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Local volume fraction fluctuations in heterogeneous media

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The volume fractions of multiphase heterogeneous media fluctuate on a spatially local level even for statistically homogeneous materials. A general formulation is given to represent the standard deviation associated with the *local* volume fraction of statistically homogeneous but anisotropic D-dimensional two-phase media for arbitrary-shaped *observation regions.* The standard deviation divided by the macroscopic volume fraction, termed the *coarseness*, is computed for D-dimensional distributions of penetrable as well as impenetrable spheres, for a wide range of densities and observation-region sizes. The effect of impenetrability of the particles, for fixed observation-region size, is to reduce the coarseness relative to that of the penetrable-sphere model, especially at high densities. For either sphere model, increasing the dimensionality D decreases the coarseness.

I. INTRODUCTION

Characterizing the microstructure of heterogeneous media, such as composite materials, porous media and cracked solids, is crucial in determining the macroscopic physical properties of such materials.[1–3] One of the most important morphological descriptors is the volume fraction of the phases; in the case of porous media, the porosity (i.e., the volume fraction of the fluid phase). Although the volume fraction is constant for statistically homogeneous media, on a spatially local level it fluctuates. An interesting and relatively unexplored question in the study of multiphase random media is the following: How does the "local" volume fraction fluctuate about its average value? The answer to this query has relevance to a number of problems, including scattering by heterogeneous media,[4] transport through composites and porous media,[1–3] and the study of noise and granularity of photographic images.[5–7] It is actually in the context of the latter problem, image science, that this question of local volume fraction fluctuations has been examined to any degree, and here primarily for simple two-dimensional models of photographic emulsions which do not account for impenetrability of the grains.[5,6]

The purpose of this paper is to provide a general means of representing and computing the *standard deviation* associated with the *local volume fraction* $\tau(\mathbf{x})$ at position $\mathbf{x}$ for arbitrary D-dimensional two-phase random media which are statistically homogeneous. The local volume fraction $\tau(\mathbf{x})$ is defined to be the volume fraction of the one of the phases, say phase 1, contained in some generally finite-sized "observation region". Clearly, $\tau(\mathbf{x})$ is a random variable and becomes a constant, equal to the volume fraction of phase 1, ϕ_1, in the limit of an infinitely large observation region. The quantity that we specifically study is the *coarseness* defined to be the standard deviation divided by ϕ_1. For concreteness, we apply our formalism to compute the standard deviation as a function of ϕ_1 and the size of the observation region for distributions of D-dimensional fully penetrable spheres (with $D=1$, 2, and 3) as well as D-dimensional impenetrable spheres with $D=1$ (hard rods), $D=2$ (hard disks) and $D=3$ (hard spheres).

In Sec. II, we define the quantities of concern and describe the basic relations. In Sec. III, we derive the general coarseness formula (for an arbitrary-shaped observation region) of any D-dimensional two-phase medium that is statistically homogeneous but anisotropic. This expression is given in terms of the two-point probability function of the medium. Using the expression for this probability function of anisotropic media composed of distributions of identical, oriented inclusions of arbitrary shape, we obtain an explicit relation for the coarseness of this wide class of media. In Sec. IV, we apply our results by computing the coarseness of D-dimensional distributions of penetrable and impenetrable spheres (with $D=1$, 2, and 3) with a D-dimensional spherical observation region. In Sec. V, we make concluding remarks.

II. DEFINITIONS AND BASIC RELATIONS

The random medium is a domain of space $\mathscr{V}(\omega\in\mathscr{R}^D)$ (where the realization ω is taken from some probability space Ω) of D-dimensional volume V which is composed of two regions: the phase 1 region $\mathscr{V}_1$ of volume fraction ϕ_1 and phase 2 region region $\mathscr{V}_2$ of volume fraction ϕ_2. Depending on the physical context, phase $i(i=1$ or $2)$ can be either void, fluid or solid.

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A. Characteristic function and *n*-point probability functions

The characteristic function $I(\mathbf{x})$ of phase 1 is defined by

$$I(\mathbf{x})=\begin{cases}1, & \mathbf{x}\in\mathscr{V}_1\\ 0, & \mathbf{x}\in\mathscr{V}_2\end{cases}. \tag{2.1}$$

To describe the structure of the system from statistical point of view, it is useful to introduce the n-point probability function S_n[8] which is defined according to the relation

$$S_n(\mathbf{x}_1,...,\mathbf{x}_n)=\left\langle \prod_{i=1}^{n} I(\mathbf{x}_i)\right\rangle. \tag{2.2}$$

The angular brackets above denote an ensemble average. S_n is the probability of simultaneously finding n points at positions $\mathbf{x}_1,...,\mathbf{x}_n$ all in phase 1 region $\mathscr{V}_1$. If the medium is statistically homogeneous, then the n-point probability function is translationally invariant and as a result only a function of the relative displacements, i.e., $S_n = S_n(\mathbf{x}_{12},...,\mathbf{x}_{1n})$, where $\mathbf{x}_{1i} = \mathbf{x}_i-\mathbf{x}_1$. For statistically homogeneous media, invoking an ergodic hypothesis, one can then equate the volume average $\overline{f}$ of some function f,

$$\overline{f}=\lim_{f\to\infty}\frac{1}{V}\int_V f\,dV, \tag{2.3}$$

to the ensemble average $\langle f\rangle$. Furthermore, if the medium is statistically isotropic, then S_n only depends upon the relative distances $x_{12},...,x_{1n}$, where $x_{1i} = |\mathbf{x}_i-\mathbf{x}_1|$. For simplicity, we shall consider statistically homogeneous but anisotropic media only.

