

Walker diffusion method for calculation of transport properties of finite composite systems

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(Received 2 August 2001; published 25 January 2002)

A heterogeneous medium may be represented by a scalar field of local transport coefficients (e.g., conductivity) or by a “resistor network” derived from that scalar field. In either case the *effective* (macroscopic) and *local* (microscopic) transport properties may be calculated by the walker diffusion method. Some sample calculations for disordered systems are presented to demonstrate the method.

DOI: 10.1103/PhysRevE.65.026144

PACS number(s): 05.10.-a, 02.70.Rr, 05.60.Cd

I. INTRODUCTION

The transport properties of porous and fractured media determine, for example, the rapid migration of chemical contaminants through fractured rock, and the displacement of oil in porous rock by injected brine. Such environmental and industrial applications motivate the many fundamental studies of transport in heterogeneous media (see the comprehensive review by Sahimi [1]).

The electrical properties are governed by the set of transport equations,

$$\begin{aligned}\nabla \times \mathbf{E}(\mathbf{r}) &= \mathbf{0}, & \nabla \cdot \mathbf{J}(\mathbf{r}) &= 0, & \mathbf{J}(\mathbf{r}) &= \sigma(\mathbf{r})\mathbf{E}(\mathbf{r}), \\ \mathbf{E}(\mathbf{r}) &= -\nabla \phi(\mathbf{r}),\end{aligned}\quad (1)$$

which relate the electric field \mathbf{E} and the current density \mathbf{J} at the point \mathbf{r} . The heterogeneity of the medium is expressed through the local conductivity $\sigma(\mathbf{r})$. This same set of equations, with appropriate vector and scalar fields, enables calculation of the thermal, dielectric, and diffusivity properties as well; again, the local transport coefficients in each case reflect the heterogeneity of the medium. The electrical analog is often extended to fluid permeability: a *local* version of Darcy’s law for fluid flow is

$$\mathbf{Q}(\mathbf{r}) = -[k(\mathbf{r})/\mu]\nabla P(\mathbf{r}),\quad (2)$$

where $\mathbf{Q}(\mathbf{r})$ is the volumetric flow rate, $P(\mathbf{r})$ is the fluid pressure, $k(\mathbf{r})$ is the permeability of the saturated medium, and μ is the viscosity of the fluid. The incompressibility of the fluid produces the additional equation $\nabla \cdot \mathbf{Q}(\mathbf{r}) = 0$.

Solution of these equations for given boundary conditions is typically accomplished by the finite difference method (FDM). This approach is equivalent to solving Kirchhoff’s laws for a resistor network, and indeed porous and fractured media are often explicitly modeled as resistor networks. For example, Bernabé [2] assigns hydraulic and electric conductances to the bonds of a network, where those conductances are derived from specified distributions of pore size and crack aperture and lateral extension. More generally, a heterogeneous medium is represented by a scalar field [comprised of the $\sigma(\mathbf{r})$ or $k(\mathbf{r})$, for example] which is effectively converted to a regular resistor network to permit FDM cal-

culations (Romeu and Noetinger [3] analyze various choices for the internodal transmissivities needed for the conversion).

Van Sicle [4] has recently introduced the walker diffusion method (WDM) for calculation of the effective transport properties of scalar fields. In contrast to the FDM, the WDM works directly with the scalar field, and does not require boundary conditions in order to determine the effective properties of the medium. The WDM also obtains the transport property correlation length ξ (the length scale above which the medium is effectively homogeneous with respect to the transport property of interest, and below which the medium is heterogeneous) and the transport property scaling law (which is relevant when the system size is less than ξ) [5]. This stochastic method has recently been applied to a study of the electric and hydraulic properties of single fractures, where a fracture is represented by a field of local transport coefficients that are functions of the local aperture value [6].

To broaden its utility, the WDM is further developed below to treat finite scalar fields (i.e., those having nonperiodic boundary conditions), and finite and infinite resistor networks. Thus the WDM enables studies of the effects of imposed boundary conditions on transport properties of heterogeneous media, and allows direct comparison of calculated properties for media represented by scalar fields and by regular resistor networks, respectively.

