Tracer dispersion in a percolation network with spatial correlations

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We analyze the transport properties of a neutral tracer in a carrier fluid flowing through percolationlike porous media with spatial correlations. We model convection in the mass transport process using the velocity field obtained by the numerical solution of the Navier-Stokes and continuity equations in the pore space. We find that the resulting statistical properties of the tracer show a transition from a subdiffusion regime at low Péclet number to an enhanced diffusion regime at high Péclet number.

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I. INTRODUCTION

The phenomenon of hydrodynamic dispersion—the unsteady transport of a neutral tracer in a carrier fluid flowing through a porous material—has been widely investigated in the fields of petroleum and chemical engineering [1–4]. One can identify different regimes of tracer dispersion according to the Péclet number $\mathrm{Pe} \equiv v\ell/D_m$, which is the ratio between the typical time for diffusion ℓ^2/D_m and the typical time for convection ℓ/v. Here v is the velocity of the carrier fluid, ℓ a characteristic length scale of the porous media, and D_m the molecular diffusivity of the tracer.

In the small-Péclet-number regime, molecular diffusion dominates the way in which the tracer samples the flow field. In the large-Péclet-number regime, also called *mechanical dispersion*, convection effects are significant; the tracer velocity is approximately equal to the carrier fluid velocity, and molecular diffusion plays little role. The tracer samples the disordered medium by following the velocity streamlines. In a random walk picture, we may think of a tracer particle following the direction of the velocity field, and taking steps of length ℓ and duration ℓ/v.

The classical approach to model dispersion in porous media is to consider microscopically disordered and macroscopic isotropic and homogeneous porous materials. Under these conditions, dispersion is said to be *Gaussian* and the phenomenon can be mathematically represented in terms of the convection-diffusion equation [2]. This traditional formalism, which is valid for Euclidean geometries, cannot be adopted to describe the global behavior of hydrodynamic dispersion in heterogeneous systems. Specifically, in the case of percolation porous media, the breakdown of the macroscopic convective-diffusion description is a direct consequence of the self-similar characteristic of the void space geometry.

The movement of a tracer in a fluid flow field with a broad velocity distribution is an interesting phenomenon that displays a rich variety of physical behaviors. Consider, e.g., fluid flow in percolation clusters near the percolation threshold—a model system relevant to a porous medium with stagnant small-velocity zones that are linked with large-velocity zones. In this case the typical time for convection ℓ/v is without bound since the velocity can be arbitrarily small in some fluid elements of the void space. Saffman showed [1] that the mean square duration of a tracer step is not finite but diverges logarithmically unless an upper cutoff is introduced into the typical time step. This upper cutoff is imposed by the mass transport mechanism of molecular diffusion.

Molecular diffusion is expected to affect the tracer motion in two ways [1].

(i) A quantity of material may cross from one streamline with fluid velocity v to another by *lateral* diffusion if the time step for convection $\ell_\parallel/v$ is larger than t_1, where $t_1 = \ell_\perp^2/2D_m$ is the characteristic time for molecular diffusivity effects to become appreciable [5] and $\ell_\parallel$ and $\ell_\perp$ are the longitudinal and lateral pore lengths, respectively (with respect to the flow direction) [1]. Thus, if $\ell_\parallel/v \gg t_1$, the tracer has enough time to diffuse across the pore, and the time step associated with such a move is $\Delta t = t_1$. When $\ell_\parallel/v \ll t_1$, the time duration of a convective step is smaller than the time required for molecular diffusion, and the tracer moves with the carrier fluid taking a step of duration $\Delta t = \ell_\parallel/v$.

(ii) An amount of material may be transported by diffusion *along the pore*. The same considerations as in point (i) lead to a time step $\Delta t = \ell_\parallel/v$ in which convection dominates when $\ell_\parallel/v \ll t_0 = \ell_\parallel^2/2D_m$. Here the typical length scale is the longitudinal length of the pore $\ell_\parallel$. If $\ell_\parallel/v \gg t_0$, diffusion dominates and the tracer takes a time step $\Delta t = t_0$.