For homogeneous media, S_1 is the probability of finding a point at $\mathbf{x}_1$ in region $\mathscr{V}_1$, and is independent of the location of the point in the space. By definition, S_1 is just the expected value of I, i.e.,

$$S_1(\mathbf{x}_1)=\langle I(\mathbf{x}_1)\rangle=\phi_1, \tag{2.4}$$

where ϕ_1 is just the volume fraction of phase 1. Now let us consider the two-point probability function

$$S_2(\mathbf{x}_{12})=\langle I(\mathbf{x}_1)I(\mathbf{x}_2)\rangle, \tag{2.5}$$

which is the autocorrelation function of I. It contains considerably more information than S_1. In the event $x_2\to x_1$, we have

$$S_2(\mathbf{x}_{12})\to\phi_1. \tag{2.6}$$

On the other hand, when the relative distance between the two points becomes very large, we have

$$\lim_{\mathbf{x}_{12}\to\infty} S_2(\mathbf{x}_{12})=\phi_1^2, \tag{2.7}$$

assuming no long-range correlations.

The fluctuations associated with the characteristic function of phase 1 can be measured by the variance σ_I^2 given by

$$\sigma_I^2\equiv\langle I^2\rangle-\langle I\rangle^2=\phi_1-\phi_1^2. \tag{2.8}$$

At fixed ϕ_1, the variance is seen to be a trivial constant and hence does not provide much useful structural information about the random medium.

B. Local volume fraction fluctuations

The volume fraction of phase 1 (or phase 2) fluctuates on a local level. That is, consider the volume fraction of phase 1 contained in an arbitrary-shaped "observation region" $\mathscr{V}_O$ of D-dimensional volume V_O whose centroid is located at $\mathbf{x}$. As the observation region is moved from point to point in the sample, it is clear that the volume fraction of phase 1 contained in it will fluctuate for finite V_O. Thus this local volume fraction $\tau(\mathbf{x})$ is a random variable defined to be

$$\tau(\mathbf{x})\equiv\frac{1}{V_O}\int I(\mathbf{z})\theta(\mathbf{z}-\mathbf{x})d\mathbf{z}, \tag{2.9}$$

where

$$\theta(\mathbf{x})=\begin{cases}1, & x\in\mathscr{V}_l\\ 0, & \text{otherwise}\end{cases} \tag{2.10}$$

is the observation region indicator function. Note that as $V_O\to\infty$, we have

$$\tau(\mathbf{x})\to\langle I\rangle=\phi_1. \tag{2.11}$$

In the limit of a very small observation region ($V_O\to 0$), $\tau(\mathbf{x})$ simply becomes the characteristic function of phase 1. i.e.,

$$\tau(\mathbf{x})\to I(\mathbf{x}). \tag{2.12}$$

The expected value of τ is clearly given by

$$\langle\tau\rangle=\phi_1. \tag{2.13}$$

A measure of local fluctuations in the volume fraction is then given by

$$C=\frac{\sigma_\tau}{\langle\tau\rangle}=\frac{\sigma_\tau}{\phi_1}, \tag{2.14}$$

where

$$\sigma_\tau^2=\langle\tau^2\rangle-\langle\tau\rangle^2=\langle\tau^2\rangle-\phi_1^2 \tag{2.15}$$

is the variance associated with the $\tau(\mathbf{x})$. We term C, a *scaled* standard deviation, the "coarseness" since it provides a quantitative measure of nonuniformity of coverage of the phases. From Eqs. (2.12) and (2.13), we have that C for infinitely large and infinitely small observation regions is given, respectively, by

$$C=0 \tag{2.16}$$

and

$$C=\frac{\sigma_I}{\phi_1}=\frac{\sqrt{\phi_1\phi_2}}{\phi_1}. \tag{2.17}$$

The dependence of the coarseness C on the observation region is, in general, nontrivial as it depends upon the details of the microstructure of the random medium. A derivation of this relationship shall be presented in the following section.

III. DERIVATION OF COARSENESS FORMULAS

In this section, we derive, using a new approach, a general expression for the coarseness of arbitrary statistically anisotropic D-dimensional heterogeneous media and an arbitrary-shaped observation region in terms of the two-point probability function S_2. For the special case of two-dimensional media ($D=2$), our general coarseness formula is equivalent to but functionally different than one due to O'Neill[5] who used a different derivation procedure. An explicit representation of S_2 for a wide class of anisotropic media composed of arbitrarily shaped particles is described. Using this expression for S_2 and the general coarseness formula, we then derive a new, explicit expression for the coarseness of the aforementioned class anisotropic media.