II. WDM FOR SCALAR FIELDS

The walker diffusion method [4] exploits the isomorphism between the transport equations and the diffusion equation for a collection of noninteracting random walkers in the presence of a driving force. The phase domains in a composite microstructure correspond to distinct populations of walkers, where the equilibrium walker density $\rho^0(\mathbf{r})$ of a population is given by the value of the transport coefficient $\sigma(\mathbf{r})$ of the corresponding phase domain. The principle of detailed balance ensures that the population densities are maintained, and provides the following rule for walker diffusion over a scalar field (or digitized microstructure): a walker at site (or pixel) i attempts a move to a randomly chosen adjacent site j during the time interval $\tau = (4d)^{-1}$, where d is the Euclidean dimension of the space; this move is successful with probability $p_{ij} = \sigma_j / (\sigma_i + \sigma_j)$, where σ_i and σ_j are the transport coefficients for the phases comprising sites i and j , respectively. The path of a walker thus reflects the phase composition (population density) and morphology of the domains

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that are encountered, and may be described by a diffusion coefficient D_w that is related to the effective (macroscopic) transport coefficient σ by

$$\sigma = \langle \sigma(\mathbf{r}) \rangle D_w, \quad (3)$$

where $\langle \sigma(\mathbf{r}) \rangle$ is the volume average of the constituent transport coefficients. The diffusion coefficient D_w is calculated from the equation

$$D_w = \langle R^2 \rangle / (2dt), \quad (4)$$

where the set $\{R\}$ of walker displacements, each occurring over the time interval t , comprises a Gaussian distribution that must necessarily be centered well beyond ξ (the walker diffusion is otherwise anomalous [5]). In practice it is convenient to eliminate the unsuccessful moves inherent in the rule stated above by use of the variable residence time algorithm [4].

Remarkably, the effective transport coefficient is obtained without solving the set of transport equations (1) or imposing boundary conditions. The WDM was verified by reproducing some exact results from percolation theory [4].

The WDM can additionally be used to *solve* the set of transport equations (1); that is, to calculate the vector fields and the potential field $\phi(\mathbf{r})$ for given boundary conditions. The concept of walker populations (representing phase domains in a composite) together with the walker diffusion rule is sufficient to *derive* the flux equation

$$\mathbf{J}(\mathbf{r}) = -\sigma(\mathbf{r}) \nabla \phi(\mathbf{r}), \quad (5)$$

where the $\phi(\mathbf{r})$ are found to be simple functions of the walker densities.

At equilibrium (i.e., in the absence of a driving force), a single diffusing walker will “populate,” or occupy, the sites of a composite in proportion to the corresponding transport coefficients $\sigma(\mathbf{r})$; thus, in the limit of infinite time, the *equilibrium* walker density at site i is given by

$$\rho_i^0 = \sigma_i = (t_i / \langle t_k \rangle) \langle \sigma_k \rangle, \quad (6)$$

where the ratio is the fraction of time spent at site i and the averages implied by the angle brackets are taken over all sites k .

The walker densities are altered when a chemical potential gradient is created by injecting walkers into the system at one boundary or point, and removing them at another. The *steady-state* walker density at site i is then

$$\rho_i = (t_i / \langle t_k \rangle) \langle \sigma_k \rangle, \quad (7)$$

where again the ratio is the fraction of time spent by the walkers at site i [this is of course different from the ratio in Eq. (6)].

The densities ρ_i^0 and ρ_i and the chemical potential μ_i are related by the principle of detailed balance

$$\rho_i p_{i \rightarrow j} = \rho_j p_{j \rightarrow i} \exp[-(\mu_j - \mu_i) / kT] \quad (8)$$

and

$$\rho_i^0 p_{i \rightarrow j} = \rho_j^0 p_{j \rightarrow i}, \quad (9)$$

where $p_{i \rightarrow j} = p_{ij} / (2d)$ is the probability that a selected walker at site i will move to the adjacent site j over the next time interval τ (the denominator $2d$ is specific to orthogonal lattices, e.g., square and cubic lattices). Together these equations produce the relation

$$\rho_i / \rho_i^0 = \exp[\mu_i / kT]. \quad (10)$$

Note that Eq. (10) is equivalent to the expression

$$\mu(\mathbf{r}) = kT \ln[\rho(\mathbf{r}) / \rho^0(\mathbf{r})], \quad (11)$$

so that the usual relation between the chemical potential gradient and a particle concentration gradient in a homogeneous system [$\rho^0(\mathbf{r}) = \text{constant}$] is recovered:

$$\nabla \mu(\mathbf{r}) = [kT / \rho(\mathbf{r})] \nabla \rho(\mathbf{r}). \quad (12)$$

