Efficient simulation technique to compute effective properties of heterogeneous media

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We present a new simulation technique to "exactly" yield effective transport properties of disordered heterogeneous media in which the transport process is governed by a steady-state diffusion equation. Hence, the algorithm, which is based upon simulating the Brownian motion of a diffusing particle, can be applied to determine the effective electrical and thermal conductivity, dielectric constant, magnetic permeability, diffusion coefficient associated with flow past fixed obstacles, and the trapping rate associated with diffusion-controlled reactions among sinks. The simulation method is shown to have a very fast execution time. The technique is illustrated by computing the trapping rate associated with diffusion-controlled reactions; it is demonstrated to have an execution time that is at least an order of magnitude faster than previous simulation methodologies.

The problem of determining the effective properties (e.g., transport, electromagnetic, mechanical, etc.) of disordered heterogeneous media, such as composites, suspensions, and porous media, is an outstanding one in science and engineering. In recent years, considerable theoretical progress has been made in this classical research area (see the reviews of Batchelor,[1] Hashin,[2] Calef and Deutch,[3] Milton,[4] and Torquato;[5] and references therein). Conversely, there is, relatively, a dearth of work on "exact" simulations of the effective property of interest, especially for "continuum" models. Such "computer experiments" could provide unambiguous tests on theories for well-defined continuum model microstructures. Computer simulations could also yield information on quantities of theoretical importance that are not readily measurable in the laboratory.

Unfortunately, most computer simulation studies carried out in the past have attempted to solve the local governing differential equations for the fields (e.g., electric, temperature, concentration, etc.), subject to the appropriate boundary conditions at the multiphase interface of the computer-generated heterogeneous system, using some numerical technique such as finite differences or finite elements. This is repeated for all possible configurations and then the fields are configurationally averaged since the effective properties depend upon ensemble averages of the fields.[1–5] This is a very inefficient and wasteful way of getting the average behavior since there is a significant amount of information lost in going from the local to the average fields. Accordingly, such calculations become computationally exorbitant, even when performed on a supercomputer.

In this letter we describe a new algorithm to compute effective transport properties associated with processes governed by a steady-state diffusion equation, e.g., electrical and thermal conductivity, dielectric constant, magnetic permeability, diffusion coefficient, and the rate constant (trapping rate) associated with diffusion-controlled reactions. The algorithm is based upon simulating the Brownian motion of a diffusing particle in the random medium and relating an appropriate mean square displacement of the particle trajectory to the effective property. This procedure provides a direct and efficient means of obtaining the average behavior.

In order to illustrate the present simulation method, we compute the trapping rate associated with diffusion-controlled reactions among static spherical traps. Our algorithm is shown to have an execution time that is at least an order of magnitude faster than two recently employed algorithms that also simulate Brownian motion.[6–8] It should be noted that the present method may be applied to compute the effective properties of general continuum model microstructures, e.g., distributions of inclusions of arbitrary shape, which, may or may not overlap one another, have some specified orientation, etc.

We first describe the recent work of Zheng and Chiew[6] and Lee *et al.*[7] who independently computed the steady-state trapping rate k associated with diffusion-controlled reactions among static, spherical traps of variable penetrability. The reactant (in the trap-free region) diffuses but is instantly absorbed on contact with any trap. At steady state, the rate of production is exactly compensated by its removal by the traps.

Consider first the algorithm of Lee *et al.*[7] which we refer to as "method A." Method A is based upon the use of a Pearson ("continuum") random walk in which the step size a is fixed and successive directions are random and uncorrelated. The trapping rate is simply the inverse of the average survival time $\bar{t}$ for the random walkers. Now if $\bar{n}$ denotes the mean number of steps taken by the random walkers and $\bar{n} \gg 1$, then the random walk becomes simple Brownian motion and one has

$$k = \bar{t}^{\ -1} = 6D/\bar{n}a^2, \tag{1}$$

or, equivalently, since the mean square displacement $\overline{r^2} = \bar{n}a^2$, one also has

