Journal of Computational Science 16 (2016) 177–189

Contents lists available at ScienceDirect

Journal of Computational Science

journal homepage: www.elsevier.com/locate/jocs

Pore networks subjected to variable connectivity and geometrical restrictions: A simulation employing a multicore system

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ARTICLE INFO

Article history:
Received 9 March 2016
Received in revised form 24 May 2016
Accepted 16 June 2016
Available online 20 July 2016

Keywords:
Pore network simulation
Percolation thresholds
Parallel computing
Variable connectivity
Geometrical restraints

ABSTRACT

Pore networks considering variable connectivity and geometrical restrictions among voids of assorted sizes are simulated using an 8-multicore computing system. The topology of the resulting networks is visualized in terms of the sizes and connectivity of the pores through color graphics. Results allow the calculation of percolation thresholds, correlation lengths among pores, fractal dimensions of percolation clusters, and conditional probabilities among connected pore sizes. Besides, it is possible to observe disconnected pore islands of different sizes, depending on the structural correlation among pores.

1. Introduction

Both natural and synthetic systems include the existence of a good deal of different networks either physical or virtual. The structural characterization of these networks is crucial to determine the application of the corresponding substrata. These applications include the separation of compounds by adsorption [1], the immiscible displacement of liquids [2,3], the imbibition and drainage of liquid phases [4], etc. In the case of our present interest, real porous substrata are complex physical networks that can be simulated by involving specific restraints. Previous studies have designed virtual pore networks based on simple physical considerations [5–8]. For instance, when pursuing the creation of a pore network from the interconnection of sites (spherical cavities) by means of cylindrical tubes (bonds), the key condition to fulfill is that the size (i.e. the diameter) of the bond that is interconnecting two neighboring cavities can never be larger than any of the sizes of the two sites in question. However, other restrictions can still surge, especially when some kind of additional evidence (e.g. microscopic visualization) is at hand. For example, mesoporous silicas with uniform pore sizes between 20 and 500 Å are synthesized in such a way as to obtain pores with very precise geometries an regularly arranged [9]. In this sense, an additional physical constraint could be that two adjacent (orthogonal) bonds (for simplicity, assumed as hollow cylindrical capillaries) are not allowed to intercept each other before meeting at the spherical cavity (site) to which these capillaries are leading to [10–13]. In the same context, a further parameter that has been subjected to scrutiny is the connectivity [14] (i.e. the mean number of bonds that are interconnecting the sites of the network). These characteristics have not yet been analyzed with respect to percolating and statistical properties.

The computing programs that our research group has developed so far, include Greedy and Monte Carlo algorithms [15,16]; in these cases, a valid porous network (i.e. that fulfilling the pertinent construction constraints) is eventually created; this is followed by the application of additional and valid exchanges of void sizes in order to achieve an isotropic configuration (i.e. that approximating to the maximum configurational entropy) of the pore arrangement. After accounting for the above observations, it seems that the in silico creation of a real pore network would be such that considers the incidence of physical restrictions. The contribution of this paper aims to structurally characterize porous networks endowed with geometrical restrictions and variable connectivity among pore entities. This model allows to calculate percolation

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http://dx.doi.org/10.1016/j.jocs.2016.06.003
1877-7503/

thresholds, while visualizing their corresponding paths. Besides, the conditional probabilities of finding interconnected sites and bonds of given sizes were numerically calculated. The scalability of small to large pore arrangements was feasible through the implementation of a parallel computing algorithm using a shared memory multicore system.

This paper is organized as follows. Sections 2–4 describe the theoretical background in the following manner: Section 2 presents the Dual Site-Bond Model (DSBM); Section 3 incorporates the concept of geometrical restraints; and Section 4 deals with the treatment of variable connectivity. Section 5 accounts the in silico parallel simulation of porous networks endowed with variable connectivity. Section 6 presents the structural characterization of the simulated pore networks, while Section 7 accounts for the computing results found after constructing such kind of pore networks. Finally, Section 8 outlines the conclusions and sets the perspectives.

2. The Dual Site-Bond Model (DSBM)

The structure of most real porous media is so irregular and intricate that a rigorous geometric description of such arrangements is frankly impossible to achieve. The number of pore entities per unit mass that are inherent to a typical porous medium, is a changeable quantity depending on the nature of the adsorbent. For instance, the number of spherical cavities that are proper of a SBA-16 material [17] can account as much as 10^{20} pores per gram of material. In the case of SBA-15 solids [18] consisting of independent (non-interconnected) cylindrical pores in hexagonal packing, and assuming a mean pore length of 1 μm, it is just necessary to have 15 g of this material in order to account for a total length equivalent to the distance of the earth to the sun. Nevertheless, for appraising or modeling valid pore networks is necessary to develop an appropriate statistical model, in which the following features converge:

1. To allow the incorporation of all those relevant characteristics that have been accounted for via careful experimental evidence.
2. To be simple enough as to grant the implementation of a rigorous mathematical treatment with respect to the calculation of pore network structural parameters.
3. To consider the principal textural characteristics of porous media that mainly control the outcome of capillary phenomena, as for instance adsorption, imbibition or intrusion.
4. The pore sizes are commonly distributed over a wide range of values, typically from 1 to 1000 nm (colloidal size range), and between 2 and 50 nm (mesoporous materials [19]). The throats that are connecting cavities are usually either cylindrical or slit-like shapes [18,20,21].
5. The mean number of throats that delimit a site in a pore network is called the connectivity of the system. This parameter is linked to the number of pathways or access routes among pores; the efficiency of the displacement of a given fluid from a pore network depends on this factor. From available evidence, it seems that in sedimentary rocks the connectivity ranges from 2 to 15 [22].
6. The mean pore width, the specific surface area, the porosity, and the permeability are determined from the first, second, third and fourth moments of the pore-size distributions of sites and bonds. In turn, the tortuosity of the porous medium is associated with the connectivity of the pore network, while the site-bond size ratio is related to the overlap existing between the site and bond size distributions.

Some models which can fulfill the previous conditions are real micromodels and virtual Dual Site-Bond Model (DSBM) structures. The micromodels are real pore structures in which capillary processes can be studied in the laboratory [23]. In contrast to this, the DSBM can be developed numerically through the in silico construction of pore networks. In this last model, a physical description of the porous medium is succinctly developed. Details of the DSBM can be found in reference [24].

As mentioned previously, porous media can be described adroitly by considering sites (antrae, cavities) and bonds (capillaries, throats, passages), which inevitably alternate to form an interconnected network. The connectivity C, is the mean number of bonds meeting at a site. For simplicity, the size of each entity is expressed by using only one quantity, R, defined as follows: for sites, considered as hollow spheres, R is the radius of the sphere, while for bonds, idealized as hollow cylinders open at both ends (owing to their function of passages), R is the radius of the cylinder. A twofold (sites and bonds) size distribution is established by means of the normalized size probability density functions, $F_S(R_S)$ and $F_B(R_B)$, of sites and bonds respectively. The fractions of sites, $S(R)$, and bonds, $B(R)$, of sizes smaller than a particular value R are respectively expressed as indicated in Eq. (1).

$$S(R) = \int_0^R F_S(R_S)\,dR; \quad B(R) = \int_0^R F_B(R_B)\,dR \tag{1}$$

Pore networks possess a very special property: the size of a site should be always larger than (or at least equal to) the size of any one of its delimiting bonds. This Construction Principle (CP) is of the upmost importance for the case of highly overlapped $F_S(R_S)$, $F_B(R_B)$ twofold structures, so that the elements are not free to distribute fully at random. Two self-consistency laws guarantee the fulfillment of the *CP*. The first law states that the proportion of bonds must be sufficiently large as to link all the sites corresponding to a given size distribution (Eq. (2)).

$$\text{First law: } B(R) \geqslant S(R) \quad \forall R \tag{2}$$

A second law is still necessary since when there exists an overlap between the site and bond size distributions topological correlations arise. Thus, the events of finding a site of size $R_S \in (R_S, R_S + dR_S)$ together with a size $R_B \in (R_B, R_B + dR_B)$ for a given one of its C bonds are not independent. In this case, the joint probability of such an event is estimated from Eq. (3).

$$F(R_S, R_B) = F_S(R_S)F_B(R_B)\phi(R_S, R_B)\,dR_S\,dR_B \tag{3}$$

An expression of the second law can be induced from the last expression as stated in Eq. (4).

$$\text{Second law: } \phi(R_S, R_B) = 0, \quad \forall R_S < R_B \tag{4}$$

Here, the correlation function $\phi(R_S, R_B)$ incorporates all the information about the site-bond assignment that will arise after constructing the pore network. For the simplest of cases, called the Self Consistent situation, sites and bonds are assigned to each other in the most random way as allowed by the CP, then, $\phi(R_S, R_B)$ attains the following form:

$$\phi(R_S, R_B) = \frac{\exp\left(-\int_{S(R_B)}^{S(R_S)} \frac{dS}{B-S}\right)}{B(R_S) - S(R_S)} = \frac{\exp\left(-\int_{B(R_B)}^{B(R_S)} \frac{dB}{B-S}\right)}{B(R_B) - S(R_B)} \tag{5}$$

$\phi(R_S, R_B)$ is a correlation function that is a measure of how much the sizes of a site and one of its connected bonds are related to. When the overlapping Ω is null between $F_S(R_S)$ and $F_B(R_B)$ this means that it is perfectly allowed to connect any bond chosen at random from $F_B(R_B)$ to any site chosen also at random from $F_S(R_S)$.

If we denote by Ω the overlapping area between the site and bond probability density functions, as shown in Fig. 1 for the case of uniform distributions, ϕ involves the following properties: (i) $\phi_{\Omega\to 0}(R_S, R_B) = 1$, $\forall R_S, R_B$; this means that sites and bonds are distributed completely at random, and (ii) $\phi_{\Omega\to 1}(R_S, R_B) \propto \delta(R_S - R_B)$;