Equation (5) may be obtained from the walker flux equation

$$J_{i \rightarrow j} \tau = \rho_i p_{i \rightarrow j} - \rho_j p_{j \rightarrow i} \quad (13)$$

which, after some algebra, produces

$$J_{i \rightarrow j} = 2(1/\rho_i^0 + 1/\rho_j^0)^{-1} (\exp[\mu_i / kT] - \exp[\mu_j / kT]). \quad (14)$$

This suggests the particular identification $\phi_i = \exp[\mu_i / kT]$, so that the potential at site i is

$$\phi_i = \rho_i / \rho_i^0 = (t_i / \langle t_k \rangle) \langle \sigma_k \rangle / \sigma_i, \quad (15)$$

where again the ratio $t_i / \langle t_k \rangle$ is the fraction of time spent by the walkers at site i . Equation (14) for the walker flux then becomes

$$J_{i \rightarrow j} = 2(1/\rho_i^0 + 1/\rho_j^0)^{-1} (\phi_i - \phi_j). \quad (16)$$

The form of this last equation indicates that the WDM for *finite* scalar fields provides a random walk solution to the set of FDM algebraic equations.

A practical issue is the implementation of boundary conditions. Clearly the potential is zero where the walkers are absorbed since ρ_i equals zero there. Relative potential differences are then established by appropriate placement and “strength” of walker sources. As an example, consider the two-dimensional, multiphase system shown in Fig. 1(a). Equipotential surfaces may be established at the left and right edges by viewing the system as an $R \times C$ grid or matrix. Then a column $C+1$ is added that is identical to column C ; when a walker reaches column $C+1$ it is immediately removed (so that column $C+1$ is an equipotential surface with all $\phi_i = 0$). To induce correct walker behavior near the R sources in column 1, a column 0 is added that is identical to column 2. If a walker placed in column 1 moves to column 0 on its *first* move, it is removed and its time spent in column 1 is disregarded; otherwise all moves from a site i contribute to the residence time t_i . If the walker diffuses in column 1 but eventually makes a move to column 0, it is removed at

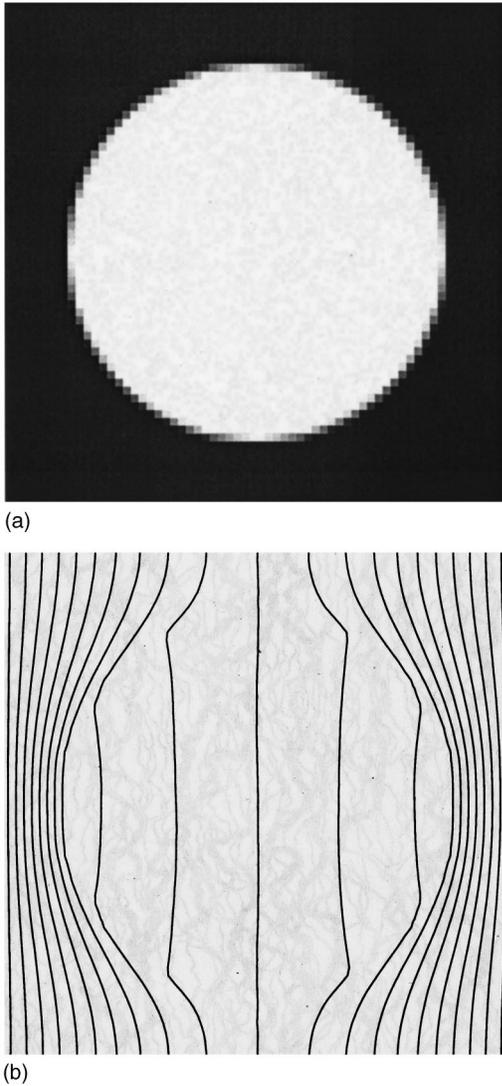


FIG. 1. (a) Two-dimensional, multiphase composite approximating a continuous system comprised of a circular region of conductivity 1.0 (white) centered in a square of conductivity 0.1 (black). The interfacial phases have intermediate conductivity values as indicated by the shade of gray. (b) Equipotential lines calculated for a potential difference applied across the composite.

