

Numerical approximation to the scaling law describing the geometrical tortuosity in porous media

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Abstract

When studying porous media transport properties, it is crucial to ascertain tortuosity (τ) and its variation with porosity (ϕ). In this work, numerical methods were used to investigate this relationship. First, a digital representation of media was derived, and thereby implement an algorithm for calculating tortuosity. The program allows deriving several statistics of the paths present within the pores. The results complement the theoretical studies that suggest the existence of a scaling law in disordered media. However, this paper proposes that the relationship between τ and ϕ depends on the average fractal dimension instead of the fractal dimensionality of the optimal path. It was also confirmed that the geometry of the latter can be considered in the same universality class as those described by loopless compressible invasion percolation.

Keywords: tortuosity; porous media; percolation; numerical method; scaling law; fractal dimension.

Aproximación numérica a la ley de escalamiento que describe la tortuosidad geométrica en medios porosos desordenados

Resumen

Cuando se estudian las propiedades de transporte de un medio poroso, es importante conocer la tortuosidad (τ) y su variación con la porosidad (ϕ). En este trabajo se utilizan métodos numéricos para buscar dicha relación. Primero se obtiene una representación digital de los medios y luego se implementa un algoritmo para el cálculo de la tortuosidad. El programa permite conocer varios estadísticos de los caminos presentes al interior de los poros. Los resultados sirven de complemento a los estudios teóricos que sugieren la existencia de una ley de escalamiento en medios desordenados. Sin embargo, se propone que la relación entre τ y ϕ depende de la dimensión fractal promedio en lugar de la dimensión fractal del camino óptimo. También se verifica que la geometría de este último, puede considerarse dentro de la misma clase de universalidad que las descritas por la percolación por invasión compresible sin bucles.

Palabras clave: tortuosidad; medios porosos; percolación; método numérico; ley de escalamiento; dimensión fractal.

1. Introduction

When studying the behavior of a porous medium, it is crucial to gain information about its effective transport properties (i.e., conductivity, permeability, permittivity, and diffusivity). For example, knowledge of the soil hydraulic permeability helps predict productivity in an oil well [1] or the pollutant migration toward aquifers or other water bodies [2]. The thermal conductivity of soil allows estimate the geothermal energy production efficiency [3]. Insight into the diffusivity of soot particles is key to designing metallic or

ceramic porous media useful for the treatment of exhaust gases [4]. It is also relevant to know the hydrogen effective diffusivity when synthesizing materials for use in energy storage devices [5]. A main subject of study in materials science is the relationship between transport properties and the microstructure descriptors of a porous medium (porosity (ϕ), pore diameter, connectivity, and tortuosity (τ), among others). Through numerical, experimental, and analytical approaches, such as percolation theory, critical path analysis, and effective medium approximation; different mathematical expressions for these relations have been obtained (see

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references [6-8] for some examples). Porosity and tortuosity are among the most widely used descriptors. Conceptually, τ can be interpreted as a measurement of the geometrical complexity of a porous structure. Despite the simplicity of this concept, some authors state that tortuosity is neither understood nor consistently defined [9-11]. Carman [12] used it for the first time. He resorted to this parameter to match his experimental permeability data with the results predicted by the analytical solution to the capillary tubes problem. In literature, four classes of τ can be found. Three of them are related to the process under study (hydraulic, electric, or diffusive tortuosity) and the fourth class is associated with the morphological properties of porous media (geometrical tortuosity). In 2013, Ghanbarian et al. [9] published a critical review of the deduction, meaning, and application field of each of these definitions. The geometrical tortuosity (τ_g) defines the relationship between the average length of the paths existing within the porous channels ($\langle L_g \rangle$) and the linear dimension of the system (L):

