ESTIMATING EFFECTIVE MODULI OF COMPOSITES USING QUANTITATIVE IMAGE ANALYSIS

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Abstract. The statistical data required for quantitative analysis of the physical properties of composite materials may be obtained using digital image processing techniques. This approach allows accurate quantitative determination of volume fraction data, of interface area per unit volume, and of more sophisticated measures of the microgeometry of material mixtures including the n-point spatial correlation functions. The two- and three-point correlation functions can be measured quite accurately both for synthetic materials and for real materials such as porous sandstones. To extract the desired parameters of the microgeometry from the three-point correlation functions, methods of interpolating and integrating these lattice-based empirical values have been developed. Methods of finessing the theory to give estimates of physical properties when the available data are not adequate to permit use of the full theory have also been developed.

1. Introduction. Various methods of estimating effective properties of composite materials require geometrical or topological information contained in statistical correlation functions. If the three-point correlation functions are measured using digital image processing methods [1], the values are computed for a discrete set of admissible triangular arguments corresponding to triangles whose vertices are commensurate with a simple cubic lattice. To extract the desired information about microgeometry from the correlation functions, methods of interpolating and integrating between these lattice-based values have been developed [2].

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Whereas the variational bounds derived by Hashin and Shtrikman [3] for elastic constants depend only on the volume fractions and the moduli of the constituents of the composite, nearly all of the improved bounds on physical properties require additional information about the microstructure of the composite. Perhaps the most common method of quantifying this microstructural information comes in the form of the n-point spatial correlation functions. These functions were introduced into this problem by Brown [4] in his early work on obtaining estimates of effective constants in series form. Debye *et al.* [5] used the angular distribution of scattered x-rays to measure the two-point spatial correlation function and estimate the specific (internal) surface area of porous materials. Prager [6] used these same functions in his early formulation of variational bounds for the fluid permeability of porous media. Beran [7] also needed them in his variational approach for finding bounds on the dielectric constant. Theoretical results that require the information contained in the correlation functions have subsequently been obtained by many authors over a period of many years [8-16], and we may expect more applications will be found in the future. It is therefore clear that methods of measuring these functions are needed in order to make these theoretical results accessible to experimentalists and engineers interested in designing new materials. Corson [13] was the first to try to measure the three-point correlation function, but — with the advent of modern image processing techniques and faster computers — better techniques are available to us now [1,17,18] than were available to him then.

In this paper we will define the correlation functions in Section 2 and present some of their more important general properties. Practical approximation methods are also discussed. In Section 3 we discuss the parameters of microgeometry that are commonly encountered in studies of electrical, thermal, and elastic composites. To provide some new insight into the structure of the correlation functions, we present a new method of visualizing the three-point correlation functions in Section 4.

2. Spatial correlation functions. A discussion of the significance of the spatial correlation functions has been presented by Torquato and Stell [14] and a detailed analysis of methods for obtaining these functions experimentally using image processing techniques was presented in [1]. We will not repeat these detailed discussions here, but it is still necessary to define the correlation functions and mention their relevant properties. The analysis presented is limited to two-phase composite media.

Let $p(\vec{x})$ be the value of some property of a random composite material (e.g., electrical or thermal conductivity, dielectric constant, bulk or shear modulus, etc.) which assumes one of two values p_0 or p_1 depending on whether $\vec{x}$ is located in a grain of material 0 or material 1. Define the indicator or characteristic function

$$f(\vec{x}) = \frac{p(\vec{x}) - p_0}{p_1 - p_0}. \tag{1}$$

Then $f(\vec{x}) = 0$ in material 0 and $f(\vec{x}) = 1$ in material 1. For example, in a porous medium we may arbitrarily label all solid regions as material 0 and all void regions as material 1. Since complete knowledge of the stochastic variable f is seldom available, our interest in the characteristic function is generally limited to a few

Figure 1. Pseudo-gray scale image of a cross section of a glass bead sample obtained by digitizing a raw scanning electron microscope image. Dark areas are solid and light areas are epoxy that has been injected into the pore space. This image must be registered (ones or zeroes must be assigned to every pixel) before the spatial correlation functions are computed. To establish the scale of this image, the apparent diameter of the largest particles is approximately 100μ.

of its statistical properties. If chosen properly, these quantities are often sufficient to provide the data needed for variational bounds on the macroscopic average of the property being studied [1]. Figure 1 shows an example of a scanning electron microscope image of a cross section of a real material (a glass bead sample) prior to registration.