that time. If the walker diffuses beyond column 1, it is immediately removed when it enters column $C+1$ or returns to column 1 (not column 0). A new walker is placed in column 1 at the site that currently has the *smallest* potential (i.e., smallest t_i/σ_i), thus ensuring (in the limit of infinite time) that column 1 is an equipotential surface. This procedure gives a uniform potential gradient in the case of a two-dimensional homogeneous system.

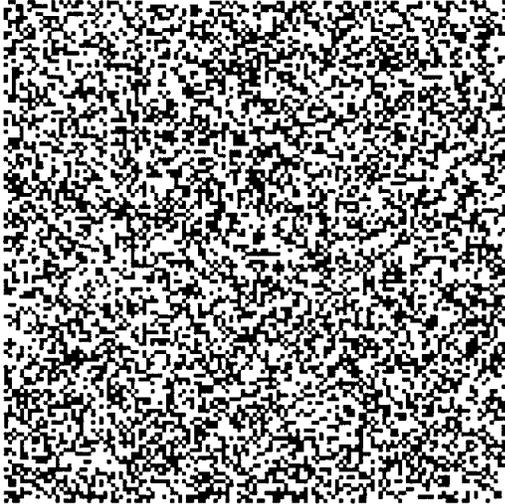
The discrete system in Fig. 1(a) was designed to approximate a continuous system comprised of a circular region of radius $\frac{3}{8}$ and conductivity 1.0 centered in a unit square of conductivity 0.1. Perrins, McKenzie, and McPhedran [7] provide a truncated series expression for the transverse conductivity of an infinite square array of cylinders that gives 0.214 619 for this continuous, two-dimensional system. Figure 1(a) was obtained by superposing a circle of radius 24 on

a 64×64 square region, then setting the conductivity of each site i intersected by the circle to the areal average value $\sigma_i = f + 0.1(1-f)$, where f is the fraction of the site contained within the circle. The electrical properties of the discrete system were obtained in the manner described above (equipotential surfaces at the left and right edges; periodic boundary conditions at the top and bottom edges) using 10^7 walkers. Figure 1(b) shows calculated equipotential surfaces (lines), the density of lines reflecting the magnitude of the local potential gradient. The calculated effective conductivity $\sigma = \langle J_{i \rightarrow j} \rangle / \Delta \phi = 0.2186$ ($\Delta \phi$ is the potential drop across the system); a somewhat smaller value would result from using the harmonic average conductivity $\sigma_i = [f + (1-f)/0.1]^{-1}$ or the geometric average conductivity $\sigma_i = (0.1)^{1-f}$ for those sites intersected by the circle. (And of course the calculated σ will approach the exact value in any case as the resolution of the system is increased from 64×64 .) This value for σ should be compared with that calculated for the discrete system with periodic boundary conditions on *all* sides; the latter value is 0.2196 ± 0.0006 obtained by calculating the diffusion coefficient D_w for a single walker diffusing over the infinite system (the uncertainty in σ may be reduced by increasing the number of walks). Note that the equipotential boundaries of the finite system coincide with the periodic boundaries of the infinite system. Thus the difference in calculated σ values is entirely due to the fact that the scalar conductivity field shown in Fig. 1(a) is converted to a conductor network when nonperiodic boundary conditions are applied [as evident from Eq. (16)].

