Chord-length distribution function for two-phase random media

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(Received 28 February 1992; revised manuscript received 13 January 1993)

A statistical correlation function of basic importance in the study of two-phase random media (such as suspensions, porous media, and composites) is the chord-length distribution function $p(z)$. We show that $p(z)$ is related to another fundamentally important morphological descriptor studied by us previously, namely, the lineal-path function $L(z)$, which gives the probability of finding a line segment of length z wholly in one of the phases when randomly thrown into the sample. We derive exact series representations of the chord-length distribution function for media comprised of spheres with a polydispersivity in size for arbitrary space dimension D. For the special case of spatially uncorrelated spheres (i.e., fully penetrable spheres), we determine exactly $p(z)$ and the mean chord length l_C, the first moment of $p(z)$. We also obtain corresponding formulas for the case of impenetrable (i.e., spatially correlated) polydispersed spheres.

PACS number(s): 47.55.Mh, 05.20.−y, 61.20.Gy

The characterization of the microstructure of two-phase random media, such as suspensions, composites, and porous media, is of great fundamental as well as practical importance [1–11]. The goal ultimately is to ascertain what is the essential morphological information, quantify it either theoretically or experimentally, and then employ the information to estimate the desired macroscopic properties of the heterogeneous material.

In this Brief Report, we concern ourselves with the so-called *chord-length distribution function* $p(z)$. Specifically, $p(z)dz$ is the probability of finding a chord of length between z and $z+dz$ in one of the phases, say phase 1. Chords are distributions of lengths between intersections of lines with the two-phase interface (see Fig. 1). Knowledge of the chord-length distribution function is of basic importance in transport problems involving "discrete free paths" and thus has an application in Knudsen diffusion and radiative transport in porous media [12–15]. The function $p(z)$ has also been measured for sedimentary rocks [16] for the purpose of studying fluid flow through such porous media. The chord-length distribution function $p(z)$ is also a quantity of great interest in stereology [11]. For example, the *mean chord (or intercept) length* l_C is the first moment of $p(z)$.

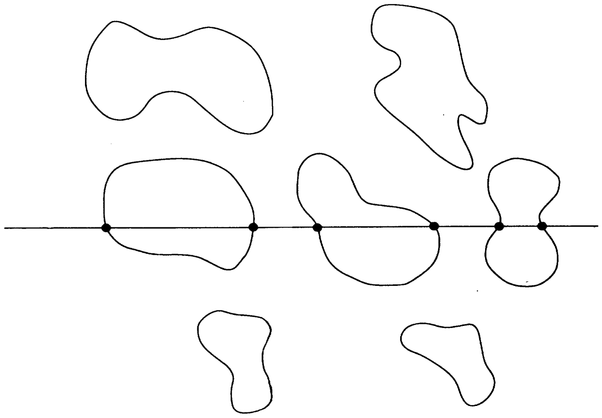

FIG. 1. Schematic of chord-length measurements for a cross section of a two-phase random medium. The chords are defined by the intersection of lines with the two-phase interface.

We first show that $p(z)$ is related to another important morphological descriptor of random media studied by us earlier [17,18], namely, the *lineal-path function* $L(z)$ which gives the probability of finding a line segment of length z wholly in phase 1 when randomly thrown into the sample. For heterogeneous media composed of spheres with a polydispersivity in size, we find an exact series representation of $p(z)$. In the special case of fully penetrable (i.e., spatially uncorrelated) spheres, we determine exact expressions for $p(z)$ and l_C. Corresponding analytical formulas are also obtained for impenetrable polydispersed spheres.

The lineal-path function $L(z)$ can be obtained by counting the relative number of times that a line segment of length z is wholly in phase 1 when thrown randomly onto an infinite line in the system. Clearly, the line segment (of length z) being wholly in phase 1 implies that all the points on the line segment (of length z) are in phase 1. The strategy now will be to express $L(z)$ in terms of $p(z)$ using the following probability argument. First, if we consider a special point on the line segment, say, the midpoint of the line segment referred as point A, then A has to be in phase 1. The probability that point A is in phase 1 is simply the porosity of the system, i.e., ϕ_1. Second, given the condition that point A is in phase 1 (it is then on a chord), we ask what is the probability that point A is on a chord with length between y and $y+dy$? Since the *length fraction* of a chord with length between y and $y+dy$ is given by

$$yp(y)dy \Big/ \int_0^\infty yp(y)dy \ ,$$

then the probability that point A is on a chord with length between y and $y+dy$ is this length fraction multiplied by the porosity ϕ_1, i.e.,