The first three spatial correlation functions are defined by

$$\hat{S}_1 =< f(\vec{x}) >= \phi, \tag{2}$$

$$\hat{S}_2(\vec{r}_1, \vec{r}_2) =< f(\vec{x}+\vec{r}_1)f(\vec{x}+\vec{r}_2) >, \tag{3}$$

and

$$\hat{S}_3(\vec{r}_1, \vec{r}_2, \vec{r}_3) =< f(\vec{x}+\vec{r}_1)f(\vec{x}+\vec{r}_2)f(\vec{x}+\vec{r}_3) > . \tag{4}$$

The brackets $< \cdot >$ indicate a volume average over the spatial coordinate $\vec{x}$. The volume fraction of constituent 1 is given by ϕ. We will refer to the three correlation functions defined this way as the one-, two-, and three-point correlation functions, respectively. Since two points lie along a line and three points lie in a plane, the two-point and three-point correlations (as well as the one-point correlation) may be measured by processing digital images of material cross sections. In the present paper, we will stress the three-point correlation functions. In general, we assume that the composite medium of interest is statistically homogeneous so that on average only the differences in the coordinate values are significant (translational invariance). Furthermore, we often assume that the material is at least locally isotropic so that the averages do not depend on orientation of the arguments. (An example of local isotropy in conjunction with global anisotropy is a transversely isotropic material composed of thin layers of isotropic materials. Such a material can be treated if the correlation functions are determined by examining cross sections taken perpendicular to the axis of symmetry.) With these assumptions, we find that the two-point correlation function simplifies to

$$\hat{S}_2(\vec{r}_1, \vec{r}_2) = \tilde{S}_2(\vec{r}_{12}) = S_2(r_{12}), \tag{5}$$

while the three-point correlation function satisfies

$$\hat{S}_3(\vec{r}_1, \vec{r}_2, \vec{r}_3) = \tilde{S}_3(\vec{r}_{12}, \vec{r}_{13}) = S_3(r_{12}, r_{13}, \mu_{12,13}), \tag{6}$$

where

$$\vec{r}_{ij} = \vec{r}_j - \vec{r}_i, \qquad r_{ij} = \mid \vec{r}_{ij} \mid,$$

and

$$\mu_{ij,ik} = cos\,\theta = \vec{r}_{ij} \cdot \vec{r}_{ik} / r_{ij} r_{ik}.$$

The two-point correlation $S_2(r)$ is the probability that two points a distance $r = r_{12}$ apart are both in material 1. The three-point correlation function $S_3(r, s, \mu)$ is the probability that all three vertices of the triangle determined by (r, s, μ) lie in material 1.

In a random material possessing no long range order, we have

$$\lim_{s fixed, r \to \infty} S_3(r, s, \mu) = \phi S_2(s) \tag{7}$$

and

$$\lim_{r,s \to \infty} S_3(r, s, \mu) = \phi^3, \tag{8}$$

assuming that $\mu \neq 1$ or that the difference $r - s$ is not fixed if $\mu = 1$. The most important property of S_3 for statistically homogeneous and isotropic media is the fact that the value depends only on the size and shape of the triangle formed by its arguments, not on the orientation of that triangle. Thus,

$$S_3(r_{12}, r_{13}, \mu_{12,13}) = S_3(r_{21}, r_{23}, \mu_{21,23}) = S_3(r_{31}, r_{32}, \mu_{31,32}). \tag{9}$$

Furthermore, the order of the first two arguments may be freely interchanged. It is important to account for the symmetries (9) when designing a scheme to calculate, sort, and store the values of S_3, otherwise the stored values could be as much as sixfold degenerate. Such a scheme has been described in [2].