$$\tau_g = \frac{\langle L_g \rangle}{L} \quad (1)$$

Some authors propose the use of the shortest pathway, or optimal path (L_{opt}), instead of $\langle L_g \rangle$ [9]. However, this definition is not the most widely adopted. Experimentally, τ_g can be directly obtained from tomographic or micrographic image registration [13-15]; or indirectly by using some function that relates it to another microstructural descriptor. The last one is the most widely used option, and porosity is the preferable descriptor. In literature, many equations that relate τ and ϕ are found. Some of them are compiled by Sobieski and Lipinski [16]. The abundance of mathematical expressions is attributed to the fact that each study was carried out for particular cases of porous media, thus their application ranges are limited. Some of them are useful for 3D beds of spherical particles but cannot be used for cubic particles. Other studies apply to 2D sections of overlapping square particles but not to the non-overlapping ones. Many of these equations have a common deficiency; they predict a value of $\tau_g = 0$ when $\phi \rightarrow 1$. Nevertheless, the geometrical tortuosity in this limit must be 1.

The lack of a predictive unified model has motivated investigations on more universal formulations. Based on the percolation theory and the finite-size scaling approach, Ghanbarian et al. [17] proposed a model that allows predict τ_g as a function of ϕ in a relatively wide range of two- and three-dimensional porous media. In this analytical model, which is described in more detail in section 2, the authors use the concepts of fractal length and dimension as well as the percolation theory and its critical exponents. They found that tortuosity follows a scaling law that gradually increases, while porosity decreases and diverges as ϕ approaches the percolation threshold (or critical porosity). The authors found that many of the equations relating τ and ϕ matched their proposal. Consequently, they could be considered within the percolation theory framework. This model has been used for predicting different transport parameters, such as effective permeability and electrical conductivity in concrete [6]; and thermal conductivity in porous alumina ceramics [6] and in materials with polyhedral porosity [7].

Using numerical methods, this work found that media with saturated, disordered, and randomly distributed pores follow a scaling law that relates these two microstructure descriptors: geometrical tortuosity and porosity. The percolation model is used for obtaining the digital representations of media with different porosities. An advanced algorithm was developed to derive tortuosity from the percolating paths that exist within the porous channels. Therefore, in addition to the average length, it is possible to obtain statistics, such as the median as well as the shortest and largest paths, which are not possible to derive using state-of-the-art algorithms. The program validity is verified by comparing its results with recently published results. Moreover, the universality class to which the shortest path belongs was found. The paper is organized as follows: the analytical model presenting tortuosity as a scaling law is described in section 2. The procedure used for generating the digital images of porous media and the algorithm developed for obtaining the percolating paths are described in section 3. Section 4 presents the scaling law obtained numerically and other results, whereas section 5 reports the conclusion.

2. Percolation-based tortuosity model [17]

According to the percolation theory, the mean distance between two points in a same finite cluster of pores, or correlation length (χ), is given by the scaling law:

$$\chi = C|\phi - \phi_c|^{-\nu} \quad (2)$$

ϕ_c is the critical porosity, a minimum porosity value needed for the existence of a cluster of pores that spans the network from one side to another (percolating cluster). ϕ_c depends on the type of percolation studied (bound or site, with or without trapping) and the lattice (square, cubic) [18]. For example, for site percolation in a square lattice, ϕ_c has an accepted value of 0.5927. Similar to other percolation properties, this is an accepted value since an analytical result does not exist. The accepted values are numerically obtained. C is a fitting coefficient with an accepted magnitude of 0.85 ± 0.4 for a square lattice [19]. ν is a critical exponent which only depends on the Euclidean dimension. Unlike ϕ_c , ν neither depends on the type of percolation nor the type of lattice and has an exact value. For this reason, it is known as a universal exponent. Its exact value in 2D is $\nu = 4/3$.

