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An optimum approximation of n-point correlation functions of random heterogeneous material systems

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An approximate solution for n-point correlation functions is developed in this study. In the approximate solution, weight functions are used to connect subsets of (n-1)-point correlation functions to estimate the full set of n-point correlation functions. In previous related studies, simple weight functions were introduced for the approximation of three and four-point correlation functions. In this work, the general framework of the weight functions is extended and derived to achieve optimum accuracy for approximate n-point correlation functions. Such approximation can be utilized to construct global n-point correlation functions for a system when there exist limited information about these functions in a subset of space. To verify its accuracy, the new formulation is used to approximate numerically three-point correlation functions from the set of two-point functions directly evaluated from a virtually generated isotropic heterogeneous microstructure representing a particulate composite system. Similarly, three-point functions are approximated for an anisotropic glass fiber/epoxy composite system and compared to their corresponding reference values calculated from an experimental dataset acquired by computational tomography. Results from both virtual and experimental studies confirm the accuracy of the new approximation. The new formulation can be utilized to attain a more accurate approximation to global n-point correlation functions for heterogeneous material systems with a hierarchy of length scales. © 2014 AIP Publishing LLC. [http://dx.doi.org/10.1063/1.4865966]
illustrate this point, the optimum approximation is implemented in the current study to approximate three-point correlation functions from two-point function in two different random heterogeneous systems; the first one comprising a hard-core platelets/polymer matrix system generated virtually, and the second one composed of a glass fiber/epoxy composite system processed and assessed experimentally.

II. APPROXIMATION OF N-POINT CORRELATION FUNCTIONS

For non-Grünberg microstructures, using n-point correlations (n > 2) of the structure can reveal more morphological details of the features and their distribution. Theoretically, non-Grünberg structures can be uniquely defined by exploiting an infinite-order correlation functions (n \(\to\) \(\infty\)). In statistical continuum theory, approximating higher-order statistical correlation functions provides a good precision and efficiency for the homogenization and reconstruction of heterogeneous microstructures.

In our previous research work, \(^{13}\) n-point correlation functions have been approximated with the aid of m-point correlation functions, where \(0 < m < n\). In that approximation, n-point correlation functions were divided into n subsets of (n-1)-point correlation functions. The key idea of this approximation was that the correlation functions can be approximated by a weighted sum of the products of n subsets of (n-1)-point correlation functions, which are linked using the conditional probabilities. In addition, the weight functions were calculated using the boundary limiting conditions. The general formulation of the approximation of n-point correlation functions (n \(>\) 3) was derived as \(^{13}\)

\[
C_n(x_1, x_2, x_3, \ldots, x_n) = \sum_{i=1}^{n} \left( W_i^n \prod_{l=1}^{(n-2)} C_{(n-1)}(x_1, \ldots, x_{(n-1)}) \right) \left( \prod_{l=1}^{(n-1)} C_{(n-2)}(x_1, \ldots, x_{(n-2)}) \right) \ldots \right),
\]

where \(W_i^n\) are the dependency weight functions. In the formulation above, \(x_{m}, x_{m} \ldots x_{y}\) is defined as the subset of \((n-1)\) points that include \(x_i\) as a member of the subset. The weight functions \(W_i^n\) can be calculated using the boundary limits. The first boundary condition is given for each i as

\[
\lim_{x_i \to \infty} C_n(x_1, x_2, x_3, \ldots, x_{(i-1)}, x_{(i)}, x_{(i+1)} \ldots, x_n) = C_1(x_i)C_{n-1}(x_1, x_2, x_3, \ldots, x_{(i-1)}, x_{(i+1)} \ldots, x_n).
\]

Here, \(C_1\) represents the one-point correlation function. This boundary limit condition can be written as

\[
\lim_{x_i \to \infty} \sum_{i=1}^{n} \left( W_i^n \left( \prod_{l=1}^{(n-2)} C_{(n-1)}(x_1, \ldots, x_{(n-1)}) \right) \left( \prod_{l=1}^{(n-1)} C_{(n-2)}(x_1, \ldots, x_{(n-2)}) \right) \ldots \right) = \ldots \right).
\]

