COMMENTS

Comments are short papers which criticize or correct papers of other authors previously published in the **Physical Review.** *Each Comment should state clearly to which paper it refers and must be accompanied by a brief abstract. The same publication schedule as for regular articles is followed, and page proofs are sent to authors.*

Comment on ''Walker diffusion method for calculation of transport properties of composite materials''

In Chan Kim,[1] Dinko Cule,[2] and Salvatore Torquato[2,3]
[1]*School of Mechanical Engineering, Kunsan National University, Kunsan City, Chonbuk 573-701, Korea*
[2]*Princeton Materials Institute, Princeton University, Princeton, New Jersey 08544*
[3]*Department of Chemistry, Princeton University, Princeton, New Jersey 08544*
(Received 15 June 1999)

In a recent paper [C. DeW. Van Siclen, Phys. Rev. E **59**, 2804 (1999)], a random-walk algorithm was proposed as the best method to calculate transport properties of composite materials. It was claimed that the method is applicable both to discrete and continuum systems. The limitations of the proposed algorithm are analyzed. We show that the algorithm does not capture the peculiarities of continuum systems (e.g., ''necks'' or ''choke points'') and we argue that it is the stochastic analog of the finite-difference method.

PACS number(s): 05.60.−k, 44.30.+v, 91.60.Pn, 66.30.−h

Van Siclen has presented a lattice random-walk method to calculate the transport properties of discrete models of composite materials [1]. The method belongs to a broad class of discrete Brownian walker diffusion models that have been extensively studied [2]. Considering a density of walker populations in distinct phase domains, Van Siclen utilized an expression for the probability $p_{i,j}$ that the walker successfully moves across a domain boundary from phase i to phase j [3]: $p_{i,j}=\sigma_j/(\sigma_i+\sigma_j)$, where σ_i, σ_j are the corresponding local conductivities. Then the *effective* conductivity σ_e of a d-dimensional composite medium can be readily obtain by monitoring the walker displacements R_i and associated mean time intervals τ_i in each visited phase, since $\sigma_e=\Sigma_i R_i^2/\Sigma_i(2d\tau_i\sigma_i^{-1})$ [1,2,4,5].

For practical calculations, Van Siclen introduced a computationally efficient ''variable residence time algorithm'' in which each walker attempts to move to one of the adjacent two-dimensional (2D) sites is always successful but it takes a variable time interval to accomplish it. The algorithm requires knowledge of the probability $P_{i,j}$ for a successful move from a given site i to an adjacent site j and the corresponding average time interval T_i over which this particular move occurs. These are given by $P_{i,j}=\tilde{p}_{i,j}+\tilde{q}_i\tilde{p}_{i,j}+\tilde{q}_i^2\tilde{p}_{i,j}+\cdots=p_{i,j}/(\Sigma_{k=1}^{2d}p_{i,k})$, and $T_i=\tau 2d/\Sigma_{k=1}^{2d}p_{i,k}$, where $\tilde{p}_{i,j}=(2d)^{-1}p_{i,j}$ (since there are $2d$ possible directions), $\tilde{q}_i=1-\Sigma_{j=1}^{2d}\tilde{p}_{i,j}$ is the probability of unsuccessful interface crossings, and $\tau=(4d)^{-1}$ is the time interval unit [1,6]. The proposed simple lattice walker method is attractive since it can be implemented easily and efficiently.

However, Van Siclen claims that the method is also applicable to continuum systems as well as to its digitized representations [1]. In the following, we show that the peculiarities of the continuum systems (digitized or not) are not captured by his algorithm. Moreover, his proposed procedure is the stochastic analog of the standard finite-difference method. Thus, Van Siclen's claim that his method is the only viable and best ''exact'' method to compute transport properties [1] is not the case and has the same limitations as the finite-difference scheme.

It is known that the contribution from transport through very narrow ''necks'' or ''choke points'' that occur in continuum systems can lead to effective transport properties that are dramatically different from lattice models [7–9]. Such choke points become numerous and especially important near the percolation threshold. These contributions are not included in Van Siclen's algorithm which involves a standard discretization of the problem. Moreover, he equates his standard lattice walk with a continuum walk on a digitized representation of a continuum system [1]. These two situations are indeed not the same for the aforementioned reasons, i.e., transport in digitized media may be dominated by choke or ''corner'' points that connect two diagonal pixel neighbors [8,9]. These are *next*-nearest-neighbor connections that are not taken into account in Van Siclen's algorithm.

To demonstrate that Van Siclen's algorithm does not capture the correct behavior of two-phase continuum systems, consider two-dimensional digitized representations. Each square pixel represents one of the phases and periodic boundary conditions are applied to the entire square (system) unit cell. Consider now one of the simplest continuum systems with natural square discretization: q regular two-phase checkerboard with phase conductivities σ_A and σ_B. It is well known that the effective conductivity of this system (as well as the random checkerboard) is exactly given by $\sigma_e=\sqrt{\sigma_A\sigma_B}$ [10]. Let the pixels cells coincide with the checkerboard cells. We now show that application of Van Siclen's algorithm to this situation leads to an incorrect result [11]. In

this simple case, it is straightforward to calculate the probability $P_{i,j}$ and time T_i. Because of the regularity of the checkerboard, one has that $P_{i,j}=1/4$, and $T_A=\tau(\sigma_A+\sigma_B)/\sigma_B$ or $T_B=\tau(\sigma_A+\sigma_B)/\sigma_A$, depending on the starting point of the walker. Thus, in this particular case, Van Siclen's procedure is exactly solvable, and gives $\sigma_e=2\sigma_A\sigma_B/(\sigma_A+\sigma_B)$, which we see is incorrect. If the ratio σ_B/σ_A differs significantly from unity, this result significantly underestimates σ_e since transport through corners is *not* included. For example, for $\sigma_A=1$ and $\sigma_B=100$, the exact result yields $\sigma_e=10$, whereas the result $\sigma_e=2\sigma_A\sigma_B/(\sigma_A+\sigma_B)$ gives $\sigma_e=200/101\approx 1.98$.