$$\phi_1 yp(y)dy \Big/ \int_0^\infty yp(y)dy \ .$$

Third, that point A of a line segment of length z *(distinct from length y)* is in phase 1, however, does not mean that the whole line segment is in phase 1. The probability that a line segment of length z is on a chord of length y under the condition that point A is on that chord is

$$(y-z)H(y-z)/y \ ,$$

where $H(x)$ is the Heaviside step function [i.e., $H(x)=1$ for $x>0$ and $H(x)=0$ otherwise]. Now, $L(z)$, the probability that the line segment of length z is entirely in phase 1, can then be obtained by combining the results given immediately above, i.e., integrating the probability for the line segment being on chords with length between y and $y+dy$ over all possible y, we find

$$L(z)=\frac{\phi_1\int_0^\infty (y-z)p(y)dyH(y-z)}{\int_0^\infty yp(y)dy} \ . \tag{1}$$

Differentiation of (1) yields

$$\frac{dL(z)}{dz}=-\frac{\phi_1}{l_C}\int_z^\infty p(y)dy \ . \tag{2}$$

Differentiation of (2) and rearrangement of terms gives

$$p(z)=\frac{l_C}{\phi_1}\frac{d^2L(z)}{dz^2} \ , \tag{3}$$

where l_C is the mean chord length given by

$$l_C=\int_0^\infty zp(z)dz \ . \tag{4}$$

Formula (3) establishes a new connection between chord-length distribution function $p(z)$ and the lineal-path function $L(z)$. It is important to note that the above relations are valid for statistically isotropic systems of arbitrary microgeometry.

Assume now that the system is composed of included particles with a continuous distribution of radius $\mathcal{R}$ characterized by the normalized probability density $f(\mathcal{R})$. This function includes the discrete particle size distribution as a special case. For example, in the discrete case with M different components, the size distribution $f(\mathcal{R}_j)=\sum_{\sigma=1}^{M}(\rho\sigma/\rho)\delta(\mathcal{R}_j-R_{\sigma_j})$, where ρ_σ is the number density of *type-σ* particles and $\delta(\mathcal{R})$ is the Dirac δ function. The system is characterized by the probability density function $\rho_n(\mathbf{r}^n;\mathcal{R}_1,\ldots,\mathcal{R}_n)f(\mathcal{R}_1)\cdots f(\mathcal{R}_n)$ associated with finding an inclusion with radius $\mathcal{R}_1$ at $\mathbf{r}_1$, another inclusion with radius $\mathcal{R}_2$ at $\mathbf{r}_2$, etc. [19]. In the instance of statistically homogeneous media, $\rho_1(\mathbf{r}_1;\mathcal{R}_1)$ is simply equal to the *total* number of density ρ. The reduced density η in D dimensions in the discrete case is defined by

$$\eta=\sum_{\sigma=1}^{M}\frac{\pi^{D/2}}{\Gamma\left[1+\frac{D}{2}\right]}\rho_\sigma R_\sigma^D \ , \tag{5}$$

where $\Gamma(x)$ is the gamma function. In the continuous case, we have

$$\eta=\frac{\pi^{D/2}}{\Gamma(1+D/2)}\rho\langle\mathcal{R}^D\rangle \ , \tag{6}$$

where the average of any function $A(\mathcal{R})$ is given by

$$\langle A(\mathcal{R})\rangle=\int_0^\infty A(\mathcal{R})f(\mathcal{R})d\mathcal{R} \ . \tag{7}$$

Only in the case of hard spheres in η equal to the sphere volume fraction ϕ_2. For penetrable-sphere systems, $\phi_2=1-\phi_1=\exp(-\eta)$ and thus $\eta\geq\phi_2$.

Lu and Torquato [17,18] have shown that the lineal-path function $L(z)$ can be represented by the following exact series:

$$L(z)=1+\sum_{k=1}^{\infty}\frac{(-1)^k}{k!}\int d\mathcal{R}_1\cdots d\mathcal{R}_k\rho_k(\mathbf{r}^k;\mathcal{R}_1,\ldots,\mathcal{R}_k)f(\mathcal{R}_1)\cdots f(\mathcal{R}_k)\prod_{j=1}^{k}m_j(x-\mathbf{r}_j;z)d\mathbf{r}_j \ , \tag{8}$$

where

$$m_j(\mathbf{y};z)=\begin{cases}1 \ , & \mathbf{y}\in\Omega_E(z,\mathcal{R}_j)\\ 0 \ , & \text{otherwise}\end{cases} \tag{9}$$

and $\Omega_E(z,\mathcal{R}_j)$ is the "exclusion" region which is a spherocylinder of cylindrical length z and radius $\mathcal{R}_j$ with hemispherical caps of radius $\mathcal{R}_j$. Here $\mathbf{y}$ is measured with respect to the centroid of Ω_E.