In general, these spatial correlation functions must be determined empirically. These one-, two-, and three-point correlation functions may be found using digital image processing techniques described previously [1]. Accurate values of S_3 are not difficult to obtain for small triangles whose vertices fit on the discrete lattice composed of the pixels in a digitized image (we call such triangles "lattice-commensurate"). However, there are several practical reasons why it is virtually impossible to obtain accurate values of the three-point correlation functions for large vertex separations: (*i*) The number of lattice-commensurate triangles of large size increases like the cube of the length of the longest side of the triangle (in pixel units), so storage of computed values of S_3 rapidly becomes difficult. (*ii*) As the triangle becomes large, fewer and fewer of them will fit on an image of fixed size, so the size of the statistical sample of S_3 for large arguments rapidly decreases and accuracy therefore suffers. Since the integrals used to define the parameters of the microgeometry are over triangles of all sizes, these practical limitations to our knowledge of S_3 must be accounted for in any image analysis scheme we devise.

Another significant practical issue arises due to the structure of the correlation function itself. Although the asymptotic value of $S_3(r,s,\mu)$ for large triangles is ϕ^3 as stated in (8), there are three exceptions and two are important: (*i*) If either $r=0$ or $s=0$, then the triangle collapses to a straight line and the value of the angle cosine μ is immaterial. In this situation,

$$S_3(0,s,\mu)=S_2(s) \qquad \text{or} \qquad S_3(r,0,\mu)=S_2(r). \tag{10}$$

However, since these results are independent of μ, we will see that (10) causes no problems in the integrals for the parameters of microgeometry. (*ii*) On the other hand, if $r=s$, then with $\mu=1$ the triangle again collapses to a straight line and we find

$$S_3(r,r,1)=S_2(r). \tag{11}$$

For large r, we find that S_3 approaches ϕ^2 instead of ϕ^3 for either (10) or (11), but (11) is more troublesome because it does depend on angle. (*iii*) Similarly, if $r\simeq s$ and r is large, then for any value of μ it is easy to see that

$$S_3(r,s,\mu)\simeq \phi S_2(t), \tag{12}$$

where $t=(r^2+s^2-2rs\mu)^{\frac{1}{2}}$ is the length of the third side of the triangle in argument of S_3. Eq. (12) follows from (4) by noting that if both r and s are large while t is small, then one of the three vertices of the triangle is far from the other two and therefore only weakly correlated with them. Thus, the distant point may be removed from the average in (4) and its contribution included through the factor ϕ in (12), *c.f.*, Eq. (7). Note that (12) is an approximation while (11) is exact. As $t\to 0$, we see that (12) smoothly approaches (11) only for such large values of r that $S_2(r)=\phi^2$.

To treat the special regions of S_3 noted in the last paragraph and to provide an approximate solution to the problem of finding accurate values of S_3 for large triangular arguments, we introduce the following approximation when at least one of the two sides r,s is large:

$$S_3(r,s,\mu)\simeq S_2(x)S_2(y)/\phi, \tag{13}$$

where x and y are the two shortest sides of the triangle whose three sides are r,s,t. When $r=0$, $x=r=0$ and $y=s$ so (13) gives the exact result (10). When $r=s$ and $\mu=1$, $x=t=0$ and $y=r$ so (13) gives the exact result (11). When $r\simeq s$ and $t<r$, $x=t$ and $y=$ the smaller of r and s (say r), then (13) gives

$$S_3(r,s,\mu)\simeq S_2(t)S_2(r)/\phi, \tag{14}$$

which reduces to (12) when r is large, as we have assumed it is.