A. General coarseness formula

From the definition of Eq. (2.15) we can get the variance for an arbitrary-shaped observation region with some fixed orientation. Substitution of Eqs. (2.9) and (2.14) into Eq. (2.15) gives

$$\sigma_\tau^2=\frac{1}{V_O^2}\left\langle \int I(\mathbf{z})\theta(\mathbf{z}-\mathbf{x})d\mathbf{z} \int I(\mathbf{y})\theta(\mathbf{y}-\mathbf{x})d\mathbf{y}\right\rangle-\phi_1^2. \tag{3.1}$$

Now since the above enemble average operator and the integral operators commute, Eq. (3.1) can be rewritten as

$$\sigma_\tau^2=\frac{1}{V_O^2}\int d\mathbf{y}\, d\mathbf{z}\, \theta(\mathbf{y}-\mathbf{x})S_2(\mathbf{z}-\mathbf{y})-\phi_1^2, \tag{3.2}$$

where $S_2(\mathbf{z}-\mathbf{y}) = \langle I(\mathbf{y})I(\mathbf{z})\rangle$ is the two-point probability function defined by Eq. (2.5). Since we are treating statistically homogeneous media, $S_2(\mathbf{z}-\mathbf{y}) = S_2(\mathbf{y}-\mathbf{z})$, and because Eq. (3.1) is independent of position, we can integrate over $\mathbf{x}$ instead of $\mathbf{y}$ and find

$$\sigma_\tau^2=\frac{1}{V_O^2}\int S_2(\mathbf{r})V_2^{\text{int}}(\mathbf{r};\sigma_o)d\mathbf{r}-\phi_1^2, \tag{3.3}$$

where

$$V_O^{\text{int}}(\mathbf{r};\sigma_o)=\int \theta(\mathbf{y}-\mathbf{x})\theta(\mathbf{z}-\mathbf{x})d\mathbf{x} \tag{3.4}$$

is the intersection volume of two observation regions whose centroids are separated by the displacement $\mathbf{r}=\mathbf{z}-\mathbf{y}$, and σ_o denote *all* of the shape parameters associated with the observation region. Dividing Eq. (3.4) by V_O^2 and integrating over $\mathbf{r}$ gives

$$\frac{1}{V_O^2}\int V_2^{\text{int}}(\mathbf{r};\sigma_o)d\mathbf{r}=1. \tag{3.5}$$

We then can write Eq. (3.3) in the following way:

$$C=\frac{1}{\phi_1 V_O}\left[\int [S_2(\mathbf{r})-\phi_1^2]V_2^{\text{int}}(\mathbf{r};\sigma_o)d\mathbf{r}\right]^{1/2}. \tag{3.6}$$

This is the desired general expression for the coarseness valid for d-dimensional statistically anisotropic media of arbitrary topology and is given in terms of $S_2(\mathbf{r})$ and $V_2^{\text{int}}(\mathbf{r};\sigma_o)$.

In the term $[S_2(\mathbf{r})-\phi_1^2]$, which appears in the integrand of Eq. (3.6), ϕ_1^2 is the long range value of S_2. Hence, $[S_2(\mathbf{r})-\phi_1^2]$ decays to zero for large r. We refer to the range over which $[S_2(\mathbf{r})-\phi_1^2]$ is nonnegligibly small as the correlation length l. Consider the case where the characteristic size of the observation region is much larger than l, then $V_2^{\text{int}}(\mathbf{r};\sigma_o)$ is approximately equal to $V_2^{\text{int}}(0;\sigma_o) = V_O$. Thus, Eq. (3.6) in such instances yields

$$C=\frac{1}{\phi_1 V_O^{1/2}}\left[\int_{r<l}[S_2(\mathbf{r})-\phi_1^2]d\mathbf{r}\right]^{1/2}. \tag{3.7}$$

The coarseness in brackets is a constant and hence for large observation regions

$$C=KV_O^{-1/2}, \tag{3.8}$$

where

$$K=\frac{1}{\phi_1}\left[\int_{r<l}[S_2(\mathbf{r})-\phi_1^2]d\mathbf{r}\right]^{1/2} \tag{3.9}$$

is a constant which depends upon ϕ_1, or, equivalently, the volume fraction of phase 2. Equation (3.9) is reminiscent of the *compressibility equation* of liquid-state theory[9] which relates the D-dimensional volume integral over the *total correlation function* to density fluctuations in the system. In the limit $V_O\to\infty$, Eq. (3.9) agrees with Eq. (2.16).

Information about the shape of the observation region enters through the intersection volume $V_2^{\text{int}}(\mathbf{r};\sigma_o)$ [cf. Eq. (3.6)]. For D-dimensional spheres of diameter σ_o with $D=1$, 2, and 3, one respectively has,

$$V_2^{\text{int}}(r;\sigma_o)=(\sigma_o-r)H(\sigma_o-r). \tag{3.10}$$

$$V_2^{\text{int}}(r;\sigma_o)=\frac{\sigma_o^2}{2}\left[\cos^{-1}\frac{r}{\sigma_o}-\frac{r}{\sigma_o}\sqrt{1-\frac{r^2}{\sigma_o^2}}\right]H(\sigma_o-r) \tag{3.11}$$

and

$$V_2^{\text{int}}(r;\sigma_o)=\frac{\pi\sigma_o^3}{3}\left(1-\frac{3r}{2\sigma_o}+\frac{r^3}{2\sigma_o^3}\right)H(\sigma_o-r). \tag{3.12}$$

