

Representation and Computational Structure-Property Relations of Random Media

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Recent trends towards integrated computational materials engineering (ICME) demand increasing reliance on modeling and simulation to estimate microstructure-property relations of materials with random microstructures.

INTRODUCTION

Digital representation of random microstructure and predictive computational structure-property relations over a range of realistic microstructures are key enabling technology elements to support integrated computational materials engineering (ICME).¹ The essence of ICME is concurrent design of products and the materials which comprise them. This is achieved by linking material models at multiple length and time scales corresponding to the hierarchy of material structure to address problems relevant to specific products and applications. ICME builds on earlier initiatives such as DARPA Accelerated Insertion of Materials (AIM)² and is consistent with the report of the National Science Foundation Blue Ribbon Panel on Simulation Based Engineering Science³ regarding the need to more fully integrate modeling and simulation within the curriculum of engineering to tackle a wide range of interdisciplinary and multiscale/multiphysics problems. Moreover, computational microstructure-property relations are a key ingredient to any strategy to support design of materials to meet a specified range of performance requirements.⁴⁻⁶

The basic notion is to generate digital microstructures, or their equivalent, in sufficient detail to estimate the dominant microstructure-response relations of interest for a given material system and application. For example, the ability to represent realistic distri-

butions of distinct grains and second phase particles or intercalating phases in metallic alloys, particles or fiber reinforcement phases in metal matrix or polymer matrix composites, and so forth enables computational estimates of the statistical distributions of a wide range of responses relevant to system performance. We use the qualifier “estimates” here to emphasize that assumptions are made in any simulation that idealize or simplify reality, whether in terms of geometric representation of microstructure, phase properties, or interactions of defects within and between phases. We seek models that are framed at the lowest relevant scale with the degree of refinement necessary to predict collective response at higher length and time scales in a manner suitable for the particular goal or end application. Such estimates potentially serve multiple goals in the context of

ICME:

- Designing new materials
- Modification and enhancement of existing materials for improved performance
- Accelerating insertion of new materials
- Quantifying variability of material performance
- Estimating durability of material systems
- Facilitating feasibility studies on potential payoff of new material systems in products

The mix of goals depend on the application; details of the microstructure representation and simulation of material responses differ in degree of rigor and/or investment, for example, when considering “what if” games for adding value by introducing new materials versus strategies for improving existing materials or developing more accurate and robust life prediction schemes for already fielded material systems. In all cases, it is important to account for dominant mechanisms that affect the material response at multiple levels of hierarchy. Applications often require control of properties or responses in multifunctional/multiphysics realms of physical and chemical properties, structural, mechanical, electronic, thermophysical, electromagnetic, phase change, etc. In the interest of brevity and specificity, this article focuses on mechanical properties and responses of polycrystalline, polyphase metallic material systems.

SCALE TRANSITIONS AND RVE FOR RANDOM MICROSTRUCTURES

Recent advances in microstructure characterization (e.g., x-ray microtomography, automated serial sectioning,

How would you...

...describe the overall significance of this paper?

This paper considers the problem of computational estimation of material properties from knowledge of microstructure. Applications include materials design, accelerated insertion of materials, durability estimates, and quantifying variability of properties, among others.

...describe this work to a materials science and engineering professional with no experience in your technical specialty?

The problem of property estimates based on microstructure characterization is critical for design of next-generation materials. Moreover, certain properties depend on average microstructure, while others depend on extreme values. Both are considered in this work.