Here we propose a model of tracer dispersion in a porous medium. The porous medium is composed of blocks of impermeable material that occupy, with a given probability p, a square lattice. We consider a lattice at the site percolation threshold, so an incipient spanning cluster is formed that connects the two ends of the lattice. Previous studies modeled the convective local ''bias'' for the movement of the neutral tracer in the porous media assuming Stokes flow [2]. Even at macroscopically small-Reynolds-number conditions, this assumption might be violated in real flow through porous media, especially in the case of heterogeneous materials (e.g., percolationlike structures) where a broad distribution of pore sizes can lead to a broad distribution of local fluxes. As a consequence, inertial effects might be locally relevant. To avoid this problem, we use the steady-state velocity field obtained by solving the full set of Navier-Stokes in the per-

colation geometry. Then we study the transport properties of a dynamically neutral tracer moving in the flow field.

II. MODEL OF TRACER DISPERSION IN A POROUS MEDIUM

We treat the competition between the effects of convection and diffusion. The velocity field presents a broad scale-invariant power-law distribution of magnitude values, and we find that there are regions of very small velocity in which the tracer can be trapped. If convection is important, the tracer follows the streamlines in the flow field. When a very small velocity region is reached, molecular diffusion effects cannot be neglected, since by diffusion the tracer may access the stagnant zones—where it then spends a long time. We shall see that the statistical properties of the tracer—e.g., the first-passage time and the root mean square displacement—show a transition from a subdiffusion regime to an enhanced diffusion regime as the Péclet number is increased. The existence of the stagnant zones is also related to the geometrical properties of the medium—whether it is correlated or uncorrelated in the occupancy variables of the percolation cluster.

We start by describing the disordered medium and the velocity field. Our basic model of a porous medium is a percolation model at threshold [6] modified to introduce correlations among the occupancy units [7]. Transport in porous media with a broad distribution of conductance values is dominated by those regions where the conductances are larger than some critical value. This critical value corresponds to the smallest conductance such that the set of conductances above the threshold forms a conducting spanning cluster [8]. This cluster is the critical percolation cluster at the threshold which we use in our simulations.

We assume the existence of long-range correlations because it provides a better mathematical representation for transport properties in real porous rocks [9]. For instance, the permeability of sandstone can fluctuate over short distances, and these fluctuations significantly affect any fluid flow through the pore space. Previous models assumed that these fluctuations were random and without short-range correlations. However, permeability is not the result of a simple random process. Geologic processes, such as sand deposition by moving water or wind, impose their own kind of correlations.

The mathematical approach we apply to describe this situation is correlated percolation. In the limit where correlations are so small as to be negligible [6], a site at position $\vec{r}$ is occupied if the occupancy variable $u(\vec{r})$ is smaller than the occupation probability $0 \leq p \leq 1$; the variables $u(\vec{r})$ are uncorrelated random numbers with uniform distribution in the interval [0,1]. To introduce long-range power-law correlations among the variables, we convolute the uncorrelated variables $u(\vec{r})$ with a suitable power-law kernel [10], and define a new set of occupancy variables $\eta(\vec{r})$ with long-range power-law correlations that decay as $r^{-\gamma}$, where $r \equiv |\vec{r}|$ (in the following we will set $\gamma = 0.4$).

