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Statistical methods for mechanical characterization of randomly reinforced media

Mikhail Tashkinov

Abstract

Advanced materials with heterogeneous microstructure attract extensive interest of researchers and engineers due to combination of unique properties and ability to create materials that are most suitable for each specific application. One of the challenging tasks is development of models of mechanical behavior for such materials since precision of the obtained numerical results highly depends on level of consideration of features of their heterogeneous microstructure. In most cases, numerical modeling of composite structures is based on multiscale approaches that require special techniques for establishing connection between parameters at different scales. This work offers a review of instruments of the statistics and the probability theory that are used for mechanical characterization of heterogeneous media with random positions of reinforcements. Such statistical descriptors are involved in assessment of correlations between the microstructural components and are parts of mechanical theories which require formalization of the information about microstructural morphology. Particularly, the paper addresses application of the instruments of statistics for geometry description and media reconstruction as well as their utilization in homogenization methods and local stochastic stress and strain field analysis.

Keywords: Heterogeneous media, Random microstructure, Stochastic fields, Correlation functions, Effective properties, Local stress fields, Multipoint statistics, Media reconstruction, 3D models

Introduction

One of the main problems of materials science is establishing connection between the physical and mechanical properties of materials, their microstructural features and parameters influenced by manufacturing process. For many centuries, the main option for investigation of this link remained direct experiments. Gathering the vast amount of empirical information was the only way to progress in creation of advanced materials. New models and numerical simulation techniques which had started to emerge in twentieth century significantly increased possibilities of production of materials with tailored parameters of microstructure to suit needs of the specific engineering applications. That allowed to substitute resource consuming experimental identification with mathematical modeling and gave a boost to creation of new classes of materials. Among the most highly potential are heterogeneous materials that consist of several phases with distinguished properties combined together. Examples of materials belonging to this class are particle and fibre reinforced composites, polycrystalline metal and ceramic alloys, poly-phase amorphous materials and some classes of polymers (Fullwood et al. 2010). In order to consider specifics of their microstructure, which frequently contains randomly placed elements, the special simulation techniques were developed.

The problem of modeling of heterogeneous structures is historically based on the mechanics of liquids and is tightly connected with the problem of statistical description of behavior of particles systems (Torquato 2000; Binder and Heermann 2010). The most common approach is to investigate heterogeneous media at different scales. The microstructural peculiarities are usually being studied within the concept of representative volume element (RVE). This element can be defined as the smallest material's volume at which the properties of phases

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can be distinguished and for which effective (homogenized) properties can be measured.

RVE is usually being characterized at two scales. At the lowest, which is yet by several orders larger than the atomic scale, constitutive behavior at the material point is described using traditional continuum equations. Local properties and fields at this scale oscillate when moving from one heterogeneity to another – that could be phases in composite materials or grains in polycrystals.

To avoid computationally expensive models involving a large number of structural elements at the microscale, the higher (macroscopic) scale RVE description is usually being used. It operates effective properties, which can be constant or varying smoothly, and are defined basing on the microstructural specifics.

Morphology of RVE plays an important role in modelling of effective thermomechanical and physical properties of random heterogeneous materials (Kanit et al. 2003; Khisaeva and Ostoja-Starzewski 2006). During the latest decades, a large number of models, theories and approaches were developed for characterization of inhomogeneous materials. Their description is given, for instance, in (Kamiński 2005; Buryachenko 2007; Kanouté et al. 2009). One of the direction is to attract statistics and theory of random function to describe random systems, which in most cases heterogeneous materials can be considered as.

There are several assumptions about the statistical properties of the microstructure that are commonly being taken. First of all, the statistical homogeneity hypothesis presumes that each RVE sampled from the material will have the same statistical properties (such as, for instance, volume fraction). This assumption is usually followed by the ergodicity hypothesis, which means that ensemble average of values over a set of materials' RVEs is equal to averaging over a single RVE.

This paper offers a review of techniques which are based on or utilize the statistical instruments for characterization of internal structure, effective behavior and local stress and strain fields of heterogeneous media. It is primarily focused on the approaches for characterization of randomly reinforced media, although some of the instruments are also suitable for the other classes of heterogeneous materials (the respective remarks are given in the text).