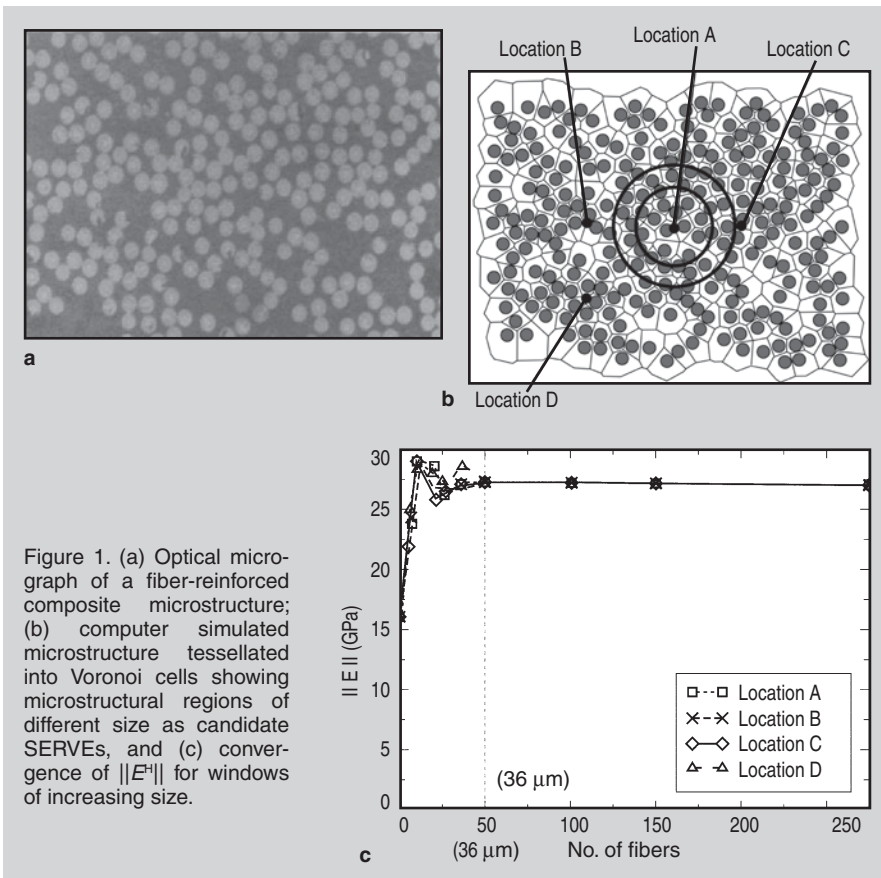


Figure 1. (a) Optical micrograph of a fiber-reinforced composite microstructure; (b) computer simulated microstructure tessellated into Voronoi cells showing microstructural regions of different size as candidate SERVEs, and (c) convergence of $\|E^h\|$ for windows of increasing size.

three-dimensional (3-D) atom probe) have demonstrated the viability of accurately capturing the 3-D details of the microstructure in selected regions of the sample at various length scales. There are three essential elements of modeling and simulation to support the goals discussed in the previous section. These include (1) geometric representation in digital or other discrete form of material structure attributes at multiple levels of spatial resolution (such as individual phases, mesoscopic spatial arrangements, and the scale of the application), (2) constitutive equations for individual phases and interfaces/interphases (kinematics, thermodynamics, and kinetics), and (3) a multiscale modeling framework. Item (1) includes all relevant spatial correlations of microstructure or mesostructure attributes.

An heuristic principle often followed is that the highest degree of resolution for a given problem is established by the scale of microstructure attributes over which control can be exerted through material processing or alteration to modify the desired responses. For example, for tailoring elastic anisotropy and yield strength of

polycrystalline alloys through texture control, the finest scale of resolution is typically that of individual grains. In contrast, the traditional approach of materials selection in design typically takes a much different approach that focuses on bulk material properties, perhaps normalized by density in lightweight designs.⁶ The identification of a minimum length scale of resolution for a given problem serves as the starting point for framing the constitutive relations in point (2) and multiscale modeling framework in point (3).

Element (3) on multiscale modeling requires some perspective.⁴⁻⁶ No single strategy is suitable or best for all cases. Concurrent schemes in which multiple length and time scales are bridged seamlessly and simultaneously are computationally intensive since they involve simulations with variable resolution over the same spatial domain. They are typically framed in terms of direct numerical simulation (DNS) of microstructure, often employing the same numerical scheme cast in multi-resolution format. Hierarchical schemes are less intensive and either consist of distinct simulations at different length scales of the microstructure

hierarchy or pass information upward in scale using handshaking or formal homogenization methods; hierarchical schemes can make use of radically different model frameworks and degrees of freedom at different scales, for example particle dynamics as in atomistic modeling of interfaces or defects at scales on the order of tens of nanometers, discrete dislocation models for line defect interactions at scales on the order of hundreds of nanometers, or continuum crystal plasticity models for responses of grains or phases at scales on the orders of several micrometers and above. In certain cases, a concurrent multiscale strategy may be desirable since it can track damage localization and growth across scales.⁷

This article focuses on scale transitions between two levels of hierarchy to arrive at effective properties or responses, based either on DNS or on influence function methods to account for interactions of microstructure heterogeneity. Regardless of the multiscale modeling strategy, the concept of a statistically “representative volume element” (RVE) is foundational to computational estimates of structure-property relations. The concept of a RVE was introduced by Hill⁸ as a microstructural subdomain that is representative of the entire microstructure in an average sense. It depends on microstructure and property/response of interest. In short, an RVE encompasses a large enough volume of microstructure such that the predicted responses or