Where $H(x)$ is the Heaviside step function. In these equations, r is the distance between the centers of the two observation regions. For D-dimensional rectangular parallelepipeds with $D=1$, 2, and 3, one has, respectively,

$$V_2^{\text{int}}(x;a)=(a-x)H(a-x), \tag{3.13}$$

$$V_2^{\text{int}}(x,y;a,b)=(a-x)(b-y)H(a-x)H(b-y), \tag{3.14}$$

$$V_2^{\text{int}}(x,y,z;a,b,c)=(a-x)(b-y)\times(c-z)H(a-x)H(b-y)H(c-z). \tag{3.15}$$

B. Series representations of the n-point probability functions

In order to apply formula (3.6), knowledge of the two-point probability function S_2 for the particular system of interest is required. Torquato and Stell[8] have given a representation of S_n for distributions of identical D-dimensional spheres. These results have been generalized to particles of arbitrary shape by a simple reinterpretation of the "particle" indicator function.[3]

Consider a statistical distribution of N identical, interacting D-dimensional particles whose positions are completely specified by center-of-mass coordinates $\mathbf{r}^N \equiv \{\mathbf{r}_1,...,\mathbf{r}_N\}$. This class of anisotropic media includes distributions of D-dimensional spheres, oriented D-dimensional ellipsoids, oriented D-dimensional rectangular parallelpipeds, oriented D-dimensional cylinders, etc. Let the space interior to the particles be phase 2. For such a distribution of particles the S_n are related to the n-body distribution function g_n according to the following relation:

$$S_n(\mathbf{x}^n) = 1 + \sum_{k=1}^{\infty} \frac{(-1)^k \rho^k}{k!} \int \cdots \int g_k(\mathbf{r}^k) \times \prod_{j=1}^{k} \left[[1 - \prod_{i=1}^{n} [1 - m(\mathbf{x}_i - \mathbf{r}_j)] \right] d\mathbf{r}_j, \tag{3.16}$$

where

$$m(\mathbf{x}) = \begin{cases} 1, & \mathbf{x} \in D_P \\ 0, & \text{otherwise} \end{cases} \tag{3.17}$$

is the particle indicator function and D_P denotes a particle region. One can now, in principle, compute the S_n for any n, given the g_n. It is useful to consider the following special cases.

1. Fully penetrable particles

In the instance of "fully penetrable" particles, the particle centers are Poisson distributed, i.e., no spatial correlations exist between the particles and

$$g_n(\mathbf{r}^n) = 1 \quad \text{for all } \mathbf{r}^n. \tag{3.18}$$

Substitution of Eq. (3.18) into Eq. (3.16) then yields the analytical expression

$$S_n(\mathbf{x}^n) = \exp[-\rho V_n(\mathbf{x}^n)], \tag{3.19}$$

where $V_n(\mathbf{x}^n)$ represents the union volume of n identical particle regions centered at $\mathbf{x}^n$, respectively. In the special instance of D-dimensional spheres of diameter σ, for example, the volume of a single inclusion is

$$V_1(\sigma) = \frac{\pi^{D/2}}{\Gamma\left(1 + \frac{1}{2}\frac{D}{2}\right)} \left(\frac{\sigma}{2}\right)^D \tag{3.20}$$

and the union volume of two such objects whose centers are separated by the distance r is given by

$$V_2(r;\sigma) = 2\sigma - (\sigma - r) H(\sigma - r), \tag{3.21}$$

$$V_2(r;\sigma) = \frac{\pi\sigma^2}{2} - \frac{\sigma^2}{2} \left[\cos^{-1}\frac{r}{\sigma} - \frac{r}{\sigma}\sqrt{1 - \frac{r^2}{\sigma^2}} \right] H(\sigma - r), \tag{3.22}$$

$$V_2(r;\sigma) = \frac{\pi\sigma^3}{3} - \frac{\pi\sigma^3}{6} \left[1 - \frac{3r}{\sigma} + \frac{r^3}{2\sigma^3} \right] H(\sigma - r) \tag{3.23}$$

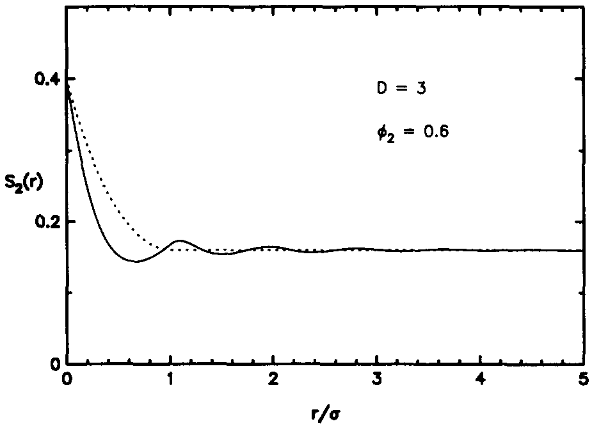

FIG. 1. The two-point probability function $S_2(r)$ for three-dimensional distributions of fully penetrable spheres (Ref. 10) (---) and totally impenetrable spheres (Ref. 13) (—) of diameter σ at a particle volume fraction $\phi_2 = 0.6$.

for $D=1$, 2, and 3, respectively. Here $\Gamma(x)$ is the gamma function.

