnected surfaces delimiting the pore network. N_3 is additive and is directly related to average Gaussian curvature of the internal interface. In the case of a collection of disconnected convex particles, N_3 is exactly the number of objects. For a pore network delimited by a unique multiconnected interface having a genus G , N_3 is equal to $1 - G$ and is strongly negative. N_3 does not fully characterize the topology of a geometrical set. Its value depends on three numbers, the Betti numbers [8], respectively, β_0 , the number of connected parts, β_1 , the number of irreducible paths and β_2 , the number of irreducible internal surfaces. An irreducible path is a close loop inside the considered set which cannot be reduced to a simple point under a continuous deformation. For example in the case of a solid torus, there is one type of irreducible path: a close loop around and inside the torus. We have the general equality in 3D:

$$N_3 = \beta_0 - \beta_1 + \beta_2. \quad (2)$$

A third way to compute N_3 is to perform a complete tilling of the considered manifold with a network made of α_3 adjacent elementary cells generating α_0 vertices, α_1 edges, α_2 surfaces. Then, the following equality holds:

$$N_3 = \alpha_0 - \alpha_1 + \alpha_2 - \alpha_3. \quad (3)$$

Let now consider some examples to illustrate the use of the three former equations. Following equation (1), a solid sphere has only one surface and then $N_3 = 1$. A collection of P disjointing solid spheres is characterized by $N_3 = P$. Using equation (2), a 3D solid cellular foam made of P close and empty cells has the following Betti numbers $\beta_0 = 1$ (one set), $\beta_1 = 0$ (no irreducible path), $\beta_2 = P$ (P irreducible internal surfaces) and $N_3 = P + 1$. In the case of a foam made of a packing of polyhedra with opened surfaces only delimited by a set of α_0 vertices and α_1 links, $N_3 = \alpha_0 - \alpha_1$. Interesting enough if this opened foam is made of α_3 polyhedra with a total of α_2 surface, we get the equality $\alpha_0 - \alpha_1 + \alpha_2 - \alpha_3 = 1$ thanks to the fact that the tilling made of these polyhedra generated a uniform and compact volume homeomorphic to a solid sphere. In the case of a pore network where there is no isolated “solid grain” floating in the pore, $\beta_2 = 0$. On the contrary, several close pores can be trapped inside the solid part of a porous material. Then, $\beta_2 \geq 0$.

In order to get a normalized topological estimator of the pore network, it is usual to use the ratio N_3/V , where V is the volume of the 3D image under analysis. Such a computation can be directly performed on the binary image thanks to the additivity of N_3 . However, this indicator stays an extensive characteristic. For example, N_3/V is not invariant under an homothetic transformation of the pore network even if the topology is conserved during this transform. It is then difficult to compare the topological evolution of different structures using the N_3/V ratio only.

A more powerful tool to characterize the topology is the skeleton graph SG [8,9,11,13]. SG is defined as the set of centers of all maximal spheres included in the pore network. This skeleton graph is obtained by progressively narrowing the pore space starting from the internal surface. This thinning retraction must satisfy both the invariance of the three first Betti numbers and some morphological constraints in order to get a skeleton graph statistically centered in the pore network. A difficult step is to reduce the numerical skeleton in terms of a simple graph made of vertices connected by edges and having the same topological properties [11,13] as the initial set. In this paper, we use a numerical scheme proposed few years ago by some of us [11]. An example of this skeletonization is shown in Figure 1 where dangling bonds (the so-called termini) are eliminated under a continuous retraction. In the case of the pore network defined by a random close packing of solid spheres, the vertexes are associated to the pores and the edges between two vertexes are passing through the pore throats. After a graph retraction, the Euler-Poincaré constant is now linked to the number of vertexes α_0 (isolated or not) and links α_1 as:

$$N_3 = \alpha_0 - \alpha_1. \quad (4)$$

In order to characterize, in a simple way, the evolution of the 3D skeleton graph, we propose to consider the following connection number [2,9]:

$$C_t = -(\alpha_0 - \alpha_1)/\alpha_0. \quad (5)$$

The definition of C_t allows us to define an *intensive* parameter related to the number of irreducible paths per

Fig. 1. (a) Binary 3D reconstruction of Vycor-like porous glass. The porosity is set to 0.5 and (b) the corresponding skeleton graph.

vertex. C_t is invariant under any continuous deformation of the pore network. For a graph without isolated vertexes, it is straightforward to check that:

$$\langle N_c \rangle = 2(C_t + 1), \quad (6)$$

where $\langle N_c \rangle$ is the first moment of the probability density function $P(N_c)$, the statistical distribution of links around a connected vertex. In this situation, C_t is then essentially positive. The more high its value, the more connected is the pore network. For a disconnected pore network with a large number of dissociated parts, a great number of isolated vertexes emerge and α_0 dominates in the right part of equation (4). In such a case, C_t is negative, ranging

between 0 and -1 . Equation (5) can be rewritten in a similar way, but with a redefinition of $\langle N_c \rangle$ as the first moment of a probability density function $P_T(N_c)$ which includes the occurrence of isolated vertexes.