Equations

$$M(r) = \frac{dH(r)}{dr} / \frac{dK(r)}{dr}, \quad (1)$$

$$\text{where } H(r) = \frac{1}{m^2} \frac{A}{N^2} \sum_i^N \sum_{k=1}^{k_i} m_i m_k(r),$$

$$\text{and } K(r) = \frac{A}{N^2} \sum_{k=1}^N l_k(r)$$

$$m_k = w_1 S_1^k + w_2 S_2^k + w_3 S_3^k, \quad (2)$$

$$\text{where } S_1^k = \frac{(LAF)^k}{\text{Max}(LAF)^j}, \quad S_2^k = \frac{(IND)^k}{\text{Max}(IND)^j},$$

$$S_3^k = \frac{(NN)^k}{\text{Max}(NN)^j}$$

$$f_r^{hh'} = \left\langle \frac{1}{S} \sum_{s=1}^S m_s^h m_{s+r}^{h'} \right\rangle \quad (3)$$

properties do not change with further increase of size; therefore, it represents all statistical moments of the microstructure-property relations. Simulating at the scale of the RVE is a rather stringent requirement. From a practical viewpoint, each response of interest may have a different RVE size. Properties that depend chiefly on higher order spatial statistics of microstructure attributes (e.g., extreme values of largest grains or particles, most severe interactions of particles or phases, etc.), such as ductility or high cycle fatigue, have potentially much larger RVE sizes than properties that depend mainly on lower order moments of microstructure spatial arrangement, such as elastic stiffness or thermal conductivity.⁹ Some authors use the term “statistical volume element” (SVE) to designate a random sample of microstructure that is too small to satisfy the statistical homogeneity requirements of the RVE for a given response function. In this case, a collection or set of SVEs must be simulated to build up the ensemble statistics required to capture suitably high order moments of a desired response distribution. The nature of applied boundary conditions also influence the issue of convergence towards RVE response.¹⁰

With these definitions in mind, we shall make use of both RVEs and SVEs. To emphasize that the size of the RVE depends on spatial statistics of microstructure, a Statistically Equivalent RVE (SERVE) is identified as the smallest volume element of the microstructure for which (i) the effective material properties of the SERVE should be equivalent to those of the entire microstructure, (ii) the spatial distribution functions of microstructure attributes should be equivalent to those for the overall microstructure, and (iii) corresponding responses are independent of location within the overall microstructure and applied stress state. Various statistical descriptors have been proposed to characterize and classify microstructures based on the spatial arrangement of heterogeneities.^{11,12} In early work, Willis¹³ proposed a two-point probability measure to arrive at statistically equivalent RVEs for composites with random variations in the microstructure. In practice, the size of the SERVE based on individual re-

sponses or properties may vary, since they are dominantly affected by different moments of the spatial distribution of microstructure attributes;¹¹ for example, elastic stiffness is insensitive to higher order moments involving phase shape or spacing, while plastic strain localization and damage evolution¹⁴ are sensitive to these higher order moments and hence require larger SERVEs.

ESTIMATING THE RVE

Identification of the SERVE is often based on effective moduli of a microstructural region subjected to non-homogeneous boundary conditions (e.g., periodic).^{15–17} As shown in Figure 1, windows of identical volume fractions but increasing size and number of inclusions may be sequentially analyzed for evaluating the components of the homogenized elastic stiffness tensor. The SERVE size is established through convergence of the Frobenius norm $\|E^H\|$ of these components at different locations of the micrograph.

Another method of evaluating the SERVE is based on use of marked correlation functions $M(r)$ introduced in Reference 18 for providing multivariate characterization of the microstructural phase distribution. A mark is an

assigned state variable field with a strong connection to the specific mechanical or failure properties and responses in question. The marked correlation functions correlate these fields in the microstructure to delineate the region of influence in a heterogeneous neighborhood. It is expressed in terms of the derivatives of relevant state variables and geometric distribution functions as Equation 1. (All equations are shown in the table.)