Applying this boundary limit, we get

\[
\lim_{x_i \to \infty} W_i^n = 0, \quad W_j^n \neq 0 \quad \text{for} \quad j \neq i.
\]

The second boundary condition is given by

\[
\lim_{x_i \to \infty} C_n(x_1, x_2, x_3, \ldots, x_n) = \prod_{i=1}^{n} C_1(x_i).
\]

This equality condition leads to

\[
x_i \to \infty \quad (i = 1, \ldots, n), \quad \sum_{i=1}^{n} W_i^n = 1.
\]

The third boundary limit is expressed as

\[
\lim_{x_i \to x_j} C_n(x_1, x_2, x_3, \ldots, x_{(i)}, \ldots, x_{(j)}, \ldots, x_n) = C_{n-1}(x_1, x_2, x_3, \ldots, x_{(j)}, \ldots, x_{(n-1)}).
\]

From this boundary condition for compatible events and using Eq. (3), we get

\[
x_i \to x_j \quad (\text{for} \quad i \neq j), \quad W_k^n = 0, \quad k \neq i \quad \text{and} \quad k \neq j.
\]

Therefore, the necessary conditions for the weight functions are summarized as follows:

\[
x_i \to \infty \quad W_i^n = 0, \quad W_j^n \neq 0 \quad \text{for} \quad j \neq i,
\]

\[
x_i \to \infty \quad (i = 1, \ldots, n), \quad \sum_{i=1}^{n} W_i^n = 1,
\]

\[
x_i \to x_j \quad (\text{for} \quad i \neq j), \quad W_k^n = 0, \quad k \neq i \quad \text{and} \quad k \neq j.
\]

In the proposed approximation, a unique solution does not exist for the weight functions. Therefore, any chosen set of the weight functions that satisfy the necessary boundary limit conditions is useful for this approximation. For example, for
the approximation of three-point correlation functions, a simple choice for the weight functions has been proposed in our previous work\textsuperscript{13} as

\[
W_m^3 = \frac{|x_kx_l|}{|x_1x_2| + |x_1x_3| + |x_2x_3|} \quad m \neq l, \quad m \neq k,
\]

where \(x_i\) are the rotation centers and \(x_jx_l\) are the correlation vectors (Figure 1). In Eq. (15), \(\{\ldots\}\) represents the subset of vector lengths of correlations which do not include \(x_j\), the exponents \(\alpha_m\) and \(\alpha_k\) are optimization parameters, and \(\Gamma_{n-1}\) is expressed using Cayley-Menger determinant\textsuperscript{14} as

\[
\Gamma_{n-1}(\{|x_1x_2|, |x_1x_3|, \ldots, |x_{(n-2)}x_{(n-1)}|\}) = \begin{vmatrix}
0 & 1 & \ldots & 1 \\
1 & 0 & \ldots & 0 \\
\vdots & \vdots & \ddots & \vdots \\
1 & 0 & \ldots & 0 \\
\end{vmatrix}
\]

We note that \(\Gamma_k\) gives a formula for \((k-1)\)-dimensional volume of convex hull of the points \((x_i)\) in terms of the Euclidean distances which are defined using the magnitude of vector lengths for \(n\)-point correlation functions with \(n < 6\) (see Figure 1). For example, the second polynomial (or \(\Gamma_3\)) yields the well-known Heron’s formula for the area \(A\) of a triangle with known edge lengths. These proposed weight functions, using Cayley-Menger determinant, are no longer physically meaningful for \(k > 4\) (or \(n > 5\)) because the Cayley-Menger determinant in the Euclidean 3D space becomes equal to zero.\textsuperscript{15} We also note that the proposed weight function in the work of Baniassadi et al.\textsuperscript{13}, for \(n < 5\), can also be used to derive the above Cayley-Menger determinant-based approximation.