We solve the full set of Navier-Stokes and continuity equations at the percolation threshold of a square lattice with 64×64 cells and cell edge $L = 1$ m. Grid element lengths
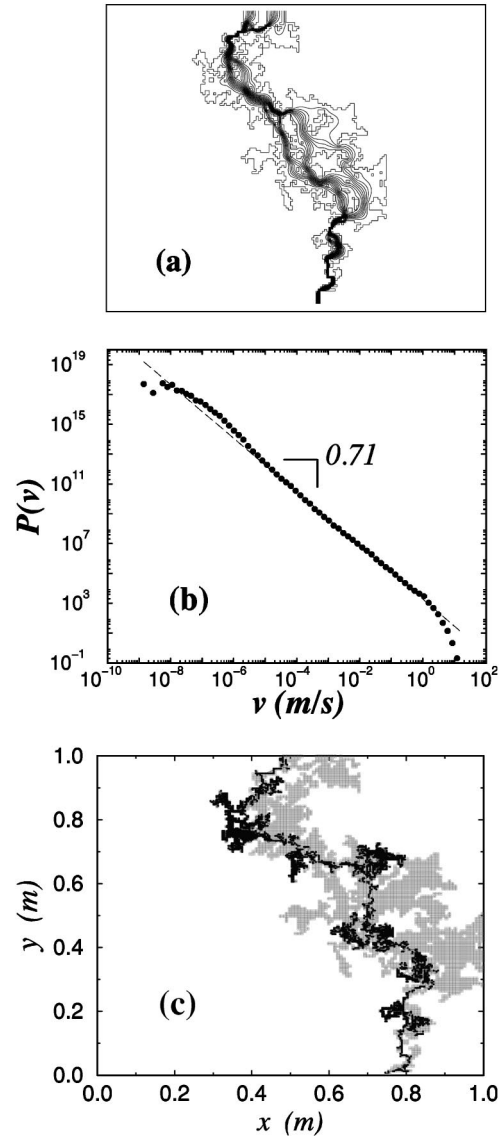

FIG. 1. (a) Typical streamlines of the velocity field in a correlated percolation cluster. (b) Velocity magnitudes probability distribution averaged over three realizations of the percolation clusters. (c) Tracer diffusion in the porous medium shown in (a), for Pe = 1.4. We release a walker and the black dots indicate the sites visited by the walker.

with 1/4 of the solid cell edge, $\ell_{\parallel} = \ell_{\perp} = \ell = L/256$, have been adopted to discretize the governing balance equations within the pore space domain [11]. Figure 1(a) shows a typical velocity field, while Fig. 1(b) shows the probability distribution of the velocity magnitudes averaged over three realizations of the percolation clusters. We find that the data are well fit by a broad power law of the type [11]

$$P(v) \sim v^{-0.71}. \tag{1}$$

Next we analyze the transport properties of a neutral tracer moving in the fluid. We use a discrete random walk model for the tracer motion. Previous discrete particle models [12] consider that the entrance probability to a region is proportional only to the flux. It has been shown [3,13] that these models fail when there is a broad distribution of ve-

locities, where regions of small velocity coexist with regions of large velocities. To circumvent this problem, we include molecular diffusion in our discrete particle model according to the Saffman theory of dispersion in porous media. We define the walker motion as a competition between flow-driven convection and molecular diffusion. To allow for comparison among different regimes of tracer dispersion, we define a macroscopic Péclet number as $Pe \equiv V \ell / D_m$, where $V = 1$ m/s is the fluid velocity at the inlet boundary of the lattice. At a given position $\vec{r}$ in the pore space, we define the time scale for convection

$$t_c \equiv \ell / v(\vec{r}), \qquad (2)$$

and a time scale for diffusion

$$t_d \equiv \ell^2 / 2D_m = Pe \ \ell / [2V]. \qquad (3)$$

Consider the walker at a site $\vec{r}$ and fluid velocity $v(\vec{r})$, with four nearest neighbor sites. The probability of choosing each of the four nearest neighbor sites is defined according to the rates of convection and diffusion which are equal to the inverse convection time and the inverse diffusion time, respectively [3,13]. We call site 1 the nearest neighbor site where the velocity $v(\vec{r})$ points to. Then the jumping rate to site 1 is

$$R_1 = 1/t_c + 1/t_d. \qquad (4)$$

The jumping rate to the rest of the sites is equal to the diffusion rate

$$R_\alpha = 1/t_d \quad (\alpha = 2,3,4), \qquad (5)$$

since the tracer can access these sites only by diffusion. Thus the probability to jump to the nearest neighbor sites is

$$p_\alpha \equiv R_\alpha \Big/ \sum_{\beta=1}^{4} R_\beta, \quad \alpha = 1, \ldots, 4. \qquad (6)$$