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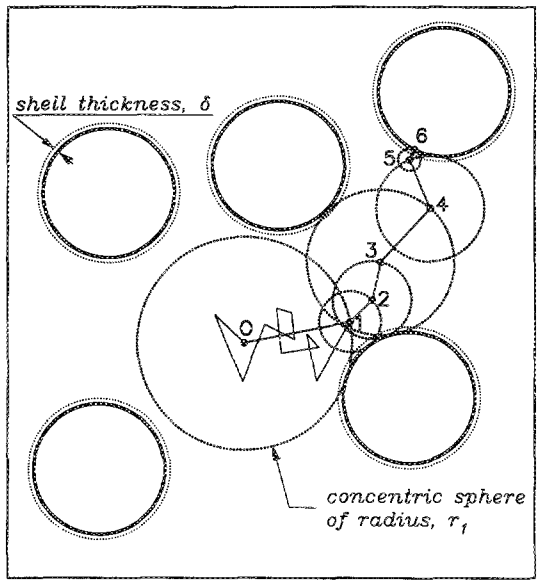

FIG. 1. Two-dimensional schematic representation of the new simulation method employed in this study. The zig-zag motion of the random walker need not be simulated step by step. Instead one constructs the largest concentric sphere of radius r_i which does not overlap any trap; the next position of the walker is taken to be on the concentric sphere surface. This process is repeated until the random walker gets trapped (i.e., comes within a distance δ of a trap). The sum over all the r_i^2 (equal to r^2) is related to the trapping rate through Eq. (2). In order to significantly reduce computer time to check for trapping, our algorithm makes use of the Grid method as described in Ref. 9.

$$k = 6D/\bar{r}^2, \tag{2}$$

where D is the diffusion coefficient. Since $\bar{n}$ must be large, then the step size a must be small compared to the trap radius R in the simulations. As a result, the trapping rate is determined by extrapolating data for a number of different step sizes to the $a/R \to 0$ limit. An important feature of method A is the use of the so-called "Grid method" which significantly reduces the computer time required to check if the random walker has been trapped. Essentially the Grid method[9] enables one to check for traps in the immediate neighborhood of the random walker instead of checking each trap. We note that method A has recently been used to determine k for cases in which the traps have a polydispersivity in size.[10]

The algorithm of Zheng and Chiew,[6] which we term "method B," is based upon the use of the first passage time probability distribution[11]:

$$P(t;r) = 1 + 2\sum_{m=1}^{\infty}(-1)^m \exp\left(-\frac{Dm^2\pi^2 t}{r^2}\right). \tag{3}$$

$P(t;r)$ is the cumulative distribution function associated with the time taken t for a random walker initially at the origin to reach the surface of a sphere of radius r. The basic idea is that the zig-zag random motion of the diffusing particle need not be simulated in detail; instead it is taken into account in a single simulation step using (3) (see Fig. 1). First one constructs the largest possible concentric sphere of radius r about the walker which does not overlap any trap particles, and then a point on the sphere surface of radius r is chosen randomly. The time required to move from the sphere center to the surface is determined by selecting a random number in the interval [0,1], inserting it into (3), and finding the corresponding time t_i. This process is repeated until the random walker is trapped and the survival time is obtained by summing over all the t_i. In practice, this can never be achieved in the course of the simulation. Zheng and Chiew assume the solute is trapped if it lies within a small distance δ from the trap surface (see Fig. 1). The average survival time $\bar{t}$ depends upon the shell thickness but they claim to obtain accurate results if $\delta/R < 0.0001$. It is important to note that the algorithm of method B does *not* make use of the Grid method to check for trapping.

Let us compare methods A and B. We have computed the scaled trapping rate k/k_s (where $k_s = 3D\phi_2/R^2$ is the dilute-limit Smoluchowski result) for fully penetrable traps (see Fig. 2)at two reduced density η values (0.3 and 0.5) using both method A and method B. (Here, $\eta = \rho 4\pi R^3/3$, where ρ is the trap number density.) It should be noted that this model, in general, is actually more computationally intensive than the case of the opposite extreme of totally impenetrable sinks (see Ref. 7 for further details). In each of the cases, we considered 490 spherical traps in a central cubical cell and employed periodic boundary conditions. The configurations of traps were generated using a standard Metropolis algorithm. We carried out 1000 random walks per configuration and considered a total of 50 configurations. The shell thickness ratio δ/R was taken to be 0.0001. In Table I we summarize these results along with the CPU times required for the calculations on a VAXstation 3200. For $\eta = 0.3$, method A is about 1.2 times faster than method B. For $\eta = 0.5$, method A is almost twice as fast as method B. Thus, even though method B avoids the need to exercise the zig-zag motion of method A, method A is faster because of its use of the Grid method to check for trapping.