The porous channels have a characteristic fractal configuration [17]. The total length of a fractal path (L_F), constructed of steps of length ε , can be obtained from eq. (3):

$$L_F(\varepsilon) = L^{D^f} \varepsilon^{1-D^f} \quad (3)$$

Where D^f is the fractal dimension

Close to the critical porosity, ε is inversely proportional to the correlation length and eq. (3) can be written as [17]:

$$L_F(\chi) \propto L^{D^f} |\phi - \phi_c|^{\nu-D^f} \quad (4)$$

Taking $L_F(\chi)$ as the average length of the paths, the geometrical tortuosity can be expressed as [17]:

$$\tau_g(\phi) \propto |\phi - \phi_c|^{v-vD^f} \quad (5)$$

The correlation length predicted by percolation theory is only independent of the system size if $\chi \leq L$. When $\chi > L$, eq. (2) and consequently eq. (5) must be modified to include this dependency. Ghanbarian et al. [17] proposed to use the scaling factor $(C/L)^{1/\nu}$.

$$\tau_g(\phi) \propto |\phi - \phi_c + (C/L)^{1/\nu}|^{v-vD^f} \quad (6)$$

The fact that $\tau_g = 1$ when $\phi \rightarrow 1$ also suggests the inclusion of the factor $|1 - \phi_c + (C/L)^{1/\nu}|$ in eq. (6) for ultimately obtaining the percolation-based tortuosity model

$$\tau_g(\phi) = \frac{|\phi - \phi_c + (C/L)^{1/\nu}|^{v-vD^f}}{|1 - \phi_c + (C/L)^{1/\nu}|} \quad (7)$$

3. Numerical approximation to the tortuosity-porosity relationship

3.1 Porous media generation

The digital representations of the microstructures are obtained by randomly selecting a fraction ϕ of the nodes constituting a square lattice of linear dimension L (in this work an $L = 120$ is arbitrarily selected). These nodes are turned into the mass center of a square pore with an edge equal to a lattice unit. The phase function $(z(x, y))$ for this system is:

$$z(x, y) = \begin{cases} 1 & \text{if } (x, y) \in \phi \\ 0 & \text{if } (x, y) \notin \phi \end{cases} \quad (8)$$

where ϕ is the porosity. A cluster of pores is defined as two or more mass centers that have $z(x, y) = 1$ and occupy consecutive sites in the lattice. The control over the distribution and number of clusters is carried out by using the labeling algorithm proposed by Ramirez et al. [20]. As in real solids, there are pores in the microstructures that do not belong to the percolating cluster (occluded pores) and therefore cannot be taken into account when calculating τ . There are also blind pores that unjustifiably increase the percolating path length. Both types of pores are eliminated by doing the following transformation $z(x, y) = 1 \rightarrow 0$ (Fig. 1 a and b).

3.2 Algorithm for calculating tortuosity (Camper)

The algorithm, named *Camper* from caminos percolantes, has the objective to find as many paths as possible, but not all. The general diagram is presented in Scheme 1. Paths are found to depart from the left side of the digital representation to arrive to the right side and also in the opposite direction. All the nodes in the lattice edges [i.e., the nodes $(0,0) \dots (0,L)$ and $(L,0) \dots (L,L)$] with $z(x, y) = 1$ are the starting point for the search. These paths are formed by steps with an equivalent length of a lattice unit. In this case, there are four possibilities. When departing from the left side, we will have steps from (x, y) to $(x, y \pm 1)$, which will be cited as right (–) or left (+), or as

$(x \pm 1, y)$, cited as forward (+) or backward (–). When departing from the right side, the signs change. The steps are preferentially taken forward provided that there are not obstacles. Obstacles are the pores walls. There are two types of pores walls in the digital representations: those that can only be evaded in one direction (to the left or right, they will be named single obstacles) and those that can be evaded in both directions (double obstacles) (Fig. 1 d). In the first case, the obstacle is bordered in the direction, in which $z = 1$, until there is the possibility of continuing the path forward. In the second case, before bordering the pore wall a direction is randomly selected. Finally, $\langle L_g \rangle$ is obtained after averaging the length of the different paths. Any configuration of steps with loops is rejected for the τ_g calculation.