In Equation 1, m_i represents a *mark* associated with the i^{th} inclusion and m is the mean of all marks. For observations within a finite window of area A , the variable r is a measure of the radial distance of influence and N is the total number of inclusions. Further, $H(r)$ is the mark intensity function and $K(r)$ is a second-order intensity function, defined in References 15–18. A declining value of $M(r)$ indicates reduced correlation between elements of the microstructure, and therefore serves as a useful metric for the estimation of the SERVE for a random microstructure with respect to a property or response of interest. Figure 2a shows results with a mark taken as the average traction at points on the fiber-matrix interface that experience the highest tensile normal

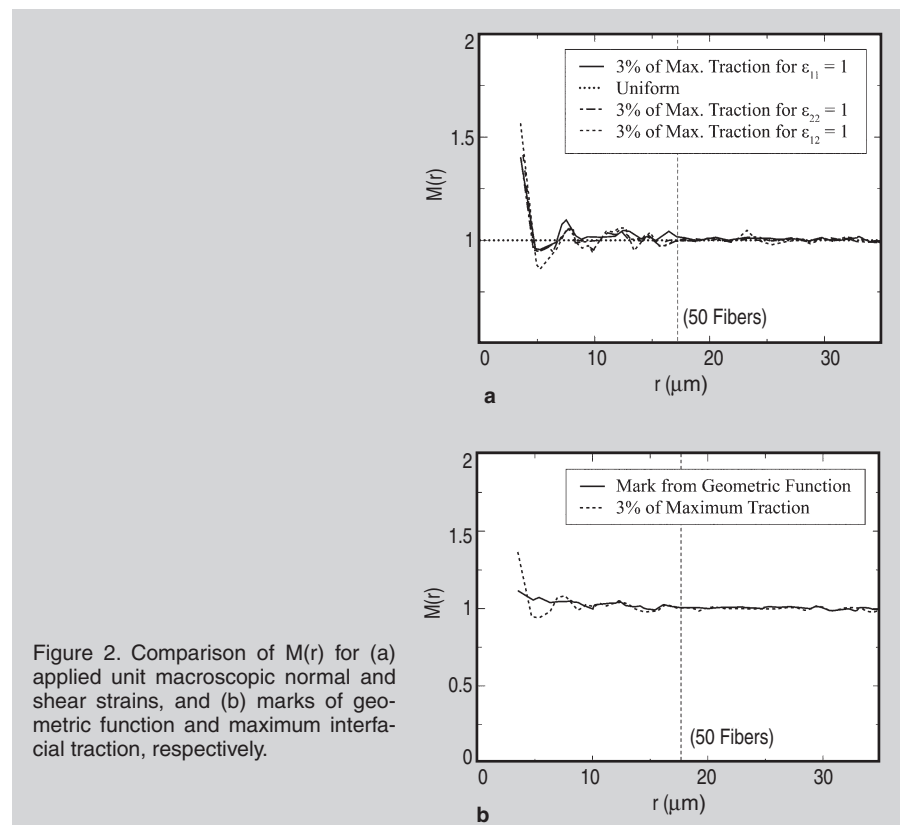


Figure 2. Comparison of $M(r)$ for (a) applied unit macroscopic normal and shear strains, and (b) marks of geometric function and maximum interfacial traction, respectively.

(i.e., 36 μm), containing about 50 fibers can be considered as the SERVE.^{15,17}

As an alternative estimator, distribution functions of critical variables in a sub-domain have been compared with those for the entire microstructure in References 16 and 20 to estimate the SERVE size in random microstructures. An example of the critical variable with regard to strength and ductility is the traction acting on inclusion-matrix interfaces, which is a precursor to interfacial debonding. Estimation of the SERVE entails extracting volume elements (SVEs) of increasing sizes from the micrograph with the phase volume fractions matching the entire microstructure, followed by micromechanical analysis of the boundary value problems subject to periodic boundary conditions and applied strain states. A critical interfacial fraction (*CIF*) is defined as the ratio of l_{tc} (interface length for which the traction exceeds a critical value T_c) to the total length of all interfaces l_{me} . The plotted distribution of *CIF* as a function of T_c for SVEs of increasing size are compared with that for the entire microstructure. The smallest element for which the two distributions match provides an estimate of the SERVE for this response function. The use of the indicators such as coefficient of variation of area fraction, two-point correlations, as well as marks based only on geometry point to the conclusion that, for problems without significant microstructure damage evolution or phase rearrangement due to inelastic deformation, the SERVE may be estimated only using spatial statistics of microstructure without having to solve micromechanics problems via DNS or other schemes.