3 Topology in the vicinity of a percolation transition

The location where C_t gets null is an interesting region where one also observes a zero crossing for N_3 . As discussed elsewhere by several authors [14–20], the null N_3 value corresponds to an hyperbolic/elliptic interface transition linked to a percolation transition. From our best knowledge, there is no mathematical proof of this coincidence. However, it is encountered in several random pore networks [16], for example, generated using a correlated Gaussian random field [17,20]. In this section, we will numerically analyze the topology in the vicinity of a percolation threshold. We first generate a fully connected pore network with a correlated Gaussian field algorithm [20] using an experimental two-point correlation function measured on a TEM section of a Vycor porous glass [21]. This binary 3D reconstruction and the corresponding skeleton graph are shown in Figure 1.

Two distinct *scenarii* of evolution of this structure are now presented: (1) a series of configurations obtained by a progressive deletion of “elementary pores”; (2) a family of pore networks generated by an iterative erosion of the pore space (dilation or expansion of the solid matrix).

3.1 Deletion of elementary pores

As discussed elsewhere [11], the pixels belonging to a defined edge of the skeleton graph can be labeled with a specific index. The segmentation of the binary pore network in a set of so-called “elementary pores” consists of giving to all pixels of the initial pore network an index equal to the index of the nearest pixel belonging to the set of edges. In such a condition, each “elementary pore” is attached to a specific edge. It is then possible to generate an evolution of pore network by a successive and random deletion of these elementary pores. A snapshot of this transform, close to the disconnectedness of the pore network, is shown in Figure 2. During the evolution process, computation of the skeleton graphs is performed in parallel with the determination of the formation factor F calculated by the classical random walk algorithm [2,22]. F is a normalized estimation of the electric resistivity and allows to follow the evolution of the self-diffusive transport in a series of evolving pore networks. As the deletion proceeds, the porosity Φ , the inverse of the formation factor $1/F$ (the normalized electric conductivity) and the connection number C_t decrease. Their respective evolutions are shown in Figure 3. Interesting enough, we observe that the zero crossing point for C_t and the disappearance of the electric conductivity appear

Fig. 2. (a) A binary pore network near the percolation transition obtained by a successive and random deletion of elementary pores from the initial structure shown in Figure 1 and (b) the corresponding skeleton graph.

simultaneously at $\Phi_c = 0.127$. Φ_c can then be used as a first estimation of the percolation threshold. As shown in Figure 4, $1/F$ evolves above this threshold as:

$$1/F = (\Phi - \Phi_c)^\mu. \quad (7)$$

The exponent μ is close to 1.85, in the same order of magnitude as the values found for the conductivity exponent associated to the bond percolation of random conductor networks (roughly 2) [3].

3.2 Erosion of the pore network

Another possible scenario of evolution is to progressively erode the pore network using a cubic structural element

Fig. 3. (Color online) Evolution of the pore network during the deletion of elementary pores. Open circles and left y axis: variation of the connection number C_t with the porosity Φ . Full triangle and right y axis: variation on the inverse of the formation factor $1/F$.

Fig. 4. Evolution of $1/F$ with the difference $(\Phi - \Phi_c)$ during the deletion of elementary pores. Φ_c is defined in Figure 3. The continuous line is the best fit according to equation (6).

of $(3 \times 3 \times 3)$ voxels [12]. In such a case, we were able to obtain seven generations of altered pore networks. One of them, close to the percolation threshold, is shown in Figure 5. The respective evolution of C_t and $1/F$ is shown in Figure 6. Again, we observe that the zero crossing point for C_t and the disappearance of the electric conductivity appear simultaneously at Φ_c but at a lower porosity ($\Phi_c = 0.067$). The progressive erosion of the pore network seems to be less efficient to disconnect the pore network than the random deletion of elementary pores. As shown in Figure 7, above the threshold Φ_c , $1/F$ evolves as:

$$1/F = (\Phi - \Phi_c)^{\mu_1}, \quad (8)$$

Fig. 5. (a) A binary pore network near the percolation transition obtained by iterative erosions of the initial structure shown in Figure 1 and (b) The corresponding skeleton graph.

with $\mu_1 = 1.24$, a value much lower than the one found for the first scenario.