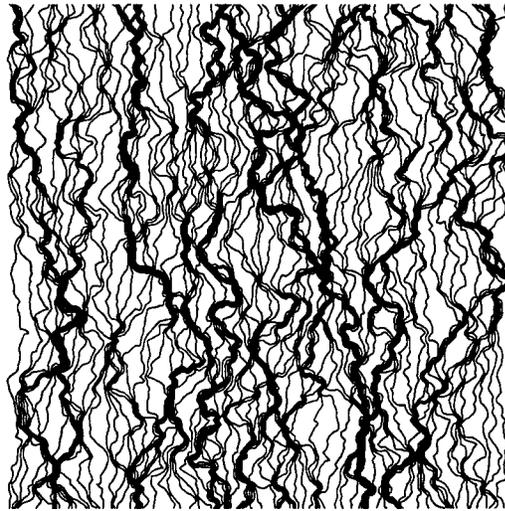
Similar calculations were made for the two-dimensional, two-phase random system shown in Fig. 2(a). The fraction of white sites is at the percolation threshold (0.592 75); those sites have the higher conductivity $\sigma_A = 1.0$ while the black sites have the lower conductivity $\sigma_B = 0.01$. The effective conductivity σ of an infinite system with these properties is exactly $(\sigma_A \sigma_B)^{1/2} = 0.1$ [8]. The 128×128 system in Fig. 2(a) should have a conductivity very close to this, as the conductivity correlation length for this choice of σ_A and σ_B ($\xi \approx 15$, Ref. [5]) is much smaller than the system length (128). Indeed, the WDM gives $\sigma = 0.0977$ when the left and right edges of the finite system are made to be equipotential surfaces by releasing and absorbing 5×10^6 walkers there. Figure 2(b) shows equipotential surfaces (lines) in the interior of the random system, their tortuosity reflecting the heterogeneity of the scalar field. In the case of fluid flow through a permeable medium, the large spatial variation in the fluid pressure gradient ensures channeling of the flow, tortuous streamlines, and a broad distribution of solute transit times.

III. WDM FOR RESISTOR NETWORKS

The isomorphism between Ohm's law for the electrical current density and the diffusion equation for a collection of noninteracting walkers in the presence of a driving force $-\nabla \phi(\mathbf{r})$ can again be invoked to develop the walker diffusion method for resistor (or conductor) *networks*. The diffusion current density (walker flux) is



(a)



(b)

FIG. 2. (a) Two-dimensional, two-phase disordered composite for which the ratio of the two conductivities is 100:1. (b) Equipotential lines calculated for a potential difference applied across the composite.

$$\mathbf{J}(\mathbf{r}) = -D(\mathbf{r})\rho(\mathbf{r})\nabla\phi(\mathbf{r}), \quad (17)$$

where $D(\mathbf{r})$ and $\rho(\mathbf{r})$ are the local walker diffusion coefficient and local walker density, respectively.

According to Ohm's law, the electrical current density $J_{i \rightarrow j}$ between adjacent network nodes i and j due to the potential difference $\phi_j - \phi_i$ is

$$J_{i \rightarrow j} = -\sigma_{ij}(\phi_j - \phi_i)/r_{ij}, \quad (18)$$

where σ_{ij} is the conductivity of the bond connecting the nodes and $r_{ij} = 1$ is the bond length (this derivation is specialized to *regular* networks; that is, networks with regularly spaced nodes). Then the local walker diffusion coefficient is identified with the conductivity such that $D_{ij} = \sigma_{ij}$, and the equilibrium walker density $\rho_i^0 = 1$ at all nodes i in the net-

work. In analogy with this local relation, the effective (macroscopic) transport coefficient σ for the regular network is given by

$$\sigma = D_w, \quad (19)$$

where D_w is the macroscopic walker diffusion coefficient.

The walker behavior again follows from the principle of detailed balance. A walker at site i attempts a move in a randomly chosen direction; that attempt (to adjacent site j) is successful with probability p_{ij} . Equation (9) gives $p_{i \rightarrow j} = p_{j \rightarrow i}$, which implies $p_{i \rightarrow j} \propto \sigma_{ij}$ and thus that $p_{ij} \propto \sigma_{ij}$. Consider now a walker diffusing over a regular conductor network with all $\sigma_{ij} = \sigma_{\max}$. Then every attempted move is successful ($p_{ij} = 1$) and the time τ associated with a move is

$$\tau = \lambda^2 / (2dD_w) = (2d\sigma_{\max})^{-1}, \quad (20)$$

where the bond length $\lambda = 1$ for convenience.