Estimating the SERVE for microstructures with evolving damage is a more involved and complex exercise since the evolution of stresses and strains are affected by the distribution of evolving damage in the microstructure as well.^{15,17} Even when the microstructure is geometrically uniform, initiation and progression of damage can result in a SERVE that is considerably larger than that for a stationary, non-evolving microstructure. Approaches similar to those used for the undamaged material are used to evaluate

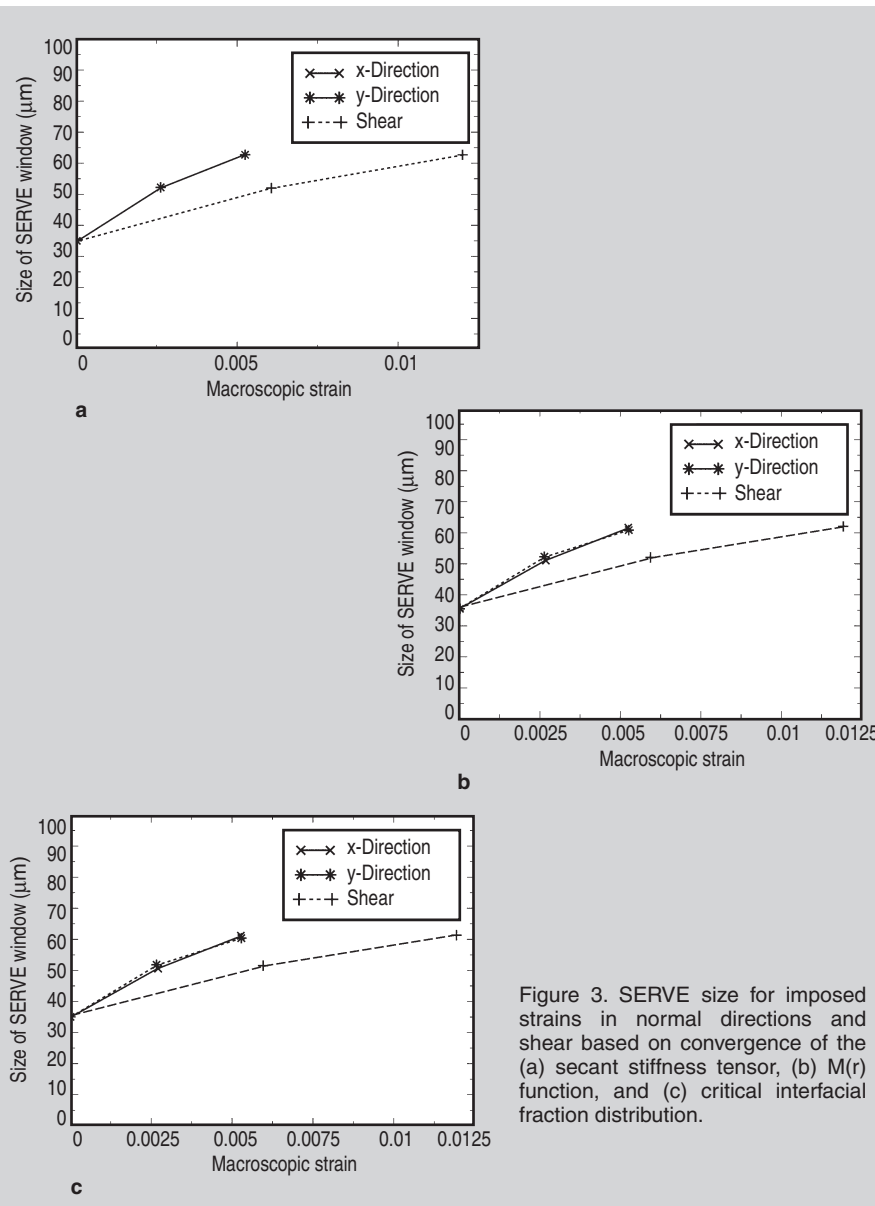


Figure 3. SERVE size for imposed strains in normal directions and shear based on convergence of the (a) secant stiffness tensor, (b) $M(r)$ function, and (c) critical interfacial fraction distribution.