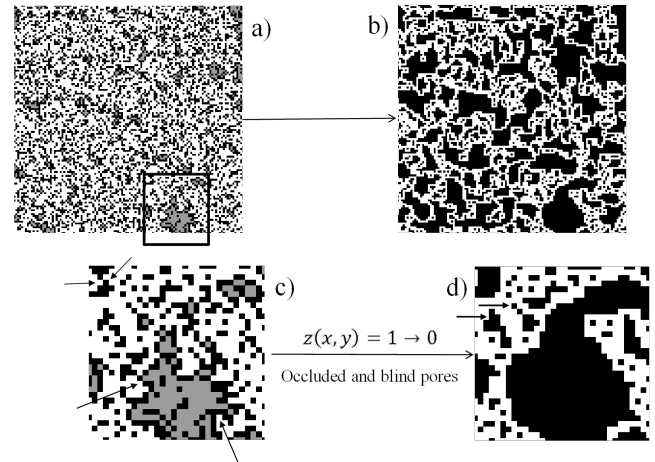


Figure 1. a) Digital representation of porous media with $\phi = 0.65$; the occluded pores are shown in gray. b) Lattice used for calculating τ , after eliminating occluded and blind pores. c) Zoomed view of the framed region, whereby arrows indicate some blind pores. d) Arrows point out a single obstacle (only evadable to the left) and a double one (Obstacles are described in section 3.2.)

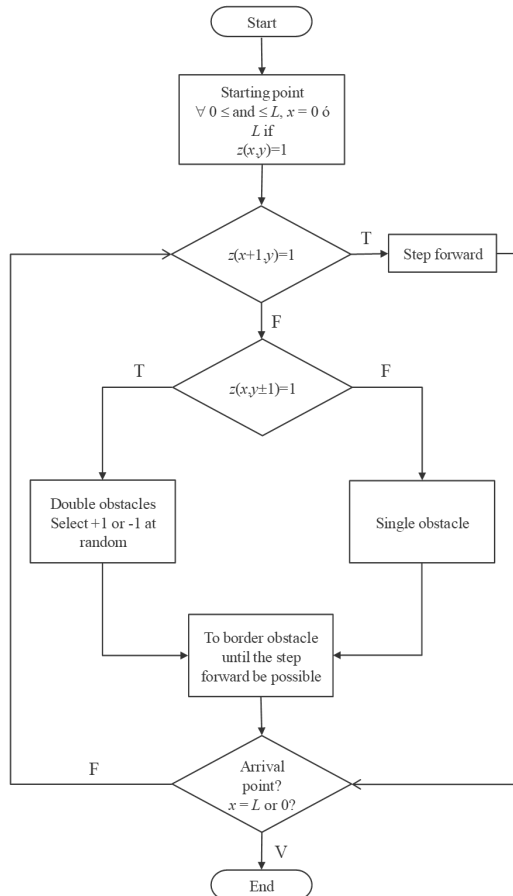
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4. Results and discussion

4.1 Algorithm validation

The algorithm validity is corroborated by comparing results with those reported by Roque and Costa in 2020 [14] and Al-raoush and Madhoun in 2017 [15]. They also developed algorithms for calculating τ_g from image registration. From reference [14], the synthetic image called curved line is taken (see Fig. 2 a). From reference [15], the micrograph of the natural media called M1, a mixed sand of silica and quartz, is taken (see Fig. 2 b). As stated in section 3.1, *Camper* only reads square images; for this reason, the rectangular image of M1 is cut as presented in Fig. 2 c. Both digital representations were adjusted to have a resolution of 240×240 pixels ($L = 240$). Table 1 presents the results.

One of the advantages of this program is the descriptive statistical analysis of the detected paths, which is not possible with the other proposals. *Camper* allows establish the number of paths used for calculating τ_g as well as the maximum and minimum lengths, and the median. In general, there is a good agreement between the tortuosities reported in literature and those calculated in this work. Particularly for M1, where the same value of tortuosity was obtained.