A new algorithm to compute effective properties of continuum models of heterogeneous media which has a substantially faster execution time than either method A or B is now described. The new procedure combines certain aspects of methods A and B. For the purposes of this letter, we illustrate the new method by computing the trapping rate k.

As noted above, the zig-zag motion of the diffusing par-

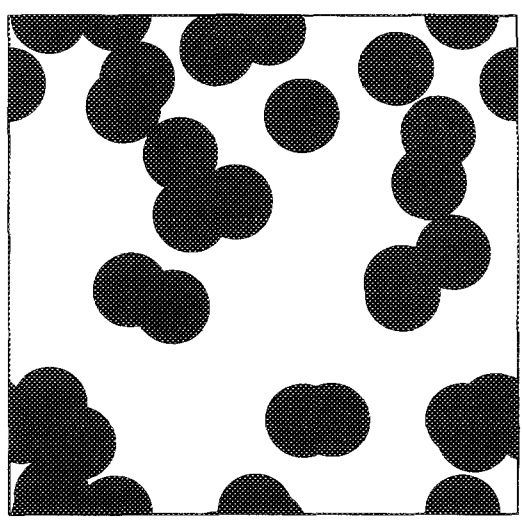

FIG. 2. Computer-generated realization of a distribution of fully penetrable traps in two dimensions. Here the trap volume fraction is about 0.4.

TABLE I. Comparison of the computing time required by the three different algorithms (method A,[a] method B,[b] and our new method) to obtain the scaled trapping rate k/k_s for a system of equisized fully penetrable traps at the reduced densities $\eta = 0.3$ and $\eta = 0.5$. Simulations were performed on a VAXstation 3200 using 490 traps, 50 configurations, and 1000 random walkers per configuration. Here $\delta/R = 0.0001$.

	$\eta = 0.3$		$\eta = 0.5$	
Algorithm	CPU time (h)	k/k_s	CPU time (h)	k/k_s
Method A	12.08	2.476	6.85	3.301
Method B	14.68	2.431	13.75	3.330
New method	0.77	2.469	0.72	3.330

[a] Reference 7.
[b] Reference 6.

ticle does not have to be simulated in detail; one can (as is done in method B) construct the largest possible concentric sphere of radius r about the solute which does not overlap any trap particles, choose a point on the surface, and compute the time taken using the first passage probability distribution, Eq. (3). However, calculation of the time using (3) is unnecessary since the mean square displacement r^2 is already known and can itself be related to the trapping rate [cf. Eq. (2)]. To prove this last point, consider obtaining the average time τ required for a random walker to first strike the surface of the largest concentric sphere:

$$\tau = \int_0^\infty t \frac{\partial P}{\partial t} dt = \frac{r^2}{6D}. \quad (4)$$

Now the total of the average times for each concentric sphere up to trapping, $\bar{t}$, is simply

$$\bar{t} = \sum_{i=1} \tau_i = \sum_{i=1} r_i^2 \quad 6D, = \bar{r^2}/6D, \quad (5)$$

which is the same as the inverse of (2), i.e., the expression used in method A. Hence, in the new algorithm we keep track of r^2 rather than t. We also assume that a solute is trapped if it lies within a distance δ from a trap surface. Moreover, as was done in method A, we employ the Grid method to dramatically reduce the computing time to check for trapping. In Table I we include corresponding calculations using the new simulation method. For a reduced density $\eta = 0.3$, the new method is about 16 and 19 times faster than methods A and B, respectively. For $\eta = 0.5$, the new algorithm is about 10 and 19 times faster than methods A and B, respectively. Thus, the new simulation method is extremely fast and accurate, requiring less than a CPU hour for each of the densities reported.

We are currently in the process of employing this new algorithm to compute the electrical (or thermal) conductivity of continuum models of multiphase media. For reasons of mathematical analogy, the results of such a study translate immediately into equivalent results for the dielectric constant, magnetic permeability, and the diffusion coefficient. Such simulations involves two new features: (1) different walking speeds in each phase and (2) a nonzero probability of reflection at the interface between the different materials.

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