Given the limitation of the above proposed approximation for the weight functions, we propose a new generalized approximation based on simple weight functions that satisfy all necessary conditions in Eq. (11)–(13) and which are valid for \(n\)-point correlation functions with \(n > 2\). These are given by

\[
W_m^n = \frac{\left(\frac{(-1)^{n-1}}{2^{n-2}((n-2)!)^2} \Gamma_{n-1}(\{|x_1x_2|, |x_1x_3|, \ldots, |x_{(n-2)}x_{(n-1)}|\})_m|m \neq l\right)^{\alpha_m}}{\sum_{k=1}^n \left(\frac{(-1)^{n-1}}{2^{n-2}((n-2)!)^2} \Gamma_{n-1}(\{|x_1x_2|, |x_1x_3|, \ldots, |x_{(n-2)}x_{(n-1)}|\})_k|n \neq k\right)^{\alpha_k}} |n < 6, (17)
\]

These weight functions are derived by simply considering multiplier correlation lengths of the subsets. Unlike the previous approximation (Eq. (15)), the new generalized approximation is a mathematical description with no particular physical meaning, particularly for \(n > 3\). In fact, we note that, for \(n = 3\), the previous (Eq. (15)) and new (Eq. (17)) approximations yield the same result, and that Eq. (17) reduces to

\[
W_m^3 = \frac{\left(\frac{|x_kx_l|^2}{|x_1x_2|^2 + |x_1x_3|^2 + |x_2x_3|^2}\right)^{\alpha_k}}{\sum_{m=1, m \neq l, m \neq k}^n \left(\frac{|x_kx_l|^2}{|x_1x_2|^2 + |x_1x_3|^2 + |x_2x_3|^2}\right)^{\alpha_k}} |m \neq l, m \neq k, (18)
\]

where \(\alpha\), \(\beta\), and \(\gamma\) are non-zero positive real numbers and \(l, m, k\) are equal to 1, 2, and 3.

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**FIG. 1.** Schematic of correlation vectors of the \(n\)-point correlation functions.

**FIG. 2.** Two-point correlation functions (TPCF) via correlation length \(r/d\), where \(d\) is the diameter of the platelets, for the two-phase heterogeneous systems (shown in inset image).
III. COMPUTATIONAL VERIFICATION

To show the validity of the proposed weight functions approximation, we use a virtual system based on random microstructures and consider the 3-point correlation functions only. The virtual microstructure consists of a representative volume element (RVE) filled with 4.5% inclusions. Isotropic realizations of randomly distributed hard-core platelet inclusions, with three different aspect ratios, are generated and used to calculate the optimum three-point correlation functions of the generated virtual microstructures. In this study, as a first step, three-point correlation functions are approximated using Eq. (18). In the next step, optimum values of the optimization parameters ($\alpha$, $\beta$, and $\gamma$) are calculated using neural networks. To calculate the two-point correlation functions, we use Monte-Carlo simulations\(^2\) for the virtual non-eigen samples. The obtained results for the two-point correlation functions are then used to get the approximate solution for the three-point correlation functions. Then, we optimize the approximation using Monte Carlo results for the 3-point correlation points. For this purpose, we use a three layer neural network with one input and one output. The network has two hidden layers with 32 and 10 neurons and one output layer with one neuron and uses Levenberg-Marquardt back propagation algorithm for training.

The platelet geometries of inclusions are defined by the corresponding radius center and the normal vector for each inclusion surface. The size of the RVE is chosen large enough to produce convergence for the two-point correlation functions. In Figure 2, we show the calculated two-point correlation functions (TPCF) of the three realized microstructures with different inclusion aspect ratios.

Three-point correlation functions (platelet/platelet/platelet) are calculated using Monte Carlo simulation, and the results are used to optimize the weight functions in Eq. (18).

In the simulation, more than 1000 three-point correlation points with different magnitude of vector lengths have been computed and used to optimize the three-point correlation functions (THPCF) given by Eq. (3). In this optimization process, we define an error function as

$$\text{Error} = \left| \frac{\text{THPCF}(r)_{\text{simulation}} - \text{THPCF}(r)_{\text{approximation}}}{\text{THPCF}(r)_{\text{simulation}}} \right|. \quad (19)$$

The average error contours are reported in Figure 3 via the optimization parameters ($\alpha$, $\beta$, and $\gamma$) of the approximation, showing a large dispersion of the error. However, we can find the optimum values of these parameters to have the best approximation of the THPCF for the considered microstructure (see Table I).

