Hydrodynamic Dispersion in Network Models of Porous Media

Lucilla de Arcangelis,[1] Joel Koplik,[2] Sidney Redner,[1,2] and David Wilkinson[2]
[1] *Center for Polymer Studies and Department of Physics, Boston University, Boston, Massachusetts 02215*
[2] *Schlumberger-Doll Research, Ridgefield, Connecticut 06877*
(Received 3 February 1986)

We consider the longitudinal dispersion of dynamically neutral tracer placed in a fluid flowing within a porous medium. For a random tube network, we derive the exact rules for tracer motion under the combined action of molecular diffusion and convection, and we introduce an efficient "probability propagation" algorithm which permits an (in principle) exact calculation of the first-passage-time distribution of the tracer as it flows through the medium. With our formalism, we exhibit both linear and nonlinear dispersion phenomena in two-dimensional random networks.

PACS numbers: 47.55.Mh, 05.40.+j, 05.60.+w

When a localized pulse of dynamically neutral tracer is released in fluid flowing in a porous medium, it disperses under the combined action of molecular diffusion and convection. Dispersion has great practical relevance since this ubiquitous phenomenon plays an important role in numerous applied fields such as oil recovery, ground water hydrology, chemical processing, and chromatography.[1] In this Letter, we develop a new methodology to understand the subtle interplay of diffusive and convective effects for dispersion phenomena in random-network models of porous media. Our method provides an (in principle) exact calculation of dispersion which overcomes the inadequacies of previous numerical treatments.

The conventional macroscopic description[1-7] of dispersion is provided by the convection-diffusion equation (CDE)

$$\frac{\partial C}{\partial t} + U\frac{\partial C}{\partial x} = D_{\parallel}\frac{\partial^2 C}{\partial x^2} + D_{\perp}\nabla_{\perp}^2 C, \tag{1}$$

where the average fluid velocity U is in the x direction, and the longitudinal and transverse dispersivities $D_{\parallel}$ and $D_{\perp}$ depend on both U and the geometry of the porous medium. The qualitative properties of $D_{\parallel}$ are particularly interesting: As $U \to 0$, molecular diffusion dominates, and $D_{\parallel}$ tends to a constant—the molecular diffusion coefficient, D_m, reduced by a factor reflecting the confined geometry of the porous medium. For high velocities, $D_{\parallel}$ is controlled by mechanical mixing: Different tracer particles are convected along different fluid streamlines, each characterized by a distinct transit time across the system. With respect to the average flow, the tracer therefore performs a random walk between pores, since sometimes the tracer is moving faster than the average flow, and sometimes slower. The characteristic time τ for a single random-walk step is of the order of l/U, the typical time to travel between pores, where l is a typical grain size in the medium. Therefore one expects $D_{\parallel} \sim l^2/\tau \sim Ul$.

However, this simple picture is inadequate as a result of important logarithmic corrections due to regions containing very slow-moving fluid.[2,3] In a random tube network, $D_{\parallel}$ is proportional to $\langle t^2\rangle - \langle t\rangle^2$, where $\langle t^n\rangle = \int dv\, t^n(v)\rho(v)$ denotes the moments of the transit time for a single tube. Here $\rho(v)$ is the probability of the tracer velocity being equal to v, and $t(v)$ is the transit time at that velocity. In the limit of pure convection, ρ is proportional to the fluid flux and therefore to v, while $t(v) = l/v$. The range of v is from zero (for steps orthogonal to the local pressure gradient) to $O(U)$ (for steps along the average gradient). Hence $\langle t^2\rangle$ diverges logarithmically for $v \to 0$ and molecular diffusion must be introduced in order to remove this singularity.[3] The divergence is cut off when the convection time reaches the diffusion time and, in consequence, $D_{\parallel} \sim Ul\log(Ul/D_m)$.

To investigate these subtle logarithmic effects in random systems, a sensitive numerical method is required. Our approach is based on three ingredients: (i) We exploit the analogy between Poiseuille flow in a random tube network, and electrical current flow in a random resistor network. Thus the steady-state electrical currents in a random-resistor network are used to specify the flow field of the background fluid which carries the tracer. (ii) We have devised an extremely efficient "probability propagation" algorithm which is an exact calculation of the first-passage probability distribution for an ensemble of tracer particles.[8] In previous numerical work,[4] individual particles were followed through a network and first-passage statistics were recorded. In the absence of slow regions this method is adequate but in general it fails unless an astronomically large ensemble is studied. The difficulty is that particles are unlikely to enter the slow regions while it is just these inadequately sampled regions which give the dominant contribution to dispersion. (iii) As discussed above, the effects of molecular dif-

fusion *must* be included if the second and higher moments of the transit time are to be finite. Here we calculate *exactly* the first-passage probability density for tracer to traverse each tube under the combined action of convection and molecular diffusion. This provides the basic "microscopic" rule for tracer motion.

