Tracer Dispersion in Porous Media with Spatial Correlations

Hernán A. Makse 1−3, José S. Andrade Jr. 1, and H. Eugene Stanley 3
1 Departamento de Física, Universidade Federal do Ceará, 60451-970 Fortaleza, Ceará, Brazil
2 Schlumberger-Doll Research, Old Quarry Road, Ridgefield, CT 06877
3 Center for Polymer Studies and Physics Dept., Boston University, Boston, MA 02215

(March 24, 2022)

We analyze the transport properties of a neutral tracer in a carrier fluid flowing through
percolation-like porous media with spatial correlations. We model convection in the mass trans-
port process using the velocity field obtained by the numerical solution of the Navier-Stokes and
continuity equations in the pore space. We show that the resulting statistical properties of the tracer
can be approximated by a Lévy walk model, which is a consequence of the broad distribution of
velocities plus the existence of spatial correlations in the porous medium.

The phenomenon of hydrodynamic dispersion—the un-
steady transport of a neutral tracer in a carrier fluid flow-
ing through a porous medium—has been widely investi-
gated in the fields of petroleum and chemical engineering
[1,2]. One can identify different regimes of tracer disper-
sion according to the Péclet number \( \text{Pe} \equiv v/\ell \), which
is the ratio between the typical time for diffusion \( \ell^2/\langle D \rangle \)
and the typical time for convection \( \ell/v \). Here \( v \) is the
velocity of the carrier fluid, \( \ell \) a characteristic length scale
of the porous media, and \( \langle D \rangle \) 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, called mechanical dispersion, convection effects are signif-
cient; 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 \( \ell \) and duration \( v \).

The classical approach to model dispersion in porous
media is to consider microscopically disordered and
macroscopic isotropic and homogeneous porous materi-
als. 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 Eu-
clidean geometries, cannot be adopted to describe the
global behavior of hydrodynamic dispersion in hetero-
gegeneous systems. Specifically, in the case of percola-
tion porous media, the breakdown of the macroscopic
convective-diffusion description is a direct consequence
of the self-similar characteristic of the void space geo-
metry.

Here we discuss the interesting physics that arises when
the tracer moves in a flow field with a very broad veloc-
ity distribution. 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 \( \ell/v \) 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 cut-off is intro-
duced into the typical time step. This upper cut-off is
imposed by the mass transport mechanism of molecular
diffusion.

Molecular diffusion is expected to affect the tracer mo-
tion in two ways [1].

(i) A quantity of material may cross from one stream-
line with fluid velocity \( v \) to another by lateral diffusion if
the time step for convection \( \ell/v \) is larger than \( t_1 \), where
\( t_1 = \ell^2/2\langle D \rangle \) is the characteristic time for molecular
diffusivity effects to become appreciable [3] and \( \ell \) and \( \ell \)
are the longitudinal and lateral pore lengths, respec-
tively (with respect to the flow direction). Thus, if
\( \ell/\ell_2 \gg 1 \), 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 \( \ell/\ell_2 \ll 1 \), the time duration of a con-
vective step is smaller than the time required for molec-
ular diffusion, and the tracer moves with the carrier fluid
taking a step of duration \( \Delta t = \ell/v \).

(ii) An amount of material may be transported by dif-
fusion along the pore. The same considerations as in
point (i) lead to a time step \( \Delta t = \ell/\ell_2 \) in which con-
vection dominates when \( \ell/\ell_2 \ll t_0 = \ell^2/2\langle D \rangle \). Here the
typical length scale is the longitudinal length of the pore
\( \ell \). If \( \ell/\ell_2 \gg t_0 \), 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 [3]. Even at macroscopically small
Reynolds conditions, this assumption might be violated
in real flow through porous media, specially in the case
of heterogeneous materials (e.g., percolation-like struc-
tures) where a broad distribution of pore sizes can lead
to a broad distribution of local fluxes.
FIG. 1. (a) Typical stream-lines of the velocity field in a correlated percolation cluster. (b) Velocity magnitudes probability distribution averaged over 5 realizations of the percolation clusters. (c) Tracer diffusion in the porous medium the tracer may access the stagnant zones—where it then spends a long time. We shall see that due to the existence of these stagnant zones, the statistical properties of the tracer—e.g., the first-passage time and the root mean square displacement—can be understood using a Lévy walk model for the tracer motion. The existence of Lévy statistics 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 modified to introduce correlations among the occupancy units. We assume the existence of correlations because we obtain a better mathematical representation of transport properties—such as sandstone permeability—by assuming the presence of long-range correlations in the permeability fluctuations of the porous rock. The permeability of rocks such as sandstone can fluctuate over short distances, and these fluctuations significantly affect any fluid flow through the rock. 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, 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 \( \gamma \), and define a new set of occupancy variables \( \eta(\vec{r}) \) with long-range power-law correlations that decay as \( \eta(r) \equiv |r|^\gamma \) (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 \( 64 \times 64 \) cells and cell edge \( L = 1 \) m. Grid element lengths 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. Figure 1a shows a typical velocity field, while Fig. 1b shows the probability distribution of the velocity magnitudes averaged over five realizations of the percolation clusters. We find that the data are well fit by a broad power-law of the type