For fully penetrable spheres, the n-particle probability densities are trivial (i.e., $\rho_n=\rho^n$) and hence we find the chord-length distribution function is exactly given by

$$p(z)=\begin{cases}-\frac{3}{4}\ln\phi_1\langle\mathcal{R}^2\rangle/\langle\mathcal{R}^3\rangle\phi_1^{3\langle\mathcal{R}^2\rangle z/\langle 4\mathcal{R}^3\rangle} \ , & D=3 \quad (10)\\ -2\ln\phi_1\langle\mathcal{R}\rangle/(\pi\langle\mathcal{R}^2\rangle)\phi_1^{2\langle\mathcal{R}\rangle z/(\pi\langle\mathcal{R}^2\rangle)} \ , & D=2 \quad (11)\\ -\ln\phi_1/\langle\mathcal{R}\rangle\phi_1^{z/\langle\mathcal{R}\rangle} \ , & D=1 \ . \quad (12)\end{cases}$$

where ϕ_1 is the volume fraction of phase 1. The mean chord length l_C for fully penetrable spherical systems is obtained by applying formula (4). We find that

$$l_C=\begin{cases}-4\langle\mathcal{R}^3\rangle/(3\langle\mathcal{R}^2\rangle\ln\phi_1)\,, & D=3 \quad (13)\\ -\pi\langle\mathcal{R}^2\rangle/(2\langle\mathcal{R}\rangle\ln\phi_1)\,, & D=2 \quad (14)\\ -\langle\mathcal{R}\rangle/\ln\phi_1\,, & D=1\,. \quad (15)\end{cases}$$

Note that for $D=1$, polydispersivity has no effect on $p(z)$ or l_C for fixed $\langle\mathcal{R}\rangle$.

In the instance of totally impenetrable or hard spheres, the exact series representation of $L(z)$ and, thus, $p(z)$ can only be evaluated exactly for the case $D=1$ (i.e., hard rods). It is impossible to evaluate the series for $D\geq 2$ exactly because the n-particle probability densities $\rho_n(\mathbf{r}^n)$ are not known exactly. One must therefore devise approximate schemes to evaluate and sum the series. Lu and Torquato [18] evaluated $L(z)$ for polydispersed hard-sphere systems in a certain accurate approximation. Using this result and expression (3) yields the chord-length distribution functions $p(z)$ as

$$p(z)=\begin{cases}\dfrac{\pi\rho\langle\mathcal{R}^2\rangle}{1-\eta}\exp\left\{-\pi\rho z\dfrac{\langle\mathcal{R}^2\rangle}{1-\eta}\right\}, & D=3 \quad (16)\\[2ex] \dfrac{2\rho\langle\mathcal{R}\rangle}{1-\eta}\exp\left[\dfrac{2\rho z\langle\mathcal{R}\rangle}{1-\eta}\right], & D=2 \quad (17)\\[2ex] \dfrac{\rho z}{1-\eta}\exp\left[\dfrac{-\rho z}{1-\eta}\right], & D=1\,. \quad (18)\end{cases}$$

The corresponding explicit expressions for the mean chord lengths are given by

$$l_C=\begin{cases}\dfrac{1}{\pi\rho}\dfrac{1-\eta}{\langle\mathcal{R}^2\rangle}\,, & D=3 \quad (19)\\[2ex] \dfrac{1}{2\rho}\dfrac{1-\eta}{\langle\mathcal{R}\rangle}\,, & D=2 \quad (20)\\[2ex] \dfrac{1-\eta}{\rho}\,, & D=1\,. \quad (21)\end{cases}$$

A commonly employed size distribution function $f(\mathcal{R})$ is the Schulz distribution function and is given by

$$f(\mathcal{R})=\frac{1}{\Gamma(m+1)}\left[\frac{m+1}{\langle\mathcal{R}\rangle}\right]^{m+1}\mathcal{R}^m \times\exp\left[\frac{-(m+1)\mathcal{R}}{\langle\mathcal{R}\rangle}\right], \quad m>-1 \qquad (22)$$

The nth moment of the Schulz distribution function is

$$\langle\mathcal{R}^n\rangle=\langle\mathcal{R}\rangle^n\frac{(m+1)^{-n}}{m}\prod_{i=0}^{n}(m+i)\,. \qquad (23)$$