Note that when $x = y = 0$, (13) gives the exact result $S_3(0, 0, \mu) = \phi$ and, when x and y are both large, (13) gives the correct asymptotic limit $S_3 \to \phi^3$. Thus, although (13) is an approximation, it does reproduce all the special limits we have discussed. Furthermore, the use of (13) for large arguments solves the problem of obtaining accurate values of S_3 for large arguments and also eliminates the storage problem, since S_2 is much easier to compute and store than S_3. Finally, note that the approximation (13) is superior to other approximations that have been used [19] such as $S_3(r, s, \mu) = S_2(r)S_2(s)/\phi$ which has no angular dependence (and therefore would produce nothing if used in the integrals) or such as $S_3(r, s, \mu) = S_2(r)(S_2(s) + S_2(t))/2\phi$ which does not satisfy all of the limiting conditions.

We will show an example later that illustrates the accuracy of the approximation (13) for large arguments.

3. Parameters of microgeometry. Milton [15,16] has introduced two parameters depending on the microgeometry of a composite through the three-point correlation function S_3. These two geometric parameters of interest for studies of electrical or thermal conductivities and elastic constants are

$$\zeta_1 = \lim_{\Delta\to 0} \lim_{\Delta'\to\infty} \frac{9}{2\phi_0\phi_1} \int_\Delta^{\Delta'} dr \int_\Delta^{\Delta'} ds \int_{-1}^{+1} d\mu \frac{S_3(r, s, \mu)}{rs} P_2(\mu) \tag{15}$$

and

$$\eta_1 = \frac{5\zeta_1}{21} + \lim_{\Delta\to 0} \lim_{\Delta'\to\infty} \frac{150}{7\phi_0\phi_1} \int_\Delta^{\Delta'} dr \int_\Delta^{\Delta'} ds \int_{-1}^{+1} d\mu \frac{S_3(r, s, \mu)}{rs} P_4(\mu), \tag{16}$$

where $\phi_1 = \phi$, $\phi_0 = 1 - \phi_1$, and $P_2(\mu)$ and $P_4(\mu)$ are the Legendre polynomials of order 2 and 4 given respectively by

$$P_2(\mu) = \frac{1}{2}(3\mu^2 - 1)$$

and

$$P_4(\mu) = \frac{1}{8}(35\mu^4 - 30\mu^2 + 3).$$

The complements of ζ_1 and η_1 are $\zeta_0 = 1 - \zeta_1$ and $\eta_0 = 1 - \eta_1$, respectively. All four of these constants lie in the range [0,1].

The integrands of both integrals (15) and (16) are singular for small values of r, s and poorly behaved for large values of r, s. The conditional convergence of the integrals requires that the integration over μ be performed prior to the integrals over r and s. The singularity at the origin may be handled easily by standard methods [20]. The exact relations (10) guarantee that triangles with at least one side of vanishing length have no angular dependence and therefore no contribution to these integrals.

Recalling the important fact (12), we can improve the rate of convergence at large values of the independent variables r and s by adding and subtracting a term that can be integrated analytically [16] giving

$$\zeta_1 = \phi_1 + \lim_{\Delta\to 0} \lim_{\Delta'\to\infty} \frac{9}{2\phi_0\phi_1} \int_\Delta^{\Delta'} dr \int_\Delta^{\Delta'} ds \int_{-1}^{+1} d\mu \frac{S_3(r, s, \mu) - \phi_1 S_2(t)}{rs} P_2(\mu) \tag{17}$$

and

$$\eta_1 = (5\zeta_1 + 16\phi_1)/21 + \lim_{\Delta\to 0}\lim_{\Delta'\to\infty} \frac{150}{7\phi_0\phi_1}\int_\Delta^{\Delta'} dr \int_\Delta^{\Delta'} ds \int_{-1}^{+1} d\mu \frac{S_3(r,s,\mu) - \phi_1 S_2(t)}{rs} P_4(\mu), \quad (18)$$

where $t = (r^2 + s^2 - 2rs\mu)^{\frac{1}{2}}$. The integrals of S_3 may be evaluated using numerical methods described in detail in references [2] and [20], or they may be evaluated more simply using methods we will describe in Section 4.