components. It compares the distribution of $M(r)$ for three different applied uniaxial macroscopic strains. The line $M(r)=1$ corresponds to a uniform distribution of fibers with identical mark. With increasing r , the local anisotropy in the mark diminishes and all three plots converge to depict the same trend for $M(r)$. The radius of convergence for all the cases is approximately 18 μm , with a SERVE containing about 50 fibers. The SERVE estimating marks may also be based purely on geometric or morphological attributes of the microstructure, especially for linear elastic problems. A weighted mark in terms of geometric parameters that contribute to microstructural damage has been constructed in References 16 and 17. The mark associated with the k^{th} inclusion is defined in Equation 2,

where S_i^k are geometric parameters characterizing the local distribution and w_i are the assigned weights. Specifically, S_1^k is a measure of the normalized local area fraction $(LAF)^k$, S_2^k is a normalized measure of the inverse of near-neighbor distance $(IND)^k$, and S_3^k is a normalized measure of the number of near-neighbors $(NN)^k$ (see Equation 2).

Clearly, a myriad of possibilities exist for such assignment of marks. Figure 2b shows a comparison of $M(r)$ from the geometric function in Equation 2 with that from maximum traction at the interface. While at lower r values, $M(r)$ for the geometric function is lower, the convergence pattern is similar for both cases. Both approach unity at around the $r_p \sim 18 \mu\text{m}$. It is estimated that a window size of twice the radius

SERVE for heterogeneous microstructures undergoing damage, specifically interfacial debonding. The assigned mark is the inverse of the magnitude of the tensile traction at the debonding fiber-matrix interface, over the length of the non-debonded or intact portion of interface. Since the tractions are expected to increase with decreasing distances between inclusions, the inverse of the traction achieves a positive correlation between marks and distances. At any given stage of damage evolution, the largest micro-region obtained by various approaches may be regarded as the instantaneous size scale of the SERVE.

The optical micrograph in Figure 1 is again examined with evolving interfacial damage due to normal and shear applied unit strains.^{16,17} Interfacial damage is modeled using cohesive zone models. Results of convergence of the secant stiffness tensor for the three loading conditions are shown in

Figure 3a. The SERVE size changes with increasing damage. The SERVE size increases at a much slower rate for the case of imposed shear strain than for imposed normal strain due to the extent of damage, which is a function of the cohesive law parameters.

For the marked correlation function, the radius of convergence r_p monotonically increases with damage evolution in the microstructure. The SERVE size at the terminal strain is $\sim 62 \mu\text{m}$, which is almost 1.7 times higher than for the undamaged interface. The rate of increase in SERVE size in Figure 3b is again found to be similar for the applied normal strains, but is slower for the shear strain. Finally, for the CIF distributions, Figure 3c depicts the change in the SERVE size with the evolution of damage from the distribution functions of critical variables. Ghosh et al.²¹ have similarly evaluated the SERVE for metals and alloys subject to evolving plastic deformation that contain

a dispersion of heterogeneities (e.g., particulates, intermetallics, or voids in their microstructure); these studies indicate that a mark associated with the micromechanical plastic work W_p at each location within the microstructure can provide the means to identify the SERVE for a given random microstructure.

N-POINT STATISTICS AND WEIGHTED RVE SETS

A very general approach to mathematical representation of random microstructures is afforded by n -point correlations or n -point statistics,^{11,22} a rigorous framework defining the spatial relations of “local states” in the microstructure. The local state is denoted by h , and is regarded as an element of the local state space H that identifies the complete set of distinct local states (microstructure attributes) that can be sampled at a selected length scale.