Low-order S_n have already been evaluated for fully penetrable spheres,[10] fully penetrable circular disks,[11] and fully penetrable oriented, circular cylinders of finite aspect ratio.[3]

2. Totally impenetrable particles

For totally impenetrable particles, the infinite series (3.16) truncates exactly after the nth term in the sum. The first two n-point probabilities, after some simplification, are given by

$$S_1 = 1 - \rho V_1, \tag{3.24}$$

$$S_2(\mathbf{x}_{12}) = 1 - \rho V_2(\mathbf{x}_{12}) + \rho^2 \int \int g_2(\mathbf{r}_{12}) \times m(\mathbf{x}_1 - \mathbf{r}_1) m(\mathbf{x}_2 - \mathbf{r}_2) d\mathbf{r}_1\, d\mathbf{r}_2. \tag{3.25}$$

Comparison of Eq. (3.23) for $n=1$ to relations (3.27) reveals that the volume fraction of phase 1, ϕ_1 ($= S_1$), for the fully-penetrable-sphere model is always greater than ϕ_1 for the totally-impenetrable-sphere model at the same number density. Low-order S_n (e.g., S_1, S_2, and S_3) have been computed for equilibrium ensembles of impenetrable, equisized rods,[12] circular disks[12] and spheres.[13] For the case of the rods, Torquato and Lado[12] gave an exact analytical solution for $S_2(r)$. In contrast, $S_2(r)$ for equilibrium distributions of hard circular disks[12] and spheres[13] were computed in the Percus–Yevick approximation.

In Fig. 1, we compare the two-point probability functions for fully penetrable spheres[10] and totally impenetrable spheres[13] at a particle volume fraction $\phi_2 = 0.6$. In the former model, $S_2(r)$ monotonically decreases from its maximum value of ϕ_1 at $r=0$ to its minimum value of ϕ_1^2 at $r=\sigma$; for $r>\sigma$, $S_2(r) = \phi_1^2$, indicating no spatial correlation for such r. In contrast, $S_2(r)$ for hard spheres oscillates about its long-range value of ϕ_1^2 for several diameters indicating short-range order as the result of exclusion-volume effects.

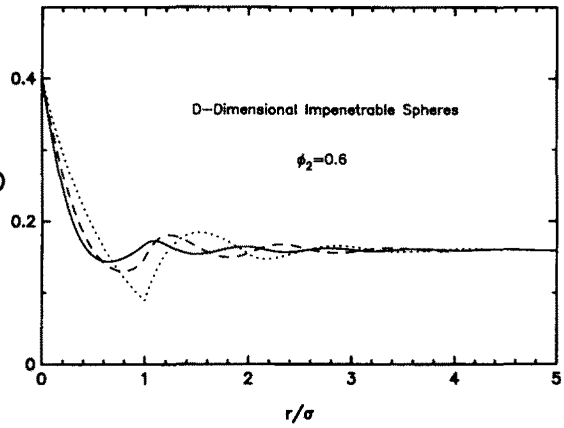

FIG. 2. The two-point probability function $S_2(r)$ for equilibrium distributions of D-dimensional totally impenetrable spheres (Refs. 12, 13) at $\phi_2 = 0.6$: hard rods (....), hard circular disks (---) and hard spheres (—).

In Fig. 2, we depict $S_2(r)$ for hard D-dimensional spheres[12,13] at $\phi_2 = 0.6$ for $D=1$, 2, and 3. Increasing the dimensionality, for small r, decreases S_2. For large r, increasing D decreases the amplitude of the oscillations.

3. Penetrable-concentric-shell model

An interesting model which lies between the two extremes of fully penetrable and totally impenetrable particles is the penetrable-concentric-shell model.[14] In this model, the particles possess a small hard core encompassed by a perfectly penetrable concentric shell. The size of the internal hard core is assumed to be proportional to an impenetrability index λ, $0 \leqslant \lambda \leqslant 1$, with $\lambda = 0$ and $\lambda = 1$ corresponding to fully penetrable and impenetrable particles, respectively. The penetrable-concentric shell model enables one to continuously change the degree of overlap, and hence the degree of connectedness of the particle phase. The two-point probability function for disks in the penetrable-concentric-shell model has been computed by Smith and Torquato.[15]