Of course, the continuum system is better approximated by a lattice random-walk model if the spatial resolution of the lattice used to discretize the system is increased. This is also true for a conventional finite-difference computation. The connection between these two techniques is well established. In the limit when the number of sampled random walk paths goes to infinity, the solution of the lattice random-walk model is *identical* to that obtained from an exact solution of the finite-difference representation [12]. It is therefore useful to analyze the dependence of the computed finite-difference results on the lattice resolution used in the calculations. These results also represent the most accurate results if the probabilistic lattice model based on the same discretization scheme is used.

To illustrate the limitations of standard network discretizations in the calculation of transport properties of continuum systems, we have calculated the effective conductivity of the two-dimensional regular checkerboard for various grid resolutions. Each square cell of the checkerboard is discretized by an $L\times L$ grid. The results for a modest contrast ratio $\sigma_A/\sigma_B=100$, obtained by employing the finite-difference method with a conjugate gradient iterative procedure, are shown in Fig. 1. We note that the $L=1$ result matches the aforementioned Van Siclen's result for that case, as expected. The approach to the exact value with increasing L is quite slow. By increasing the contrast ratio, the discrepancy is even greater. In the extreme limit $\sigma_A/\sigma_B\rightarrow 0$ or ∞, the finite-difference method (or Van Siclen's method) is not applicable to the checkerboard problem.

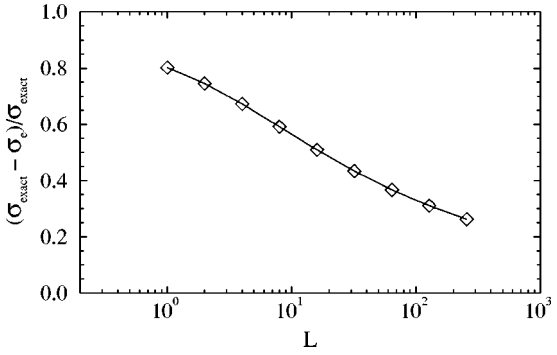

FIG. 1. Effective conductivity of a two-dimensional, two-phase regular checkerboard with phase conductivities $\sigma_A=1$, $\sigma_B=100$. The normalized difference between the finite-difference and exact result $\sigma_{exact}=(\sigma_A\sigma_B)^{1/2}$ is shown as a function of the resolution L used in the finite-difference method.

In conclusion, the lattice random-walk method proposed in Ref. [1] cannot be utilized generally to compute transport properties of digitized representations of continuum composite materials, much less actual continuum composites. There are very general random-walk methods based on first-passage time equations to compute the transport properties of continuum composite materials [4,5]. Reference [5] provides a theoretical foundation for the first-passage time equations used in Ref. [4] and shows how to apply the analysis to digitized continuum systems. This continuum random-walk method can correctly capture the effect of ''touching corners,'' which is very important for certain morphologies, such as those characteristic of systems at the percolation threshold. The differences between discrete lattice and continuum models are even greater in higher-dimensional systems ($d>2$) near the percolation threshold [7].

This work was supported by the Engineering Research Program of the Office of Basic Energy Science at the Department of Energy under Grant No. DE-FG02-92E4275.

[1] C. DeW. Van Siclen, Phys. Rev. E **59**, 2804 (1999).
[2] D. C. Hong, H. E. Stanley, A. Coniglio, and A. Bunde, Phys. Rev. B **33**, 4564 (1986), and references therein.
[3] A first principles derivation of this formula for continuum systems can be found in Refs. [4] and [5].
[4] I. C. Kim and S. Torquato, J. Appl. Phys. **68**, 3892 (1990); **69**, 2280 (1991).
[5] S. Torquato, I. C. Kim, and D. Cule, J. Appl. Phys. **85**, 1560 (1999).
[6] We note that the time T_i is related to mean-first-passage time from lattice site i to site j (other adjacent sites are excluded) since $T_i=P_{i,j}^{-1}[(1\tau)\tilde{p}_{i,j}+(2\tau)\tilde{q}_i\tilde{p}_{i,j}+(3\tau)\tilde{q}_i^2\tilde{p}_{i,j}+\cdots]=\tau 2d/\Sigma_{k=1}^{2d}p_{i,k}$.
[7] S. Feng, B. I. Halperin, and P. N. Sen, Phys. Rev. B **35**, 197 (1987).
[8] P. Sheng and R. V. Kohn, Phys. Rev. B **26**, 1331 (1982).
[9] L. Berlyand and K. Golden, Phys. Rev. B **50**, 2114 (1994).
[10] A. M. Dykhne, Zh. Éksp. Teor. Fiz. **59**, 110 (1970) [Sov. Phys. JETP **32**, 63 (1971)]; K. S. Mendelson, J. Appl. Phys. **46**, 917 (1975).
[11] This example serves to illustrate the limitations of standard network discretizations of the problem. We are not suggesting that Van Siclen would choose such a coarse lattice for the checkerboard.
[12] A. Haji-Sheikh, in *Handbook of Numerical Heat Transfer*, edited by W. J. Minkowycz *et al.* (Wiley-Interscience, New York, 1988).