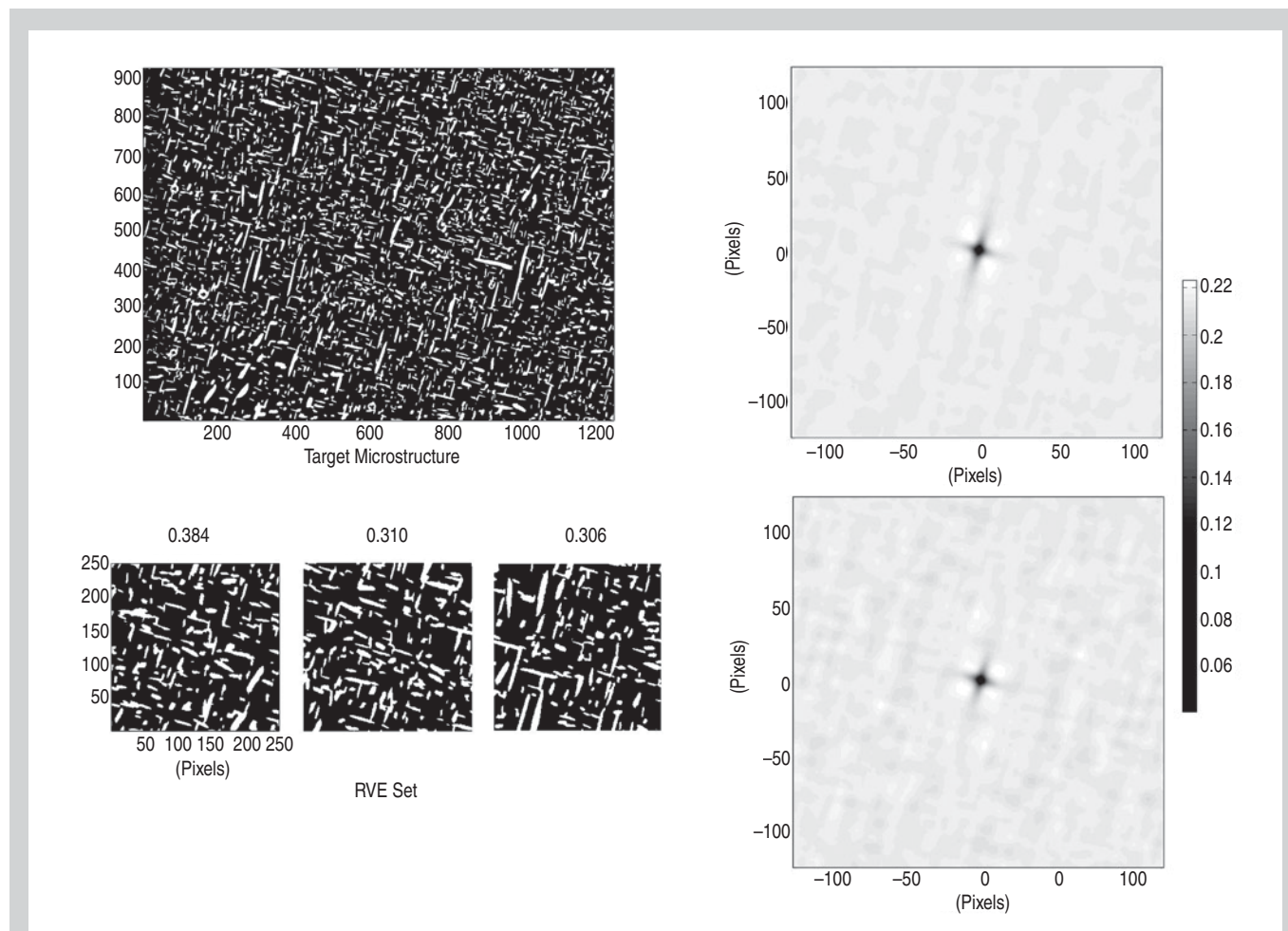


Figure 4. Illustration of the construction of a 3-member RVE Set comprised of weighted SVEs for an experimentally characterized precipitate microstructure.²⁴

For example, at the scale of individual grains (i.e., crystals), the local state may be characterized by the thermodynamic phase, the elemental composition of the phase, and the crystal lattice orientation. It is therefore important to recognize that the definition of the local state and the corresponding local state space is inherently tied to the length and time scales salient to the physics of the phenomena being modeled.

The n -point correlations provide a hierarchy of statistical measures of the microstructure. The simplest of these are the 1-point correlations, f^h , which essentially reflect the probability of finding local state h at a randomly sampled point in the microstructure. The phase volume fractions, which do not capture any spatial information regarding the microstructure, are prime examples of 1-point statistics. Expanding on this basic concept, the 2-point correlations, denoted $f_r^{h_1 h_2}$, capture the probability density associated with finding local states h_1 and h_2 at the tail and head, respectively, of a prescribed vector r randomly placed into the microstructure. With the advent of CCD cameras, SVEs are typically captured as digital images composed of a two-dimensional (2-D) or three-dimensional (3-D) array of discrete pixels or voxels. Consider one such SVE where each pixel is indexed, $s = \{1, 2, \dots, S\}$. For mathematical manipulation, it is convenient to represent the characterized SVE as a digital array m_s^h , which denotes the volume fraction of local state h at pixel s . The 2-point correlations for a given sample are commonly estimated as Equation 3 where the brackets $\langle \rangle$ indicate ensemble averaging over a number of SVEs. In the notation used here, both s and r are treated as vectors. The 3-point and higher-order correlations are defined in an analogous manner. Equation 3 is readily identified as a discrete convolution and is best computed by exploiting the well known convolution properties of the discrete Fourier transforms (DFTs). There is a tremendous leap in the amount of microstructure information contained in the 2-point statistics compared to the 1-point statistics. Many of the commonly used microstructure metrics, such as the average grain or precipitate size and shape, and the grain boundary

character distribution, can be recovered from appropriately defined 2-point statistics and their related measures.^{11,22,23}