\[
P(v) \sim v^{-0.71}.
\]  

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. Following 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 \( Pe \equiv v_m \ell / D_m \), where \( v_m = 1 \) 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 \( t_c \equiv \ell / v(\vec{r}) \). We choose a convective or diffusive move, and a corresponding time step \( \Delta t \) according to:

\[
\begin{align*}
t_c < t_d & , \quad \text{convection, } \Delta t = t_c \quad (2) \\
t_c > t_d & , \quad \text{diffusion, } \Delta t = t_d. \quad (3)
\end{align*}
\]

Here \( t_d \equiv \ell^2 / 2D_m = Pe \ell / (2v_m) \) is the characteristic time above which diffusion effects become relevant. If the convection move is accepted, then the tracer moves to the nearest-neighbor site in the direction given by the velocity of the fluid and the clock is updated according to \( t \rightarrow t + t_c \). If the diffusion move is accepted, then the tracer moves to one of the four nearest-neighbor positions with equal probability and the clock is updated according to \( t \rightarrow t + t_d \).

We next discuss the case of large Péclet number, \( Pe = 1.7 \), so the value of \( t_d \) is such that diffusion only occurs in regions of small fluid velocity. Typical tracer trajectories are shown in Fig. 2a. We see that the tracer particles perform walks with very long straight trajectories followed by periods where they get 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 almost ballistic trajectory until it penetrates into the next small velocity region. The tracer trajectory resembles a quasi-one-dimensional channel of “tubes and blobs.” The “tubes and blobs” picture is the analog for this problem of the traditional “links and blobs” picture associated with anomalous diffusion in percolation clusters [9,10].

We analyze the transit time, i.e., the average time required for the tracer to traverse a given distance \( x \) from the inlet line, \( 0 < x < L \), for different Péclet numbers. We find (Fig. 2a) that the transit times follow a power law

\[
\langle t \rangle \sim x^\beta \quad (4)
\]

where \( \beta \approx 1.26 \) when \( Pe = 1.7 \).

In the “tubes and blobs” picture, we define a tube as the set of steps taken by the tracer following a fixed direction, and we analyze the statistical distribution of the tube length \( s \). In stagnant zones where diffusion is dominant, the tracer is expected to change direction every time step, so that \( s \simeq \ell \). In regions in which convection dominates, the tracer moves in ballistic trajectories limited only by impermeable obstacles. Since long-range correlated clusters are very compact (Fig. 2b), we expect \( s \gg \ell \) and the tube length distribution to be broad. Both expectations are corroborated by our calculations.

Figure 2b shows the probability distribution \( P(s) \) for two different values of \( Pe \). We find that for sufficiently large \( Pe \), the step lengths follow a scale invariant power law distribution

\[
P(s) \sim s^{-2.35} \quad [Pe = 1.7],
\]

while for small \( Pe \) values, when diffusion is the dominant mechanism in the entire pore space, the distribution is Gaussian

\[
P(s) \approx e^{-\frac{1}{2}(s/s_0)^2} \quad [Pe = 0.1],
\]

FIG. 2. (a) Transit times for different Péclet numbers, averaged over five realizations of the percolation clusters. (b) Probability distributions of steps for the case of \( Pe = 1.7 \) (power-law distribution) and \( Pe = 0.1 \) (Gaussian distribution). (c) Transit time exponent as a function of the Péclet number.
with $s_0$ a characteristic jump length.