C. Explicit relation for C for a class of anisotropic media

Using the results of the previous two subsections, we can obtain an explicit relation for the coarseness of the wide class of anisotropic media described in Sec. III B, for arbitrary-shaped observation regions. Recall that this class of media contains all statistically anisotropic distributions of identical, oriented inclusions of arbitrary shape. Substituting Eq. (3.17) with $n=2$ into the general coarseness formula (3.6) yields, we substitute Eq. (3.17) with $n=2$ into the general coarseness formula (3.6) yields

$$C=\left[\frac{1-\phi_1^2}{\phi_1^2}-\frac{\rho}{\phi_1^2 V_o^2}\int V_2(\mathbf{r};\sigma)V_2^{\text{int}}(\mathbf{r};\sigma_o)d\mathbf{r} + \frac{1}{\phi_1^2 V_o^2}\int \sum_{k=2}^{\infty} S_2^{(k)}(\mathbf{r})V_2^{\text{int}}(\mathbf{r};\sigma_o)(\mathbf{r})d\mathbf{r}\right]^{1/2}, \tag{3.26}$$

where

$$S_2^{(k)}(\mathbf{x}_{12})=\frac{(-1)^k\rho^k}{k!}\int\cdots\int g_k(\mathbf{r}^k) \times \prod_{j=1}^{k}\left[1-\prod_{i=1}^{2}\left[1-m(\mathbf{x}_i-\mathbf{r}_j)\right]\right]d\mathbf{r}_j. \tag{3.27}$$

Note that anisotropic structural information enters through the terms involving two and higher-body effects, i.e., in the sum of Eq. (3.26). Equation (3.26) is valid for distributions of identical particles whose positions are completely specified by center-of-mass coordinates. It is important to emphasize that the interparticle interactions that one can consider are perfectly general; hence, the particles may partially overlap one another, interact through repulsive forces (e.g., impenetrable cores and Coulombic forces) as well as attractive forces, etc.

IV. RESULTS FOR THE COARSENESS OF DISTRIBUTIONS OF *D*-DIMENSIONAL SPHERES

The results of Sec. III are applied here to obtain explicit relations for the coarseness of distributions of identical D-dimensional spheres of diameter σ, in both the fully penetrable particle and totally impenetrable particle models. A D-dimensional spherical observation region of diameter σ_o is employed. These coarseness expressions are then computed for various values of the particle volume fraction ϕ_2 and the observation volume V_O.

A. Coarseness expressions for *D*-dimensional spheres and observation regions

1. D-dimensional fully penetrable spheres

For the case of D-dimensional fully penetrable spheres, the coarseness formula, for a D-dimensional spherical observation region of diameter σ_o, is easily obtained by substituting Eq. (3.19) into Eq. (3.6). After some simplication, one finds

$$C=\frac{\sqrt{2}\pi^{D/4}}{V_O\sqrt{\Gamma(D/2)}} \times\left\{\int_0^{\sigma_o}\left[\exp[\rho V_2^{\text{int}}(r;\sigma)]-1\right]V_2^{\text{int}}(r;\sigma_o)r^D\,dr\right\}^{1/2}. \tag{4.1}$$

Here $V_2^{\text{int}}(r;\sigma_o)$ is given by relations (3.21)–(3.23). Expression (4.1) is the D-dimensional generalization of the result obtained by Bayer[6] for two-dimensional distributions of penetrable disks.

2. D-dimensional totally impenetrable spheres

In the instance of an isotropic distribution of D-dimensional totally impenetrable spheres of diameter σ and spherical observation region of diameter σ_0, Eq. (3.26) yields

$$C=\left[\frac{1-\phi_1^2}{\phi_1^2}-\frac{\rho}{\phi_1^2 V_O^2}\int V_2(r;\sigma)V_2^{\rm int}(r;\sigma_o)d\mathbf{r}+\frac{1}{\phi_1^2 V_O^2}\int S_2^{(2)}(r)V_2^{\rm int}(r;\sigma_o)d\mathbf{r}\right]^{1/2}, \quad (4.2)$$

where

$$S_2^{(2)}(r_{12})=\rho^2\int\int g_2(r_{34})m(r_{13})m(r_{14})d\mathbf{r}_3\,d\mathbf{r}_4. \quad (4.3)$$

The particle indicator function for D-dimensional spheres is simply given by

$$m(r)=\begin{cases}1, & r\leqslant\sigma/2\\ 0, & r>\sigma/2\end{cases}. \quad (4.4)$$

B. Calculations of the coarseness for D-dimensional fully penetrable and impenetrable spheres

We now calculate the coarseness C for D-dimensional distributions of fully penetrable and totally impenetrable spheres of diameter σ for selected values of the sphere volume fraction ϕ_2 and observation volume V_O. In the case of fully penetrable particles, the integral of Eq. (4.1) is numerically evaluated using a trapezoidal rule. In the instance of impenetrable particles, the second integral of Eq. (4.2) is also computed employing a trapezoidal rule. Here the equilibrium result for the two-point probability function $S_2(r)$ of hard rods,[12] hard circular disks,[12] and hard spheres[13] are utilized.

In Fig. 3 we display the coarseness for penetrable and impenetrable rods ($D=1$) as a function of the ϕ_2 for three values of the scaled observation volume V_O/V_1. In Figs. 4 and 5 we depict corresponding results for $D=2$ and $D=3$, respectively. It is seen that for fixed D and volume fraction V_O/V_1, the coarseness is always smaller for impenetrable particles than for the penetrable particles at the same value of ϕ_2. This effect becomes more pronounced at higher values of the particle volume fraction. Physically speaking, this is true because exclusion-volume effects associated with the impenetrable particles results in a distribution which is less "random" than that with randomly centered particles. For fixed D and V_O/V_1, the coarseness for fully penetrable particles is a monotonically decreasing function of ϕ_2. This functional dependence is in contrast to that of the impenetrable-particle model in which C first increases with increasing ϕ_2 for small to moderate ϕ_2, reaches some maximum value, and then decreases with increasing ϕ_2. Moreover, C decreases with increasing volume ratio V_O/V_1 for either penetrable or impenetrable particles. Finally, from Figs. 2–4, it is obvious that for either particle model, increasing the dimensionality has the effect of decreasing the coarseness since the particle coverage is more uniform in higher dimensions.

