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

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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.

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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 \( \sigma_i \) and \( \sigma_j \) are the corresponding local conductivities. Then the effective conductivity \( \sigma_e \) of a \( d \)-dimensional composite medium can be readily obtained by monitoring the walker displacements \( R \) and associated mean time intervals \( \tau \) in each visited phase, since

\[
\sigma_e = \frac{\sum R^2_j / \sum_2 (2d \tau, \sigma_i^{-1})}{1, 2, 4, 5}.
\]

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 in a natural square unit cell. Consider now one of the simplest continuum systems with natural square discretization: \( q \) regular two-phase checkerboard with phase conductivities \( \sigma_A \) and \( \sigma_B \). It is well known that the effective conductivity of this system is exactly given by

\[
\sigma_e = \sqrt{\sigma_A \sigma_B}.
\]

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 \( \sigma_A \) and \( \sigma_B \). It is well known that the effective conductivity of this system is exactly given by

\[
\sigma_e = \sqrt{\sigma_A \sigma_B}.
\]

Let the pixels cells coincide with the checkerboard cell. 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 $T_{A} = \pi(\sigma_{A} + \sigma_{B})/\sigma_{B}$ or $T_{B} = \pi(\sigma_{A} + \sigma_{B})/\sigma_{A}$, depending on the starting point of the walker. Thus, in this particular case, Van Siclen’s procedure is exactly solvable, and gives $\sigma_{x} = 2\sigma_{A}\sigma_{B}/(\sigma_{A} + \sigma_{B})$, which we see is incorrect. If the ratio $\sigma_{B}/\sigma_{A}$ differs significantly from unity, this result significantly underestimates $\sigma_{x}$ since transport through corners is not included. For example, for $\sigma_{A} = 1$ and $\sigma_{B} = 100$, the exact result yields $\sigma_{x} = 10$, whereas the result $\sigma_{x} = 2\sigma_{A}\sigma_{B}/(\sigma_{A} + \sigma_{B})$ gives $\sigma_{x} = 200/101 \approx 1.98$.

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 $\infty$, the finite-difference method (or Van Siclen’s method) is not applicable to the checkerboard problem.

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].

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[3] A first principles derivation of this formula for continuum systems can be found in Refs. [4] and [5].
[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 $T_{i} = P_{i,j}^{-1}[(1/2)\tau_{i,j} + (2/3)\tau_{j,i} + (3/4)\tau_{k,i} + \ldots]$.
[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.