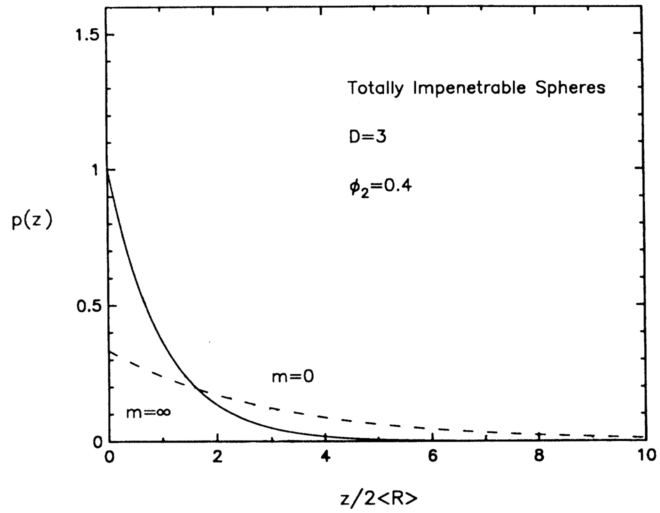

FIG. 2. Chord-length distribution function $p(z)$ vs dimensionless distance $z/2\langle\mathcal{R}\rangle$ for a three-dimensional totally impenetrable polydispersed system characterized by a Schulz distribution (22) with $m=\infty$ (solid line) and $m=0$ (dashed line) at a sphere volume fraction of $\phi_2=0.4$ as obtained from (16).

By increasing m, the variance decreases, i.e., the distribution becomes sharper. In the monodisperse limit, $z\to\infty$, $f(\mathcal{R})=\delta(\mathcal{R}-\langle\mathcal{R}\rangle)$. Note that for homogeneous and isotropic media, the density of the particles with radius between $\mathcal{R}$ and $\mathcal{R}+d\mathcal{R}$ is $\rho f(\mathcal{R})d\mathcal{R}$ with ρ the total density.

To illustrate the results given above, we consider polydispersed-sphere systems characterized by a Schulz distribution (22). In Fig. 2, we plot our analytical results of $p(z)$ for totally impenetrable polydispersed systems characterized by a Schulz distribution with $m=0$ and $m=\infty$ at the sphere volume fraction $\phi_2=0.4$ for $D=3$

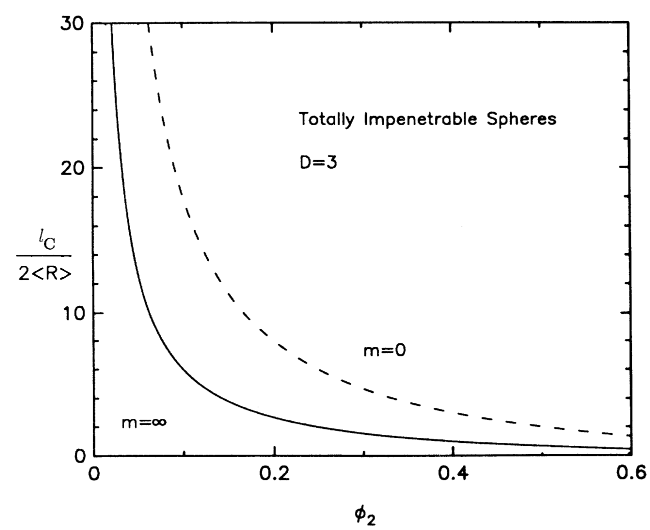

FIG. 3. Mean chord length l_C vs the sphere volume fraction ϕ_2 for a three-dimensional totally impenetrable polydispersed system characterized by a Schulz distribution (22) with $m=\infty$ (solid line) and $m=0$ (dashed line) as obtained from (19).

[Eq. (16)]. As expected, $p(z)$ is a monotonically decreasing function of z. The figure shows that the effect of increasing the degree of polydispersivity is to broaden the distribution function $p(z)$, i.e., increasing polydispersivity decreases $p(z)$ for small z but increases $p(z)$ for large z. The same general trends are found for $D=2$. In Fig. 3, we depict the analytical results for the mean chord length [Eq. (21)] for totally impenetrable spheres for $D=3$ at a sphere volume fraction $\phi_2=0.4$ with $m=0$ and $m=\infty$.

What is the effect of increasing the degree of particle *penetrability* on $p(z)$? If one compares expressions (10) and (11) for fully penetrable spheres to relations (16) and (17) for impenetrable spheres, one sees that increasing penetrability, at fixed volume fraction, broadens $p(z)$. Otherwise, the behavior of $p(z)$ for these two models are qualitatively similar.

Elsewhere, [20] we study a more general lineal-path function $L(z,a)$ associated with the *space available* to a spherical "test" particle of radius a which is inserted or diffusing in the two-phase random medium. The corresponding chord-length distribution, termed the *free-path distribution function* $p(z,a)$, is also examined.

The authors gratefully acknowledge the support of the Office of Basic Energy Science, U.S. Department of Energy under Grant No. DE-FG02-92ER14275.

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