In Fig. 6 we depict the quantity $C(V_O/V_1)^{1/2}$ as a

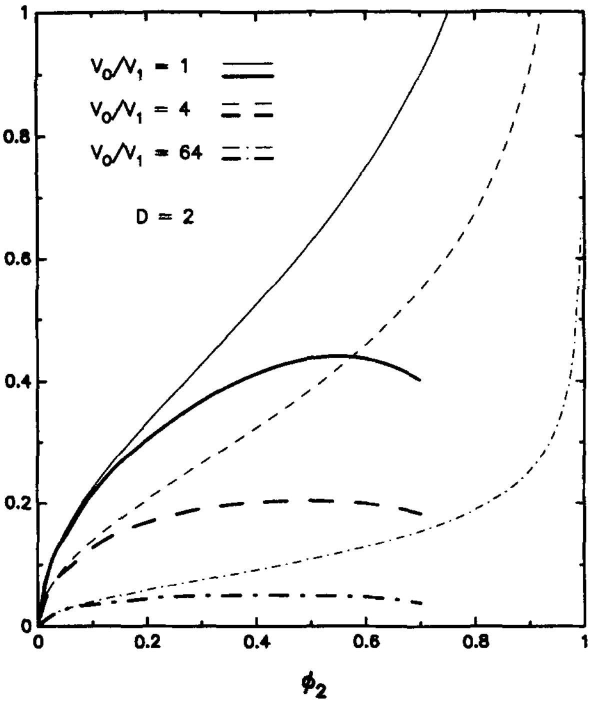

FIG. 4. As in Fig. 3 for the case of two-dimensional distributions of circular disks of diameter σ. $V_O=\pi\sigma_o^2/4$ and $V_1=\pi\sigma^2/4$.

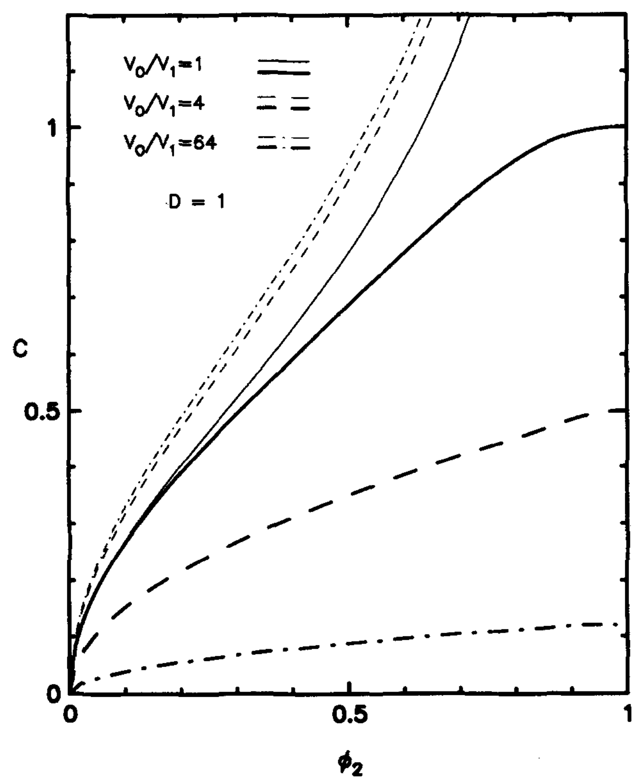

FIG. 3. The coarseness C vs the particle volume fraction ϕ_2 for three different values of the scaled observation-region volume V_O/V_1 for one-dimensional distribution of fully penetrable rods (lighter curves) and totally impenetrable rods (heavier curves) of diameter σ. V_O is the observation volume equal to σ_o and V_1 is the particle volume equal to σ.