More generally, a walker at site i attempts a move to a randomly chosen adjacent site j during the time interval $\tau = (2d\sigma_{\max})^{-1}$; that move is successful with probability $p_{ij} = \sigma_{ij} / \sigma_{\max}$, where σ_{\max} is the network conductor with largest conductivity value. The walker diffusion coefficient D_w is then calculated from the total displacement of the walker over the time corresponding to the number of attempted moves [Eq. (4)].

This diffusion procedure is computationally inefficient, since not all attempted moves by the walker are successful. This is overcome by statistically weighing the behavior of the walker such that every attempt is successful but the move is accomplished over a variable time interval. Thus the actual behavior of the walker is well approximated by a sequence of moves in which the direction of each move from a site i is determined randomly by the set of probabilities $\{P_{ij}\}$, where

$$P_{ij} = \frac{p_{i \rightarrow j}}{\sum_k p_{i \rightarrow k}} = \frac{\sigma_{ij}}{\sum_k \sigma_{ik}} \quad (21)$$

and the time interval over which the move occurs is

$$T_i = \frac{\tau}{\sum_k p_{i \rightarrow k}} = \frac{1}{\sum_k \sigma_{ik}} \quad (22)$$

[the last equality in Eq. (22) is specific to orthogonal networks, e.g., square and cubic networks]. This variable residence time algorithm was verified analytically (by use of Markov chain theory [9]) for one-dimensional conductor networks.

A further check is made by calculating the effective conductivity of square random networks where half the bonds have conductivity σ_A and half have conductivity σ_B (so each phase is at the bond percolation threshold). These results, for ratios $\sigma_B / \sigma_A = 0.5, 0.1, 0.01,$ and 0.001 , are presented in Fig. 3. Each data point is the average value obtained from 15 different networks of size 512×512 (with periodic boundary

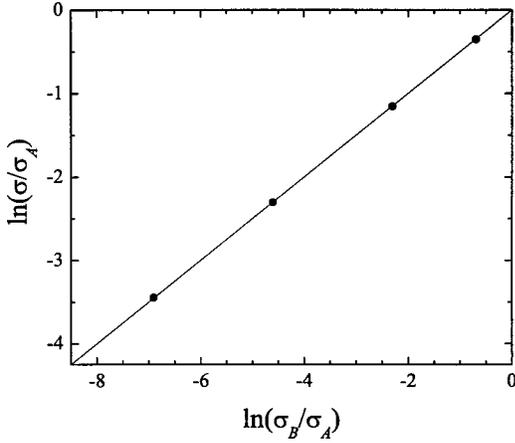


FIG. 3. Calculated values of the conductivity ratio σ/σ_A for square random networks where half the bonds have conductivity σ_A and half have conductivity σ_B . The four data points are taken for $\sigma_B/\sigma_A=0.5, 0.1, 0.01, \text{ and } 0.001$.

conditions); standard deviations about each average are much smaller than the size of the plotted point. The points all fall on the straight line of slope 0.5, in agreement with the analytical relation $\sigma=(\sigma_A\sigma_B)^{1/2}$ [8,10].

The effective conductivity was also calculated for square random networks where a fraction f_A of the bonds have conductivity σ_A and the rest are nonconducting. These results, for $f_A=0.55, 0.6, 0.65, 0.7, \text{ and } 0.8$, are presented in Fig. 4. Each data point is the average value obtained from 15 different networks of size 1024×1024 (with periodic boundary conditions); standard deviations about each average are smaller than the size of the plotted point. The equation for the *curve* fit through these points (and passing through the origin) is

$$\sigma/\sigma_A=(2f_A-1)^{\mu+v \ln(2f_A-1)} \quad (23)$$

with $\mu=1.02771\pm 0.00196$ (standard error) and $v=-0.02988\pm 0.00103$. Thus the conductivity exponent for