III. NUMERICAL SIMULATIONS

We first discuss the case of the large Péclet number, Pe $= 1.4$, so the value of t_d is such that diffusion only occurs in regions of small fluid velocity. A typical tracer trajectory is shown in Fig. 1(c). We see that the tracer particle performs a walk with long trajectories following the streamlines of the fluid followed by periods where it gets trapped in small velocity zones. These ''stagnant zones'' in the pore space differ significantly from the dangling ends of the analogous electrical problem (i.e., the parts of the infinite cluster connected by only one site to the backbone). The tracer enters these regions by diffusion, and requires a long time to escape. After escaping, the particle performs another trajectory following the streamlines until it penetrates into the next small velocity region. The tracer trajectory resembles a quasi-one-dimensional trajectory of ''channels and blobs.'' The ''channel and blobs'' picture is the analog for this problem of the traditional ''links and blobs'' picture associated with anomalous diffusion in percolation clusters [14–16].

We analyze the probability density of transit time $P(t)$ for a given system size L, i.e., the time required for the tracer to
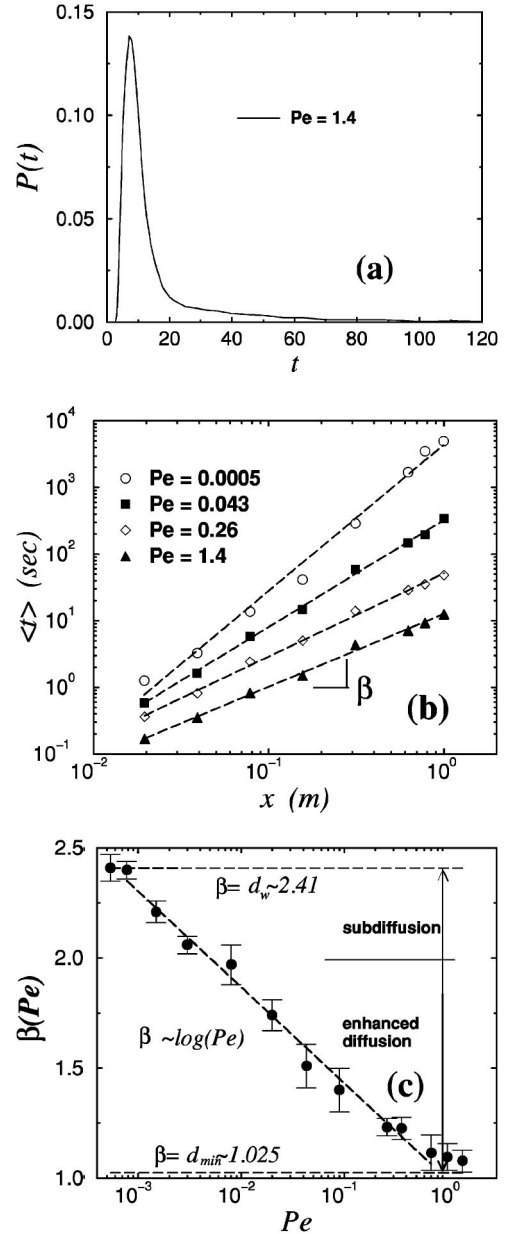