It is often desirable to identify a statistically representative microstructure that corresponds to the ensemble averaged statistics computed using Equation 1 (ideally extracted from multiple SVEs or at least from a very large microstructure dataset). An instantiation of a microstructure that is large enough to correspond to complete ensemble-averaged statistics constitutes the RVE, as discussed earlier. This definition of a RVE can differ substantially from the approach taken in Figure 1c based on convergence of homogenized elastic stiffness via DNS. It is often impossible to reconstruct a single microstructure in a small window that accurately reflects the ensemble-averaged 2-point statistics computed on multiple SVEs (or equivalently on a very large SVE or RVE). Computationally, it is significantly much simpler to identify a set of weighted SVEs with the weighting optimized to match the ensemble averaged statistics to desired accuracy. These sets of optimized weighted SVEs are referred to as RVE Sets.²⁴ Figure 4 shows an example of the construction of a three-member RVE Set for an experimentally characterized two-phase precipitate microstructure. The elements of the RVE Set were constructed such that the weighted average of the 2-point statistics of the three elements (weights shown on top of the RVEs) is in excellent agreement with the 2-point statistics obtained from a very large scan (top left), as seen in the plots on the right. This construction was further validated by the observed match in a number of elastic and plastic properties/responses for the RVE Set against the corresponding properties/responses for the large microstructure dataset.²⁴

These n -point statistics can be inserted naturally into the higher-order homogenization schemes used to predict macroscale effective properties of a heterogeneous material system. Such relations are often achieved through perturbation expansions.^{22,25,26} The Green's function (or more generally, influence function) solution used in these methods provides a natural link between terms of the series expansion and correlation functions that contain

different orders of geometrical information. As a further extension of this approach, one may focus on the local material response as determined by the Green's function rather than the homogenized response. Such an approach may be used as the basis of microstructure design to optimize performance related to failure criteria that are concerned more with local than with global response. A localization tensor arises naturally in the Green's function approach, and benefits from the same approach as taken with the homogenization relations mentioned above.²⁷ Furthermore, calibration techniques may be combined with the spectral methods to result in very accurate and fast computation of the localization tensors.^{28,29} Moreover, n -point statistics can be matched using simulated annealing or other methods to reconstruct SVEs for DNS to obtain a wide range of properties/responses for stationary and evolving microstructures, the latter involving processes such as plasticity and damage. In all cases, it is hypothesized that responses computed using the RVE set will correspond to that of the RVE for a given microstructure and response(s) of interest. Significant computational advantage is afforded by using such weighted SVEs since computational time increases in a highly nonlinear manner with size of the SVE (see also References 30, and 31).

It is also noted that for responses such as minimum high cycle fatigue lifetime or minimum true fracture ductility that depend on higher order moments of spatial statistics (e.g., minimum spacing, maximum size of two adjacent particles or grains, etc.), the weighted RVE set can be extracted from marked correlation functions using "hot spots" of plastic strain intensification obtained from DNS of SVEs as the marks.³² In this way, attention is focused on the tail region of the probability distribution of the effective response rather than the lower moments of the distribution. In general, the RVE is untenably large for such extreme value responses to consider DNS at the RVE scale as an option.

CONCLUSION

The use of microstructure attribute/geometry-based indicators such as

coefficient of variation of area fraction, two point correlations, and marks based on geometry indicates that for certain problems without damage, the SERVE can be estimated without having to solve the micromechanics problem. The continual increase in the SERVE size in the microstructure with evolving damage provides grounds for its restricted use in homogenization schemes that utilize RVEs for evaluating constitutive models at the continuum level; the concept of RVE sets may provide a viable option for such cases. The general utility of n-point statistics is considered as a basis for efficient and fairly general methods to characterize microstructure-property relations. Although not addressed here for the sake of brevity, experiments play a pivotal role by providing insight into key mechanisms to be addressed in constitutive modeling at the microstructural level, as well as properties of individual phases. Moreover, the responses of interest must be experimentally validated for selected microstructures.

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