Comparing Figures 3 and 6, we observe a shift of C_t and $1/F$ curves to lower porosity when the erosion scenario is applied. This evolving process is less efficient to disconnect the pore network than the random deletion of “elementary pores”. The spatial distribution of pore throats and their elimination monitor the disconnection by erosion. As shown in Figure 5, this generates a connected filamentous pattern for the residual pore network. The exponent μ_1 should be compared to numerous data available in the literature and related to the analysis of grain consolidation during sintering, hot pressing or diage-

Fig. 6. (Color online) Evolution of the pore network during successive erosions. Open circles and left y axis: variation of the connection number C_t with the porosity. Full triangle and right y axis: variation on the inverse of the formation factor.

Fig. 7. Evolution of $1/F$ with the difference $(\Phi - \Phi_c)$ during successive erosions. Φ_c is defined in Figure 6. The continuous line is the best fit according to equation (7).

nesis of granular porous materials. During these processes, often simulated by a progressive erosion of the pore space, the connected paths have been observed to vanish at very low porosity and it was assumed in several cases that the percolation threshold could occur at precisely zero porosity [23]. This is why the experimental and the simulated results are either analyzed using equation (7) or with an empirical power law known as Archi’s law:

$$1/F = a(\Phi)^m. \quad (9)$$

The two exponents m and μ_1 are clearly related when Φ_c is rather low. In the literature, exponent m is found to be really sensitive to the pore network geometry [24]:

Fig. 8. (Color online) Finite size effect on the determination of the connection number C_t . Open squares: computation performed on a binary 3D image shown in Figure 1 and having a volume V . Full squares: computation performed on a subset having a volume $V/8$.

e.g., $m = 1.5$ for an assembly sphere, $m = 1.8$ to 2 for consolidated sandstone and $m = 1.3$ for unconsolidated sand pack.

In our case, our initial matrix is not a granular material but bicontinuous system where no generic convex particles can be identified locally. The erosion generates a clear disconnection of the pore network, appearing at well-defined porosity.

3.3 Comment on the finite size effect

To end this section, we have tested how the determination of C_t is influenced by the size of the initial binary image. For this prospect, we have compared the evolution of C_t during a progressive deletion of “elementary pores” for the reference volume V (the one used Sect. 3.1) and for a subvolume $V/8$. As shown in Figure 8, the two evolutions are almost similar with the same zero crossing point. A slight discrepancy is observed at low porosity where the pore network is constituted of separated small clusters. Fluctuation in the cluster number inside the small volume $V/8$ is certainly the main origin of this discrepancy.

4 Experimental example of evolving pore network: the setting of a cement paste

In this last section, we present an experimental example of an evolving pore network: the setting of a cement paste in its early age, ranging from few hours to several days [25–27].

4.1 Sample preparation and 3D X-ray microtomography

A CEM I 42.5 Portland cement (OPC) was used for this study. Pastes were prepared at a water-to-cement ratio W/C of 0.4. Cement and distilled water were mixed by hand for 5 min and then injected with a syringe into thin cylindrical glass tubes with a diameter of 600 μm and a wall thickness of 10 μm . The glass tubes were of Lindemann glass 14 type (which minimizes the scattering of X rays by the capillary). The use of a thin capillary has significant advantages in reducing the acquisition time for the projections and reducing absorption effects in the center of the reconstituted sections. After one day, the cylindrical plastics are immersed into mixing water in a sealed plastic box and, only during the tomography acquisition, the cylindrical plastics were removed. To avoid the variations due to sample preparation, the same cylindrical plastics were studied over time. Tomographic scans of these samples were performed at the Swiss light source (SLS) in Villigen (Switzerland) on the TOMCAT beamline [27]. Since X-ray tomography is a non-invasive technique, the samples were imaged in their native state (i.e., wet) at the age of 34, 83 and 150 h. Beam energy was set to 25 KeV with a constant intensity of 300 mA. Projections (1501) with an angle step of 0.119° and an exposure time of 1.8 s each were acquired on a 2048 px CCD camera equipped with a 1400 mm field of view and a $10\times$ magnification optical objective. The pixel resolution under these conditions was 0.685 μm . Reconstructed slices (tomograms) were computed using the filtered back projection algorithms [5]. It should be noted that there was no prior drying or other preparation of the sample before imaging. The short exposure times and configuration of the line ensure that there is no significant heating or drying of the sample during the acquisition of the images.