In case (3), the step lengths statistics can be considered as a Lévy walk, i.e., a random walk process in which the jump distribution is a power law [1]

$$P(s) \sim s^{-\mu}. \quad (7)$$

A random walker with a distribution (3) travels a typical distance $r \sim t^{2-\mu/2}$, when $2 < \mu < 3$. Thus, the transit time for a Lévy walker with jump statistics given by (3) is [1]

$$\langle t \rangle \sim x^{2/(4-\mu)}. \quad (8)$$

For $\mu = 2.35$— the value we find in our simulations for $Pe = 1.7$— we obtain $\langle t \rangle \sim x^{1.21}$, which agrees with the scaling found when we calculate the transit time directly, $\langle t \rangle \sim x^{1.26}$ from Fig. 2a, and confirms the validity of the Lévy walk picture as an accurate description of the tracer motion at large $Pe$.

The transit time exponent $\beta$ is not universal and depends on $Pe$ (Fig. 3). In fact we find that the Lévy statistics approximates well the value of $\beta$ in the entire enhanced diffusion regime $1 < \beta < 2$, while in the sub-diffusion regime $\beta > 2$, the Lévy statistics Eq. (3) ceases to be valid. 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 $\ell_{\text{min}}$ scales as $\ell_{\text{min}} \sim x^{d_{\text{min}}}$ where $d_{\text{min}}$ is the fractal dimension of the minimum path distance between two points separated by a linear distance $x$. If the tracer moves with a constant velocity, we can identify the minimum path distance with the transit time, so $\beta = d_{\text{min}}$. This is the lower limit of the transit time exponent, and we confirm this prediction since we obtain $\beta \sim d_{\text{min}}$ when $Pe$ is large (Fig. 3).

The other limit at larger diffusivities—the anomalous diffusion case (1)—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 [4] and $d_w = 2.41$ [5] 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). Between these two limiting cases, we find that the transit time exponent can be approximated by

$$\beta(\text{Pe}) \sim \log(\text{Pe}). \quad (9)$$

We also perform simulations on uncorrelated percolation clusters. We find an enhanced diffusion regime and a sub-diffusion regime as well. However, due to the tortuosity of the uncorrelated percolation clusters at the threshold, the distribution of steps is not a scale-free power-law, as we find in the case of enhanced diffusion in correlated clusters Eq. (3). Thus, we conclude that the Lévy statistics found in the case of dispersion in correlated clusters 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 ballistic steps without encountering obstacles during the random walk process.

In summary, we find that, at sufficiently large Péclet numbers, there is a regime of dispersion for correlated porous media in which the trajectory of the tracer particle should be better described by a Lévy statistics Eq. (3) instead of the Gaussian behavior Eq. (1). 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 percolation-like porous materials.

[1] P. G. Saffman, J. Fluid Mech. 6, 321 (1959).
[2] See, e.g., the comprehensive review M. Sahimi, Flow and Transport in Porous Media and Fractured Rock (VCH, Boston, 1995) and extensive references therein.
[3] G. I. Taylor, Proc. R. Soc. London, Ser. A 219, 186 (1953); R. Aris, Proc. R. Soc. London, Ser. A 235, 67 (1956).
[4] A. Bunde and S. Havlin, eds., Fractals and Disordered Systems, 2nd ed. (Springer, Berlin 1996).
[5] S. Prakash, S. Havlin, M. Schwartz, and H. E. Stanley, Phys. Rev. A 46, R1724 (1992); H. A. Makse, et al., Physica A 233, 587 (1996).
[6] J. S. Andrade Jr., Phys. Rev. Lett. 79, 219 (1997).
[7] H. A. Makse, S. Havlin, M. Schwartz, and H. E. Stanley, Phys. Rev. E 54, 3129 (1996).
[8] J. S. Andrade Jr., et al., Phys. Rev. Lett. 79, 3901 (1997).
[9] H. E. Stanley, J. Phys. A 10, L211 (1977); A. Coniglio, Phys. Rev. Lett. 46, 250 (1981).
[10] S. Havlin and D. Ben-Avraham, Adv. Phys. 36, 695 (1987).
[11] M. F. Shlesinger, G. Zaslavsky, and U. Frisch, eds., Levy Flights and Related Topics in Physics (Springer, Berlin, 1995); J. Klafter, A. Blumen, and M. F. Shlesinger, Phys. Rev. A 35, 3081 (1987); J.-P. Bouchaud and A. Georges, Phys. Rep. 195, 127 (1990).
[12] Notice that the value $d_{\text{min}} \approx 1.025$ corresponds to the correlated percolation value [4], 10% smaller than the value $d_{\text{min}} \approx 1.135$ for the uncorrelated percolation problem [4].