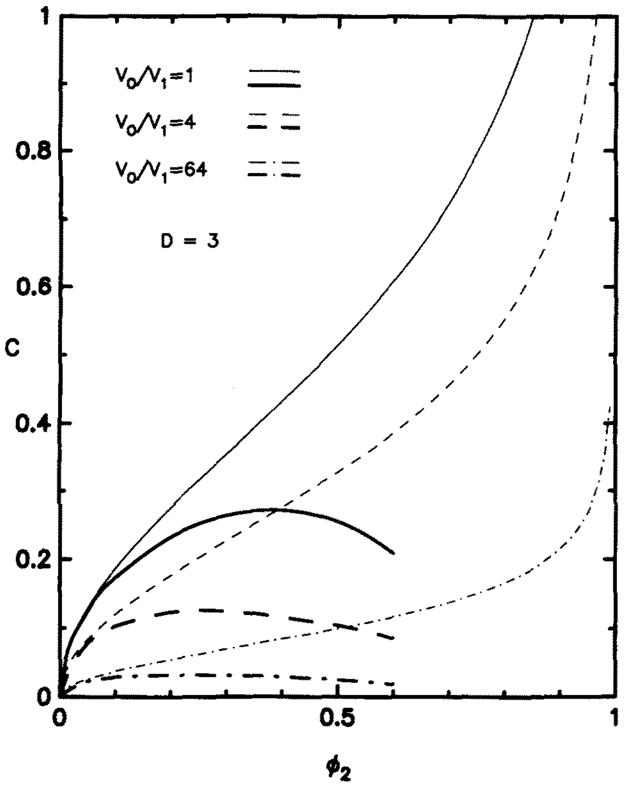

FIG. 5. As in Fig. 3 for the case of three-dimensional distributions of circular disks of diameter σ. $V_O = \pi\sigma_O^3/6$ and $V_1 = \pi\sigma^3/6$.

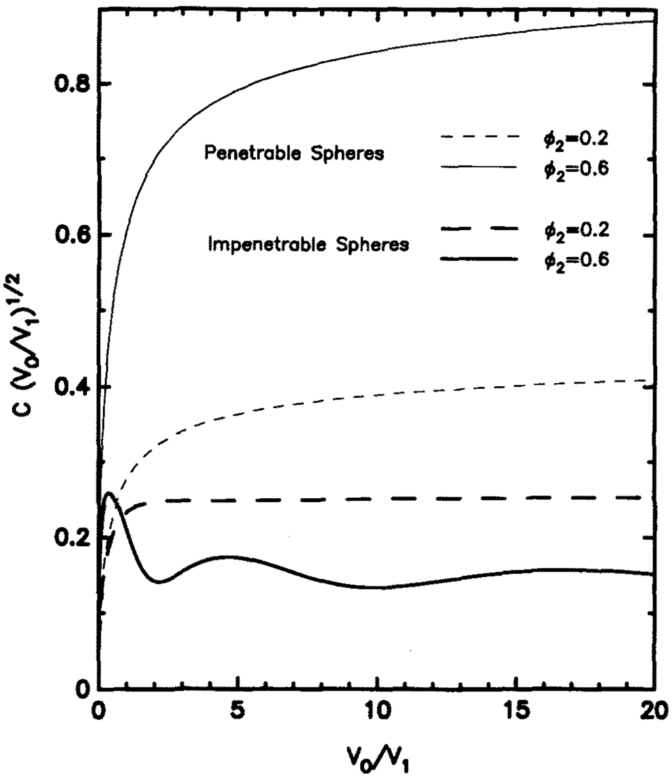

FIG. 6. The coarseness C multiplied by $(V_O/V_1)^{1/2}$ as a function of the scaled observation volume V_O/V_1 for both three-dimensional fully penetrable (lighter curves) and impenetrable spheres (heavier curves) at $\phi_2 = 0.2$ and 0.6. $V_O = \pi\sigma_O^3/6$ and $V_1 = \pi\sigma^3/6$.

function of the volume ratio V_O/V_1 for both models with $D=3$ for $\phi_2 = 0.2$ and $\phi_2 = 0.6$. Recall that for large observation regions $C \sim V_O^{-1/2}$ [cf. Eq. (3.8)], and thus $C(V_O/V_1)^{1/2}$ approaches the constant $KV_1^{-1/2}$ [where K given by Eq. (3.9)], for large V_O. At $\phi_2 = 0.6$, $C(V_O/V_1)^{1/2}$ for penetrable spheres is generally considerably larger than the corresponding quantity for impenetrable spheres. Note that at $\phi_2 = 0.6$ for impenetrable spheres, $C(V_O/V_1)^{1/2}$ oscillates about its long-range value of approximately 0.15 due to exclusion-volume effects. Oscillations begin to become noticeable for impenetrable spheres at $\phi_2 = 0.4$ (not shown). Observe also that for the same model at $\phi_2 = 0.2$, the long-range value of $C(V_O/V_1)^{1/2}$ is achieved for considerably smaller values of the volume ratio ($V_O/V_1 \geqslant 2$). The same general trends described here for $D=3$ are observed for arbitrary D.

V. CONCLUSIONS

In this paper we have defined the *coarseness C* which gives a measure of local volume fraction fluctuations in heterogeneous media. This definition provides a means of obtaining C either experimentally or theoretically. The coarseness, for arbitrary D-dimensional two-phase anisotropic media and observation region, is shown to be related to an integral involving the two-point probability function. Using this general relation, we then obtained an explicit relation for the coarseness of anisotropic media composed of identical particles of arbitrary shape whose positions are completely specified by their center-of mass coordinates (ellipses, rectangles, ellipsoids, cylinders, etc.). For concreteness, C was computed for D-dimensional distributions of penetrable as well as impenetrable spheres (with $D=1$, 2, and 3), for a wide range of particle densities and observation-region sizes. The effect of impenetrability of the particles, for fixed observation-region size, is to reduce the coarseness relative to that of the penetrable-particle model, especially at high densities. The effect of increasing the dimensionality is to decrease the coarseness.

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