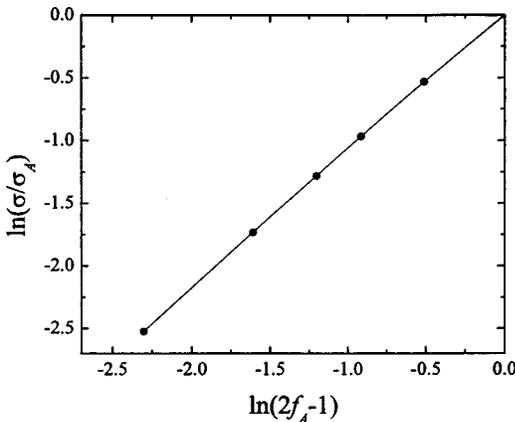


FIG. 4. Calculated values of the conductivity ratio σ/σ_A for square random networks where a fraction f_A of the bonds have conductivity σ_A and the rest are insulating. The five data points (from the right) are taken for $f_A=0.8, 0.7, 0.65, 0.6, \text{ and } 0.55$.

two-dimensional bond percolation is *greater* than 1.096 (since the slope of the curve in Fig. 4 is increasing as the percolation threshold is approached from above), which is consistent with exponent values obtained by, e.g., Straley [8] (1.10 ± 0.05), Stinchcombe and Watson [11] (1.13 ± 0.09), Fisch and Harris [12] (1.43 ± 0.02), Ben-Mizrachi and Bergman [13] (1.00), Derrida and Vannimenus [14] (1.28 ± 0.03), Sahimi *et al.* [15] (1.264 ± 0.054), and Grassberger [16] (1.3100 ± 0.0011). It is interesting to note that the conductivity exponent obtained by the WDM for two-dimensional *site* percolation is 1.253 ± 0.005 [4].

The transport properties of *finite* (nonperiodic) resistor networks can be found in a manner similar to that described in the preceding section for finite scalar fields. A chemical potential gradient is created by injecting walkers at selected nodes and removing them at other nodes (internal as well as boundary nodes can be walker sources and sinks). Then the *steady-state* walker density at node i is the fraction of time spent by the walkers at node i ,

$$\rho_i=t_i/\langle t_k \rangle \quad (24)$$

(in the absence of a chemical potential, $\rho_i=\rho_i^0=1$). Otherwise the treatment for resistor networks follows that for scalar fields; in particular, Eqs. (8)–(13) hold here as well. The counterpart to Eq. (14) is

$$J_{i\rightarrow j}=\sigma_{ij}(\exp[\mu_i/kT]-\exp[\mu_j/kT]). \quad (25)$$

This suggests the particular identification $\phi_i=\exp[\mu_i/kT]$, so that the potential at node i is

$$\phi_i=\rho_i=t_i/\langle t_k \rangle \quad (26)$$

and Eq. (25) for the walker flux becomes

$$J_{i\rightarrow j}=\sigma_{ij}(\phi_i-\phi_j). \quad (27)$$

This last equation is identical to Eq. (18), which indicates that the WDM for *finite* resistor networks provides a random walk solution to the set of FDM algebraic equations.

The WDM may be straightforwardly applied to *irregular* resistor networks as well, as those can be converted to (possibly higher-dimensional) regular networks for which the walker diffusion rule is given above. Actual conversion of an irregular network is not necessary, however, when the variable residence time algorithm [Eqs. (21) and (22)], with the conductivities σ_{ij} replaced by the conductances σ_{ij}/r_{ij} , is used. In this manner the transport properties of complex, two-dimensional fracture networks can be calculated, for example.

IV. CONCLUDING REMARKS

The WDM can be used to calculate the *effective* (macroscopic) and *local* (microscopic) transport properties of a heterogeneous medium represented by a scalar field of local transport coefficients (e.g., conductivity) or by a “resistor network” derived from that scalar field. As a calculation can be accomplished by monitoring the position of a single

walker, the method requires very little computer memory and so is especially suitable for highly complex material systems such as geomedia. Because the walkers “solve” the transport equations “on the fly,” the WDM may also be useful in studies of dynamic phenomena such as miscible fluid invasion in permeable media.

ACKNOWLEDGMENTS

This work was supported in part by the U.S. Department of Energy, Office of Environmental Management, under DOE Idaho Operations Office Contract No. DE-AC07-99ID13727.

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