IV. EXPERIMENTAL VALIDATION

To scrutinize the accuracy of the proposed approximation, a composite specimen composed of 52 vol. % unidirectional glass fibers loaded into an epoxy matrix is analyzed. The internal microstructure of the specimen has been obtained using a high-resolution 3D X-ray imaging system.

<table>
<thead>
<tr>
<th>Aspect ratio</th>
<th>$\alpha$</th>
<th>$\beta$</th>
<th>$\gamma$</th>
<th>Average of minimum error</th>
</tr>
</thead>
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<tr>
<td>10</td>
<td>2.2</td>
<td>2.2</td>
<td>2.2</td>
<td>0.12</td>
</tr>
<tr>
<td>15</td>
<td>2.1</td>
<td>2.1</td>
<td>2.1</td>
<td>0.13</td>
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<tr>
<td>20</td>
<td>2</td>
<td>2</td>
<td>2</td>
<td>0.16</td>
</tr>
</tbody>
</table>

FIG. 4. (a) Sample x-ray projection image used for reconstruction; (b) 2D cross-section view of binary label matrix; and (c) 3D volume rendering of the arrangement of the glass fibers in the unidirectional composite specimen.
FIG. 5. In-plane variation of TPCF\textsubscript{01} (a) and TPCF\textsubscript{11} (b), with 1 and 0 respectively denoting the glass and epoxy phases; (c) Variation of the four two-point correlation functions for a random in-plane direction, showing a correlation length $\lambda = 10 \mu$m for all in-plane directions (All dimension are in $\mu$m).

(MicroXCT-400, Xradia). Figure 4(a) shows a sample 2D projection generated by the X-rays passing through the specimen. A number of these X-ray projections have been acquired from the specimen in different angles (from $-170^\circ$ to $170^\circ$ around the main axis of the specimen). Using a filtered back projection method, the 3D microstructure of the specimen has been reconstructed from these projection images. To eliminate noise and improve quality, a Gaussian smoothing filter has been applied to the raw data. The binary representation of the microstructure has been segmented from gray-scale data using a threshold filter. A 2D cross-section of the binary matrix is shown in Figure 4(b). Each voxel of the binary matrix (also known as label matrix) represents a cubic chunk of the material and a non-zero value is assigned to each voxel corresponding to the phase occupying the location of the voxel. These operations have been performed in MATLAB using the Image-Processing toolbox. Figure 4(c) also shows a volumetric rendering generated from the acquired data revealing the anisotropic arrangement of unidirectional glass fibers in the composite specimen.

Two-point correlation functions (TPCFs) have been evaluated directly from the binary label matrix. Global 3D TPCF have been constructed by systematically evaluating these two-point functions in three orthonormal directions over the correlation range. Figures 5(a) and 5(b) show the two-point correlation functions TPCF\textsubscript{11} and TPCF\textsubscript{01} with 1 representing glass phase and 0 the epoxy evaluated along a random in-plane direction. Figure 5(c) shows a 2D slice of the global two-point functions.

Next the three-point correlation functions (THPCF) are approximated from the two-point functions utilizing Eqs. (3) and (18). For comparison, the reference value of the THPCFs are also evaluated directly from binary label matrix and compared against their approximate counterparts. An error cost function is constructed simply by taking absolute difference between the approximate and reference values and it is minimized to find best values for $\alpha$, $\beta$, and $\gamma$. Unique weighting considered for all three optimization parameters in the cost function. Using a collocation approach, a number of points have been selected randomly in the correlation range for the specimen; i.e., about $10 \mu$m as shown in Figure 5(c), and the average value for each parameter are reported in Table II. The distribution of error corresponding to the proposed approximation is depicted in Figure 6 showing a maximum of 15% error.

V. CONCLUSIONS

In the present work, a previously developed approximation for n-point correlation functions based on the conditional probability theory has been modified. In this study, a set of weight functions has been proposed to obtain an accurate approximation for n-point correlation functions of heterogeneous materials or systems. The approximation can be adapted to different microstructures. Two examples have been shown to validate our approach. In both examples, three-point correlation functions are approximated from the set of two-point functions using our proposed methodology for optimizing the accuracy. This methodology can be readily extended to higher-order correlation functions that are needed for applications such as cosmology, biology, and materials science.