To calculate the first-passage-time probability density for the network it is both elegant and convenient to work in the Laplace-transform domain. Let $\tilde{p}_{ij}(s)$ be the Laplace transform of $p_{ij}(t)$, the first-passage probability density that a tracer at node i reaches a neighboring node j in a time t, and let $\tilde{P}(s)$ be the Laplace transform of the first-passage probability density $P(t)$ for the entire network. With the assumption that the $\tilde{p}_{ij}(s)$ are known, then $\tilde{P}(s)$ is given in terms of the $\tilde{p}_{ij}(s)$ by

$$\tilde{P}(s)=\sum_{\Gamma}\prod_{ij\in\Gamma}\tilde{p}_{ij}(s), \tag{2}$$

where the sum is over all paths Γ from the inlet I to the outlet O of the network.[9]

To compute this sum over paths efficiently, we first order the nodes of the lattice in decreasing pressure order, starting with the inlet and finishing with the outlet. At each node i, we introduce a quantity $Q_i(s)$ which is a partial sum of the form (2), over paths running from the inlet to site i. Initially $Q_I=1$ at the inlet I (for a delta-function input of tracer) and $Q_i=0$ elsewhere. We then proceed recursively through the pressure-ordered node list (starting with the inlet) propagating the quantity Q from each node i to its lattice neighbors j according to the rule

$$Q_j(s)\rightarrow Q_j(s)+Q_i(s)\tilde{p}_{ij}(s),\quad Q_i(s)\rightarrow 0. \tag{3}$$

The lowest (outlet) node is not propagated. After all the internal nodes have been propagated once in this way, the quantity $Q_0(s)$ at the outlet contains *all* the terms in (2) corresponding to purely downstream paths. If there were no molecular diffusion, all the tracer would be collected at the outlet after a single such downstream "sweep" through the network. In an $L\times L$ network, there are $O(e^{L^2})$ independent downstream paths, and our propagation method sums over all these paths in an algorithm time that is of the order of $L^2\log L$.

Because of molecular diffusion, however, the tracer motion includes upstream paths as well. In this case, after one sweep through the network, one has $Q_n\neq 0$ in general for internal nodes. By a sweep downstream through the network again and propagation of the residual probability, the contribution of all paths involving exactly one (diffusive) upstream step are included. By repeated sweeps through the network, the contribution of paths with progressively more upstream steps are included. We find that the first-passage probability [which is just $\tilde{P}(0)$] converges to unity geometrically in the number of sweeps, in that a roughly constant fraction of the residual probability is extracted in each sweep. Thus the propagation algorithm performs a rearrangement of the sum over paths in (3), and gives $\tilde{P}(s)$ *exactly,* up to geometric convergence.

To complete our calculational approach, we must specify the microscopic rule for tracer motion under the combined influence of convection and diffusion, namely the quantity $\tilde{p}_{ij}(s)$ for each node i. At the level of a single tube, the tracer concentration, $c(x,t)$, satisfies the one-dimensional CDE[10]

$$\partial c/\partial t+u\,\partial c/\partial x=D_m\,\partial^2 c/\partial x^2, \tag{4}$$

where x is the longitudinal coordinate along the tube, D_m is the molecular diffusion coefficient, and u is the average fluid velocity in the tube. Now consider a network of such tubes $\{ij\}$ of common length l, but with a distribution of cross-sectional areas S_{ij}, and focus on node i which is connected to z distinct nodes $\{j\}$. The concentrations in each tube ij, $c_{ij}(x_{ij},t)$, each satisfy (4), with initial condition $c_{ij}(x_{ij},0)=0$, and three boundary conditions: (i) a unit pulse input of flux at node i at $t=0$,

$$\sum_j S_{ij}\left\{u_{ij}c_{ij}-D_m\frac{\partial c_{ij}}{\partial x_{ij}}\right\}_{x_{ij}=0}=\delta(t);$$

(ii) a common concentration $\xi_i(t)$ at the starting junction, $c_{ij}(0,t)=\xi_i(t)$ for all J; and (iii) a sink at each tube end, $c_{ij}(l,t)=0$ for all j, corresponding to the fact that tracer reaching the end acts as a source for the junction problem at the new node. The first-passage probability is then simply the flux, $p_{ij}(t)=-S_{ij}D_m\times\partial c_{ij}(l,t)/\partial x_{ij}$.