4.2 Image segmentation in three phases

A simple, generic and robust method is used to extract the components from experimental tridimensional images obtained by X-ray tomography. This extraction is decomposed in two steps: segmentation and splitting. For the segmentation step, if there is sufficient brightness between the different components, we apply a classical threshold [28] procedure followed by a succession of morphological filters [12]. If not, we apply a watershed transformation procedure. The localization of two seeds is done using a soft threshold followed by a succession of morphological filters. Generally, the segmentation visualization shows one problem: some pairs of grains close one to each other become connected. To overcome this problem, we applied a splitting procedure. This procedure is a watershed transformation applied on the inverse of the distance function of segmented image after a dynamic filter [29]. The complete segmentation strategy is presented in a comprehensive way in [30]. In the specific case of the cement paste, this strategy allows to separate the 3D gray images in three distinct phases, the unreacted solid,

Fig. 9. (Color online) Part of a 3D tomography of a cement paste after segmentation. In blue, unreacted grains, in green hydrated phase (nanoporous pore network). Complementary space: the capillary pore network.

Fig. 10. A full image of the segmented capillary pore network of the cement paste after 150 h.

the hydrated phase, also called the CSH phase which is the major and essential product of the dissolution and reprecipitation process and finally the capillary pore network full of water and observed at the length-scale resolution of about $0.7 \mu\text{m}$. A part of a 3D image after the three-phase segmentation is shown in Figure 9. A full image of the segmented pore network of the cement paste after 150 h is also shown in Figure 10. The large extension of these tomographic reconstructions makes a visual inspection of this pore network hopeless (Fig. 11). A more quantitative image analysis is needed.

Fig. 11. Time evolution of the skeleton graph associated with the capillary pore network (a) after 34 h, (b) after 83 h and (c) after 150 h.

4.3 Topology of the evolving pore network

The skeletization graphs of the capillary pore network after a setting of 34, 83 and 150 h are shown in Figure 10. With aging, one observes a progressive simplification of the graph which looks less and less dense. This evolution is essentially due to the dissolution of the initial anhydrous phases (C3S) and the precipitation of the hydrated phase (CSH). This process consumes progressively the liquid water localized initially in the capillary pore network. Meanwhile, the hydrated phase invades the capillary pore network and replaces a part of the anhydrous phases. The evolution of the connection number with the porosity is shown in Figure 12. In early time, at 34 h, the C_t reaches a relatively high value, around 0.8, very much

Fig. 12. (Color online) Evolution of the connection number C_t with the porosity of the capillary pore network.

Fig. 13. (Color online) Evolution of $1/F$ with the connection number C_t . Computation is performed on 3D binary images of the capillary pore network at three different ages.

comparable to what is found for a well-connected network. We observe that C_t changes its sign after about 100 h to become strongly negative. As previously described, the formation factor F is computed for the three binary images of the pore network. Its evolution with the connection number C_t is shown in Figure 13. A “depercolation” process is observed between 83 h and 150 h in parallel with a change of sign for C_t . At the spatial resolution of these tomographic images, typically the μm , our analysis shows that evolution of the capillary pore network with age induces an arrest of the diffusive and/or the electric long-range transport in brine. However, some recent electric measurements [31] performed on the same sample clearly show that a weak electric conductivity can be measured even after an aging of one year and half. The main con-

clusion is threefold: (i) the hydrated phase is in fact a nanoporous phase [32] and our experimental spatial resolution is not sufficient to image correctly its structure; (ii) at long time, an intermittent transport can be considered with successive localizations in the capillary and the hydrated phase pore networks; (iii) 3D multimodal analysis of the cement paste at different length-scale is needed [6].

5 Conclusion

In this paper, we have discussed the interest to use the skeleton graph to analyze the topology of evolving pore networks. This graph prevents the topology of the pore network and can be computed on segmented 3D tomographic images. It provides a relatively simple representation of a complex structure. More especially, we have discussed some topology properties of a pore network near a disconnectedness and/or a percolation threshold and their relation with diffusive transport. We show how the knowledge of the skeleton graph allows quantifying such a transition. For this, we have introduced a connection number C_t which is an intensive value, negative between -1 and zero for a set of isolated vertexes and positive for a well-connected structure. The location where C_t gets null is an interesting region where a percolation and/or disconnectedness transition is observed. This transition is analyzed using two distinct *scenarii* of evolution of a pore network: (1) a series of configurations obtained by progressive deletion of “elementary pores”; (2) a family of pore networks generated by an iterative erosion of the pore space (dilation or expansion of the solid matrix). The last part of this paper is dedicated to an experimental example of evolving pore network: the setting of an ordinary cement paste probed in its early age by synchrotron X-ray microcomputerized tomography. At the spatial resolution of these tomographic images, typically the μm , our topological analysis allows us to discuss the evolution of the diffusive and/or the electric long-range transport inside the capillary pore network. We finally emphasize the need for a 3D multimodal analysis of this material at different length-scales.

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