A physical interpretation for the parameters ζ_1 and η_1 is lacking at the present time. However, it is known that for the relevant applications (effective electrical or thermal conductivity, dielectric constant, elastic constants, etc.) the effective constants are themselves scale invariant and, therefore, any pertinent parameters of the microgeometry should also be scale invariant. Eqs. (15) and (16) clearly satisfy the scale invariance criterion, and it is hard to construct any other scale invariant scalars depending on S_3 or integrals of S_3 (at least this author has not been able to find any). The scale invariance argument also shows why the Hashin-Shtrikman bounds depend only on volume fraction information even though S_2 appears at intermediate stages of the derivation [9].

One significant practical observation about the results for ζ and η concerns the choice of image magnification. The digital image used to produce the empirical values of S_3 may or may not contain the level of detail required for accurate computation of these integrals. We have found [2] that our integration scheme produces good results for ζ when the effective radius R of the penetrable spheres satisfies $R/H \simeq 12$ where H is the pixel width. For η the best results were obtained when the effective radius of the penetrable spheres satisfied $R/H \simeq 25$. Thus, the image magnification required for the two integrals differs by about a factor of 2. This result is very reasonable when we compare the Legendre polynomials P_2 and P_4 appearing in the integrands. Since P_4 has twice as many zeroes as P_2, it is clear that, whatever resolution is required to obtain accurate values of ζ, about twice that resolution will be needed to obtain comparable accuracy for η. The observed behavior conforms to this expectation.

Although we expect this factor of 2 difference in optimum image magnification to be maintained for real materials, we do not expect that these particular values of (particle radius/pixel size) will be appropriate for all materials. The spherical particle shape and the lack of both cracks and surface roughness make the penetrable sphere model somewhat unrealistic. If the magnification of an image must be increased substantially beyond the values quoted in the preceding paragraph, then it is likely that the number of pixels in the image will also have to increase beyond the value of 512×512 assumed here. With the largest triangles having side $l \leq 64$, the total number of S_3 values computed from an image is 47377. Increasing the largest triangle size by a factor of 2 means increasing the total number of S_3 values to 364065 [2]; such an increase entails storing more computed values of S_3 than the total number of pixels in the original image. If such an increase in the data base is really needed for accurate determination of the integrals of interest, it might be preferable to use the digitized image itself as the data base for the larger triangles and compute the values of S_3 for these triangles as needed during the integration. Another approach would be to increase the number of pixels per image to 1024×1024 or 2048×2048 directly or by constructing a mosaic. The interpolation technique developed in [2] could be used with any of these approaches to the integration problem, and is used with still another approach in the next section.

4. Visualization of three-point correlations. Because the three-point correlation function $S_3(r, s, cos\,\theta)$ is a function of triangle shape and size and therefore of three arguments, it is somewhat difficult to display the function. If we fix the angle θ between the sides r, s of the triangle, then it is possible to display the resulting function of two variables. The display can take the form of a contour plot in the (r, s)-plane showing lines of constant $S_3(r, s, \mu)$; the display could be performed using three-dimensional plotting routines that actually show the surface of $S_3(r, s, \mu)$ above the (r, s)-plane; or the display can be presented in color as we will do here (see Figure 2). The color display has the advantage of a contour plot in that all of the function can be shown simultaneously (three-dimensional surface plots always suffer from the fact that part of the surface is hidden behind the peaks of the high values closest to the viewer). Furthermore, it has the advantage that quantitative information about the values of the function are coded into the color mapping.