To solve, we take the Laplace transform of (4), obtaining z second-order ODE's for the Laplace transform concentrations $\tilde{c}_{ij}(x,s)$. The general solution is

$$\begin{aligned}&\tilde{c}_{ij}(x,s)=A_{ij}e^{\alpha_{ij}x}+B_{ij}e^{\beta_{ij}x},\\&\alpha_{ij},\beta_{ij}=(1/2D_m)[u_{ij}\pm(u_{ij}^2+4D_m s)^{1/2}],\end{aligned} \tag{5}$$

where the $2z$ coefficients A_{ij} and B_{ij} may be determined from the boundary conditions (i) to (iii) above. After several steps we find

$$\begin{aligned}&\tilde{p}_{ij}(s)=\tilde{\xi}_i(s)S_{ij}\frac{\alpha_{ij}-\beta_{ij}}{e^{-\beta_{ik}l}-e^{-\alpha_{ik}l}},\\&\frac{1}{\tilde{\xi}_i(s)}=\sum_k S_{ik}\frac{\alpha_{ik}e^{-\beta_{ik}l}-\beta_{ik}e^{-\alpha_{ik}l}}{e^{-\beta_{ik}l}-e^{-\alpha_{ik}l}}.\end{aligned} \tag{6}$$

The limiting behavior of the rather untransparent expressions in (6) is familiar, in the diffusive limit ($u_{ij}\rightarrow 0$ for all j) we find that the probability to exit via tube ij and the average time required for this event

[obtained by differentiating $\tilde{p}_{ij}(s)$] are $\tilde{p}_{ij}(0) = S_{ij}/\sum_k S_{ik}$ and $\langle t_{ij}\rangle = l^2/2D_m$, while for the opposite convective limit (all $|u_{ij}| \to \infty$) we have, for u_{ij} positive, $\tilde{p}_{ij}(0) = u_{ij}S_{ij}/(\sum_k u_{ik}S_{ik})$ and $\langle t_{ij}\rangle = l/u_{ij}$, where the sum is over the outward-flowing tubes. For the inward-flowing tubes (u_{ij} negative) the corresponding exit probability vanishes exponentially as $|u_{ij}| \to \infty$.

It is important to realize that the quantities $\tilde{p}_{ij}(s)$ in (6), together with (2), provide the *exact* solution to the set of tube-level convective diffusion equations for the whole lattice. This is because the $\tilde{p}_{ij}(s)$ yield not only the exact exit probability and mean transit time for each link, but also the effects of longitudinal dispersion within each link exactly. Thus our approach gives the exact probability distribution for a random-walk model in which there is a different time for each "step," and where each "step" represents the continuum solution of the traversal time problem for each link. From this viewpoint our method should have wide applicability to transport in random media.[11]

To demonstrate the power of these methods we investigate dispersion in a square lattice of tubes with cross-sectional areas uniformly distributed in the interval $[1-\frac{1}{2}w, 1+\frac{1}{2}w]$, with w representing the degree of disorder in the system. Two sides of the lattice are set at constant pressure and the other two are connected by periodic boundary conditions. The lattice axes are oriented at 45° with respect to the average flow direction. The constant-pressure lattice edges are identified as inlet and outlet nodes for tracer, and we use probability propagation and the exact expression for $\tilde{p}_{ij}(s)$ to compute $\tilde{P}(s)$, and thereby obtain moments of the transit time distribution.

We analyze the data by matching the standard deviation $\sigma_T^2 \equiv \langle t^2\rangle - \langle t\rangle^2$ found in the simulation with the corresponding quantity obtained by treating the entire network macroscopically. The standard deviation can be obtained directly from $\tilde{P}(s)$ via $\langle t^n\rangle = (-\partial/\partial s)^n \tilde{P}(s)|_{s=0}$, and these derivatives may be approximated numerically in terms of $\tilde{P}(s)$ for discrete values of s near $s=0$. At the macroscopic level, we assume that the system satisfies the CDE (1) with boundary conditions of unit input at the inlet, and a "sink" at the outlet. We solve this equation by the same method used to obtain (6), thus obtaining

$$\tilde{P}(s) = \frac{M_s e^M}{M_s \cosh M_s + M \sinh M_s}, \tag{7}$$

where $M_s \equiv (M^2 + sL^2/D_{\parallel})^{1/2}$ and M is the macroscopic Peclet number $M = UL/2D_{\parallel}$. This represents the reduction of expressions (6) to the case of a single bond. Then the standard deviation of the transit time is obtained by differentiation of this expression, yielding

$$\sigma_T^2 = (L/U)^2 f(M), \tag{8}$$

where L is the system length, and

$$f(M) = M^{-2}[M - \tfrac{5}{4} + (2M+1)e^{-2M} + \tfrac{1}{4}e^{-4M}]. \tag{9}$$