FIG. 2. (a) Probability density of the transit time $P(t)$ for a given system size and Pe$=1.4$, averaged over three realizations of the percolation clusters, showing a non-Gaussian behavior. (b) Mean value of the transit times as a function of the traverse distance x for different Péclet numbers, averaged over three realizations of the percolation clusters. (c) Transit time exponent as a function of the Péclet number. The upper limit corresponds to the value of the exponent of the anomalous diffusion in correlated percolation clusters d_w. The lower limit corresponds to the minimum path exponent d_{min} [17], and in between those limits we find that our data can be approximated by a logarithmic dependence with Pe.

traverse the system size L from the inlet line, for a given Pe$=1.4$. We find [Fig. 2(a)] that the transit time probability density for a fixed L and Pe has a non-Gaussian shape as found in experiments and simulations [2–4]. Moreover, we find that the average time required for the tracer to traverse a given distance x from the inlet line, $0 < x < L$, follows a power law [Fig. 2(b)]

$$\langle t \rangle \sim x^\beta \qquad (7)$$

where $\beta \simeq 1.08$ when Pe$=1.4$.

The transit time exponent β is not universal and depends on Pe [Fig. 2(b)]. In fact, we find that there is a regime of subdiffusion at low Péclet numbers where $\beta > 2$, and diffusion dominates over convection. At higher Péclet numbers there is a transition to an enhanced diffusion regime dominated by convection where $1 < \beta < 2$. Moreover, we expect two limiting regimes. If convection dominates completely (mechanical dispersion), then the tracer should follow the minimum path along the spanning percolation cluster. The minimum path length $\ell_{\min}$ scales as $\ell_{\min} \sim x^{d_{\min}}$ where $d_{\min}$ is the fractal dimension of the minimum path distance between two points separated by a linear distance x [6]. If the tracer moves with a constant velocity, we can identify the minimum path distance with the transit time, so $\beta = d_{\min}$. This is the lower limit of the transit time exponent, and we confirm this prediction since we obtain $\beta \gtrsim d_{\min}$ when Pe is large [Fig. 2(c)] [17].

The other limit at larger diffusivities—the anomalous diffusion case [16]—corresponds to the regime dominated completely by diffusion, and the transit time scales as $\langle t \rangle \sim x^{d_w}$, where d_w is the random walk fractal dimension. The value d_w depends on the degree of correlation, with $d_w = 2.87$ for the uncorrelated percolation limit [6] and $d_w = 2.41$ [7] for the correlated percolation problem we study ($\gamma = 0.4$). We see that the limiting cases of our calculations agree with these predictions [Fig. 2(c)]. Between these two limiting cases, we find that the transit time exponent can be approximated by

$$\beta(\mathrm{Pe}) \sim \log(\mathrm{Pe}). \tag{8}$$

We also perform simulations on uncorrelated percolation clusters. We find an enhanced diffusion regime and a subdiffusion regime as well. However, due to the tortuosity of the uncorrelated percolation clusters at the threshold, the regions of low velocity where the walker is trapped are not present as we found in the case of enhanced diffusion at high Péclet numbers in correlated clusters shown in Fig. 1(c). Thus, we conclude that the existence of the ''channels and blobs'' structure found in the case of dispersion in correlated clusters at high Péclet numbers is a by-product of the dynamical properties of the tracer moving in a broadly distributed velocity field plus the geometrical properties of the particular porous medium treated here. The compact features of long-range correlated percolation clusters allows the tracer to perform large steps following the streamlines of the fluid without encountering obstacles during the random walk process.

IV. DISCUSSION

In summary, we find a transition from a subdiffusion regime to an enhanced diffusion regime as the Péclet number is increased. In the enhanced diffusion regime the trajectory of the tracer particle is reminiscent of a ''channels and blobs'' picture. Interestingly, this fact should be relevant to elucidate the mass and momentum transport mechanisms responsible for the dispersion regime called ''holdup dispersion'' [2]. Tracer experiments indicate that this regime of strong dependence between dispersion measurements and Péclet number is typical of percolationlike porous materials.

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