Scheme 1. Flowchart of the algorithm used to find the percolating paths. Source: elaborated by the author

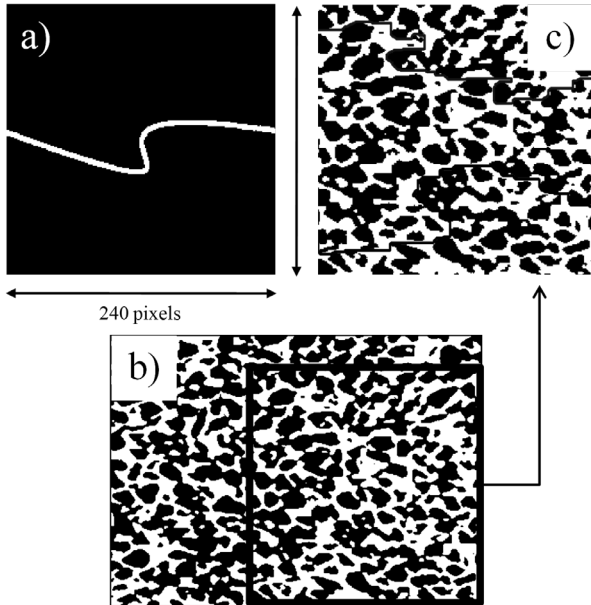


Figure 2. Digital representations used for corroborating the algorithm. a) Curved line, b) Micrography of the mixed sand (M1 sample), the frame encloses the area used for the calculation. c) Zoomed view of the used area. As an example, in c) two of the paths found by the program are presented. Note how they evade the blind pores. Source: adapted from references [14] and [15] with the author's permission

Table 1. Results used for validating the algorithm

	Medium	Curved line	M1
Total number of paths	9		90
$\langle L_g \rangle$	291.2		375.8
Standard deviation	28.2		50.4
L_{opt}^a	260		296
L_{Me}^b	313		367
L_{max}^c	317		504
$\tau_g [14]^d$	1.36		
$\tau_g [15]^d$			1.57
$\tau_g Camper^d$	1.21±0.1		1.57±0.2

^a Length of the shortest percolating path or optimal path

^b Median for paths

^c Largest path length

^d Tortuosities published in [14] and [15] and the one obtained by *Camper*

Source: elaborated by the author

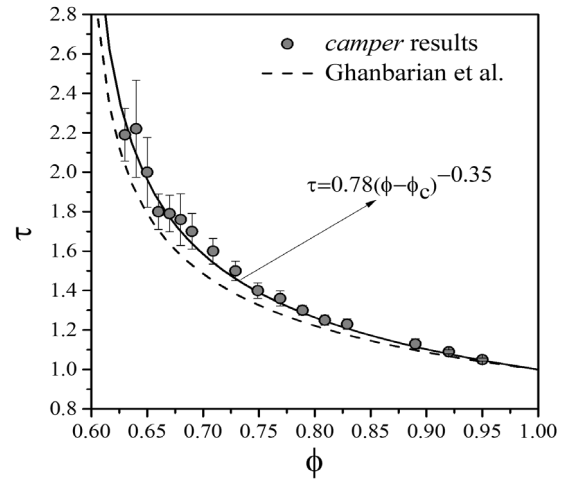


Figure 3. Relationship between the geometrical tortuosity and porosity. The circles indicate the numerically obtained results. The vertical bar represents the standard deviation. The solid line is the equation that better fits the data. The dashed line is the scaling law analytically obtained by Ghanbarian et al. (eq. 8). Source: elaborated by the author

4.2 Detection of a scaling law

With the algorithm, τ_g is calculated in media with porosity from 0.63 to 0.95. Fig. 3 presents the results. This range of porosity was used because $\chi \leq L$; hence, the correlation length is independent of the size of the system and the scaling factor can be eliminated in eq. (7). However, while we are closer to $\phi = 0.63$, the finite-size effects appear as greater standard deviations, which lead to greater uncertainty in the calculation of τ_g . Ghanbarian et al. [17] proposed to use the fractal dimension of the optimal path [21, 22] ($D^{opt} = 1.22$ in 2D) as D^f in eq. (7). By replacing the accepted values for ϕ_c and D^{opt} , the universal exponent in (7), and by solving the denominator such equation can be rewritten as:

$$\tau_g(\phi) = 0.78|\phi - \phi_c|^{-0.29} \quad (9)$$

This equation is presented as a dashed line in Fig. 3. The solid line is the scaling law that better fits the simulated tortuosity values and is

$$\tau_g(\phi) = 0.78|\phi - \phi_c|^{-0.35} \quad (10)$$

There is a good correlation between both equations. After solving D^f from $\nu - \nu D^f = -0.35$, a fractal dimension value of 1.26 was found. As expected, $D^f > D^{opt}$ since all percolating paths, and not only the shortest one, were used for its calculation. Consequently, D^f can be interpreted as an average fractal dimension. When D^{opt} is used in eq. (7), as Ghanbarian et al. do so, the geometrical tortuosity is obtained in terms of the L_{opt}/L relationship and not as the relationship between the average length of the paths and the linear dimension of the system. To adjust to the most accepted definition of tortuosity (eq. 1), the use of the average fractal dimension found in this work, instead of D^{opt} , is proposed. Eq. (9) fulfills $\tau_g \rightarrow 1$ when $\phi \rightarrow 1$.

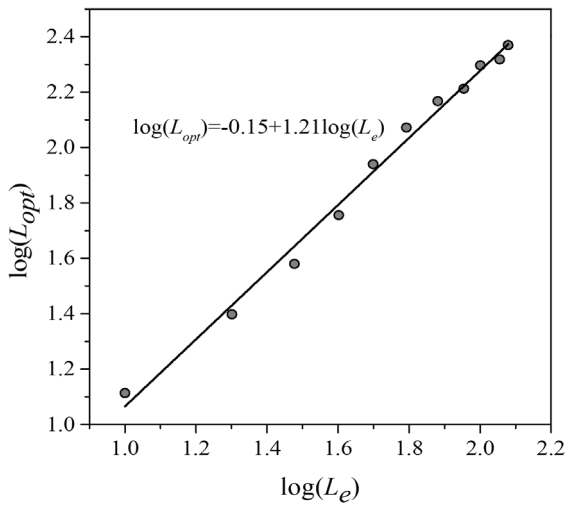


Figure 4. Logarithmic relationship between the optimal path length and the linear dimension of the system. The slope gives the value of the fractal dimension, D^{opt}

Source: elaborated by the author

4.3 Optimal path and fractal dimension

In this section, the capacity of *Camper* for assigning a value to the optimal path length was used. As mentioned before, when $\chi > L$ the percolation properties depend on the system dimension. It has been possible to establish that under these conditions and in the percolation threshold, the length of such path scales with its Euclidean distance, meaning that $L_{opt} \sim L_e^{D^{opt}}$ [22,23]. L_e is the distance between the starting point and the arrival point, $L_e \geq L$. It is proposed to obtain D^{opt} from the slope of the relation $\log_{10}(L_{opt})$ vs. $\log_{10}(L_e)$ constructed from twelve media with $\phi = 0.6$ and $L = 10, 20, \dots, 110, 120$. The critical porosity ($\phi_c = 0.5927$) could not be used because, in some cases, the program does not find a percolating path. Results are presented in Fig. 4. Despite not using ϕ_c , the value obtained for D^{opt} was 1.21, which agrees adequately with the accepted value of 1.22 [21,22]. This suggests that the optimal path existing within the porous channels can be considered as in the same

universality class of the geometries described by loopless compressible invasion percolation [21,22,24].

5. Conclusions

The sequence followed by the developed algorithm, i.e., removing occluded and blind pores, paths starting in different points of each media edge, steps forward, which border obstacles, and avoiding loops allows to derive a solid statistical description of the percolating paths existing within porous media. For media with saturated, disordered, and randomly distributed pores, there is a power law that relates geometrical tortuosity and porosity. The exponent of such law can be obtained from the critical exponent of the correlation length and the average fractal dimension instead of the fractal dimension of the optimal path. The fractal dimension of the optimal path that exists within this type of microstructures is in the same universality class that the Eden growth observed in tissue or bacterial cultures or the geometry of the undirected polymers.

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