From our computed value of σ_T^2, we obtain $D_{\parallel}$ by a graphical solution of (8), as indicated in Fig. 1. Notice that only in the limit $M \to \infty$ (pure convection), where $f(M) \to 1/M$, do we recover the widely quoted[4] simple relation $D_{\parallel} = (U^3/2L)\sigma_T^2$. In general, the right-hand side of (8) is nonmonotonic, so that the extraction of $D_{\parallel}$ from numerical data requires considerable care. As seen in Fig. 1, for any value of U there are in general two possible values of M; the correct one is determined by the requirements that the ob-

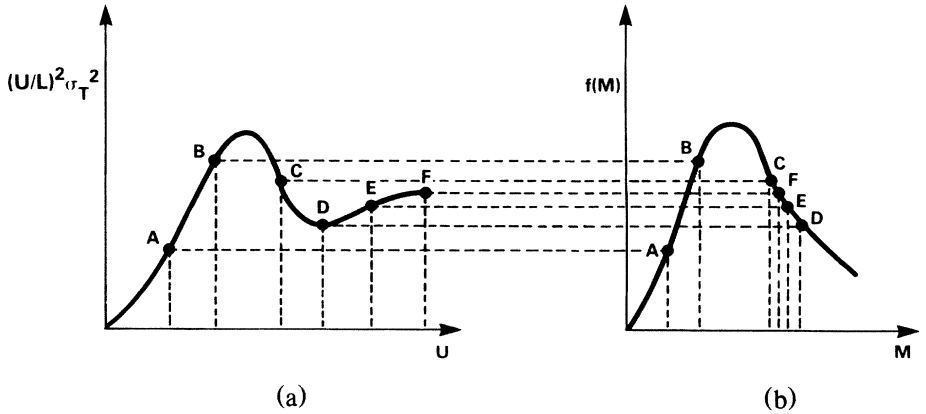

FIG. 1. Schematic numerical data for $(U/L)^2\sigma_T^2$ as a function of U together with a plot of $f(M)$ of Eq. (8) vs M. Each datum point is matched with a point on the $f(M)$ curve as indicated by the letters. From the value of M so obtained, a curve of $D_{\parallel}$ as a function of U is inferred. At high velocity, $(U/L)^2\sigma_T^2$ shows a second increase as a function of U, a behavior which corresponds to $D_{\parallel}$ increasing faster than linearly with U.

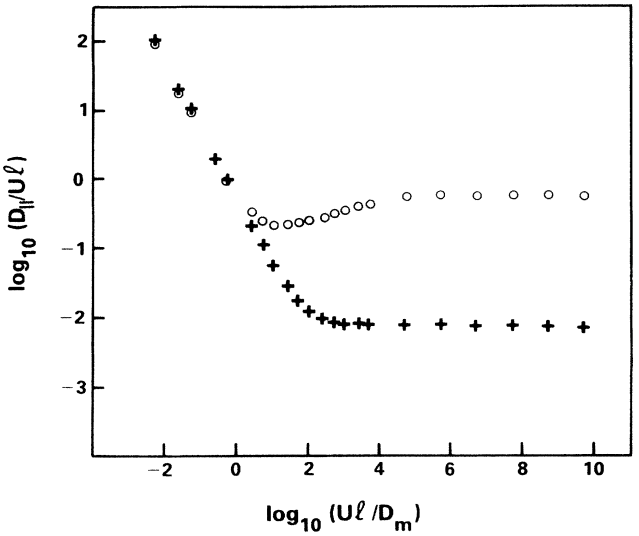

FIG. 2. Comparison of the velocity dependence of the macroscopic dispersion coefficient, $D_{\|}$, for a 10×10 network with $w=0.5$ (plusses) and 1.5 (circles). Plotted is $D_{\|}/Ul$ vs the (microscopic) Peclet number Ul/D_m. The data represent averages over 1000 and 2000 configurations, respectively. The latter case gives rise to a regime of nonlinear velocity dependence where $D_{\|}/Ul$ is increasing.

tained value of M is a smooth function of U, and that $M\to 0$ as $U\to 0$. The consistency of this picture requires that $(U/L)^2\sigma_T^2$ is nonmonotonic and that its maximum value matches that of $f(M)$. That this indeed happens in our simulations provides strong evidence that the system is well described by the CDE (1), with a velocity-dependent macroscopic dispersion coefficient $D_{\|}$.