Figure 2 shows nine views of the three-point correlation function computed from the cross section of the glass bead sample shown in Figure 1. The nine views were chosen for equally spaced angles from $\theta = 0°$ to $\theta = 180°$. The superimposed protractor shows clearly what the angle is for each frame displayed. The color bar at the bottom shows the mapping between color and the numerical value of the three-point correlation function. Since the porosity (volume fraction of void space) for the glass bead sample was around 30%, the spectrum of colors has been compressed in this example so the interesting features are most easily seen.

There were 512×512 pixels in Figure 1. The two-point correlation function was computed to a lag of 256 pixel widths. The full three-point correlation function was computed only for triangles with largest side $\le$ 64 pixel widths, and the display in each frame of Figure 2 shows the composite three-point correlation function for triangles with largest side $\le$ 128 pixel widths. The values for S_3 in the upper left corner of each frame were computed using the interpolation scheme of [2]. Note that the region where this interpolation scheme is used depends on the angle: For $\theta = 0°$, this region is a 64×64 pixel square. For $\theta = 90°$, this region is one-quarter of a circle with 64 pixel radius. For $\theta = 180°$, this region is an isosceles right triangle with adjacent sides 64 pixels in length. The remaining values of S_3 were obtained using the approximation (13). The accuracy of (13) for large arguments is demonstrated in Figure 2 by the high degree of continuity observed between the regions calculated using the different methods.

The main features of the three-point correlation function are easily deduced from this display. All nine frames show the characteristic behavior along the top and left hand sides that the three-point correlation function is independent of angle if either $r = 0$ or $s = 0$ — the reason being that if one side of the triangle vanishes then the triangle reduces to a line and the three-point correlation function is just the two-point correlation function for the length of that line. The single frame that is most distinct from all the others is for $\theta = 0°$. When the angle between r and s vanishes and $r = s$, then again the value of the three-point correlation function is given by the two-point correlation function $S_2(r)$. This behavior dominates the frame in the lower right of Figure 2. As the angle increases monotonically from zero, we see that the correlation function becomes nearly independent of angle for $\theta > 90°$, but the significantly higher values of the correlation function at small angles when $r = s$ is still very apparent for $\theta = 22.5°$.

Once we have the capability of displaying the three-point correlation function for different choices of angle as in Figure 2, it is easy to make the next step of computing the parameters of the microgeometry ζ and η. To do so, we must first do the angular integration by multiplying the values of S_3 by the appropriate values of the Legendre polynomials for each angle and then combining them in quadrature. This step produces the integrals $\int d\mu S_3(r,s,\mu)P_2(\mu)$ and $\int d\mu S_3(r,s,\mu)P_4(\mu)$. The remaining integrals over the square domain in the (r,s)-plane are straightforward. The accuracy of the resulting values of ζ and η will depend strongly on the choice of image magnification used. In particular, the examples chosen for display in Figure 2 were chosen to illustrate the characteristics of the correlation function, but higher resolution is required to obtain accurate values of the parameters of microgeometry if this integration scheme is to be used. Other integration schemes have been used to produce more accurate results [2, 19, 20].

5. Discussion. We have shown that it is possible to obtain useful measured values of three-point correlation functions for real materials, to interpolate and integrate those values to find the parameters of the composite microgeometry, and to use new visualization techniques to gain insight into the structure of the correlation functions themselves. If our information is incomplete — so that accurate values of both ζ and η cannot be obtained from image analysis techniques, other methods of attack may be required. It has been shown elsewhere [21] that much can be learned about effective properties when other information is available. For example, the existence of the bounds on elastic constants using the microgeometry parameters is enough to show that some improvements can be made to the Hashin-Shtrikman bounds.

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COLOR FIGURE CAPTION

Figure 2. Display of the three-point correlation function $S_3(r, s, cos\,\theta)$ as the angle θ between the sides r, s of the triangular argument varies from 0° to 180° in steps of 22.5°. In each frame, the origin for the (r, s)-plane is in the upper left corner.