In Fig. 2 we show simulation results on 10×10 lattices with two different values of the disorder parameter w. For small disorder ($w=0.5$), there is a well-defined value for the ensemble average of the "slowest" bond in the network. Consequently, there is a sharp transition from a low-U regime, where all the bonds are diffusive and $D_{\|}$ is U-independent, to a high-U regime, where all the bonds are convective and $D_{\|}$ varies linearly with U. However, for larger disorder ($w=1.5$), an infinite ensemble of systems will contain arbitrarily slow bonds. In this case, the dispersion at high velocity will be dominated by the small fraction of tracer which spends time in these slow bonds, while the majority of the tracer is convected through the rest of the network. In this regime, we find that $(U/L)^2\sigma_T^2$ exhibits a second increase for increasing U, as illustrated schematically in Fig. 1(a). This means that the corresponding point in Fig. 1(b) retraces back along the $f(M)$ curve, corresponding to $D_{\|}$ increasing faster than linearly in U (Fig. 2). At very high velocities, all the bonds are convective, because for any finite ensemble there is a lower limit to the slowness of the bonds, and therefore $D_{\|}$ is exactly proportional to the velocity.

In summary, we have developed a new approach which provides an exact description of dispersion and other transport phenomena in disordered media. We have derived the exact rules for tracer motion in random tube networks under the combined action of molecular diffusion and convection, and combined this approach with probability propagation to yield a numerical method which is both very powerful and accurate. We are able to compute the macroscopic behavior of $D_{\|}$ over the whole velocity range. Furthermore, for strongly disordered networks, we have found a regime of faster than linear velocity dependence of $D_{\|}$ which is indicative of the logarithmic behavior predicted by Saffman. We are now applying our method to dispersion in poorly connected media, in order to elucidate the very interesting effects due to stagnant regions and large-scale heterogeneities.

We thank H. T. Davis, E. Guyon, and M. Sahimi for informative discussions and S. Roux for correcting an earlier typographical error in the manuscript.

[1]J. J. Fried and M. A. Combarnous, Adv. Hydrosci. **7**, 169 (1971); J. Bear, *Dynamics of Fluids in Porous Media* (Elsevier, Amsterdam, 1971); A. E. Scheidegger, *The Physics of Flow in Porous Media* (Univ. of Toronto Press, Toronto, 1974).

[2]G. de Josselin de Jong, Trans. Am. Geophys. Union **39**, 67 (1958).

[3]P. G. Saffman, J. Fluid Mech. **6**, 321 (1959), and **7**, 194 (1960).

[4]M. Sahimi, H. T. Davis, and L. E. Scriven, Chem. Eng. Commun. **23**, 329 (1983); M. Sahimi, B. D. Hughes, A. L. Heiba, H. T. Davis, and L. E. Scriven, Chem. Eng. Sci. (to be published).

[5]J. A. Aronovitz and D. R. Nelson, Phys. Rev. A **30**, 1948 (1984).

[6]J.-C. Bacri and D. Salin, J. Phys. (Paris), Lett. **45**, L767 (1984).

[7]D. L. Koch and J. F. Brady, J. Fluid Mech. **154**, 399 (1985).

[8]For probability propagation in the context of random walks see, e.g., D. Ben-Avraham and S. Havlin, J. Phys. A **15**, L691 (1982).

[9]This exact sum was apparently first discussed in M. Leitzelement, P. Maj, J. A. Dodds, and J. L. Greffe, in *Solid-Liquid Separation,* edited by J. Gregory (Ellis Horwood, Chichester, England, 1984).

[10]For given U and l, our treatment is exact when the radius a for each tube satisfies $a/l \ll 1$ and $a/l \ll D_m/Ul$. The second condition may be relaxed to $a/l \ll (D_m/Ul)^{1/2}$ if we replace D_m by the Taylor-Aris effective dispersivity (Ref. 1) $D_m + U^2a^2/48D_m$.

[11]As an example we could easily simulate the random-walk model of a two-component composite, where there are two possible values for the time step; see D. C. Hong, H. E. Stanley, A. Coniglio and A. Bunde, Phys. Rev. B **33**, 4564 (1986).