The section 2 of the paper is related to application of statistical descriptors to obtaining formalized information about microstructural morphology. Such information can be used both for stochastic mechanical models or as a ground for geometrical operations like media reconstruction. The most commonly used descriptors are introduced, their development in terms of various applications is discussed. Methods of geometrical modelling of random structures are also addressed.

The section 3 contains a summary of the homogenization approaches which in certain ways rely on statistical information about the microstructure. Recent advances, which justify involvement of multipoint and high order correlation functions, are presented.

The section 4 continues discussion of the methods of stochastic mechanics in application to investigation of local fields of stress and strain in components and interfaces between them. This is determined by necessity of rigorous estimation of the critical zones when it comes to analysis of damage and non-linear behavior. Statistical moments for fields of stress and strain can be introduced for particular elements of the microstructure and give opportunity to indicate local changes and the respective response of microscopic fields.

Microstructural description

Heterogeneous media characterization

Among the important problems of modelling of heterogeneous materials is consideration of peculiarities of their random microstructure to the most possible extent. The morphology of random media microstructure can be studied as a complex system of many interacting components. Mathematical instruments that are used for description of such systems were developed within the individual discipline – mathematical morphology (Matheron 1975; Serra 1982).

Mathematical morphology was initially introduced for image analysis and was primarily aimed on formalization of microscopy images by extracting quantitative information (Matheron 1971, 1975; Najman and Talbot 2010). It is mostly based on set theory, lattice theory, topology and random functions. Capabilities of the mathematical morphology were significantly expanded from its emerging in 1964 during the last decades of twentieth century, from processing of 2D binary black and white images to 3D geometrical structures.

Particular instruments of mathematical morphology are widely used in materials science for characterization of complex internal structures of heterogeneous materials, such as experimentally obtained data from scanning electron microscopy (SEM), transmission electron microscopy (TEM), X-ray tomography (see Fig. 1). According to the methodology, images and structures are subject to discretization and transformation prior to quantification. For instance, microstructure image data in a form of 2D and 3D arrays can be reduced by thresholding to a binary array, which is mathematically described as a random set (Chiu et al. 2013). Later, automatic procedures can be applied to determine desired measurements and various statistical estimates.

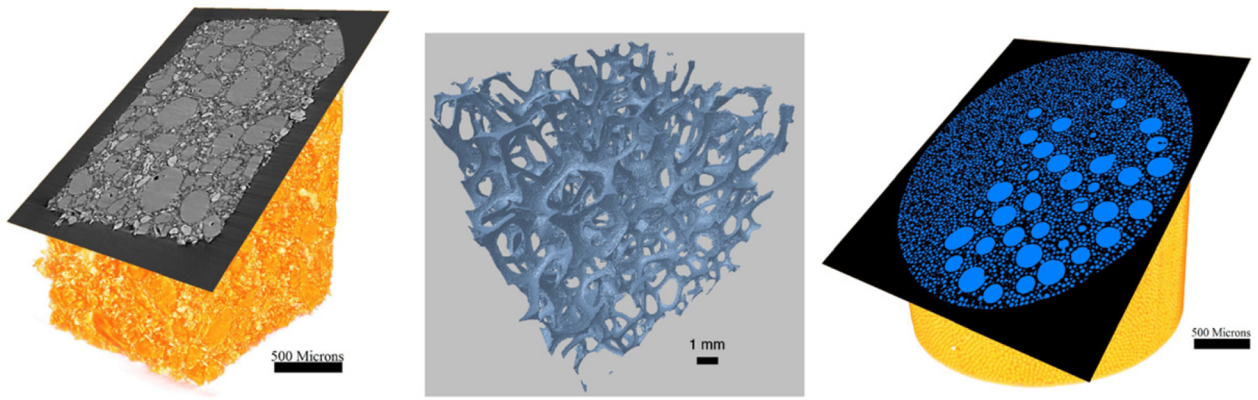

Fig. 1 Examples of micro-CT obtained microstructure: (**a**) Heterogeneous propellant, (**b**) Synthetic foam, (**c**) Glass beads (Lee et al. 2011)

Calculating the average values (or first (lower)-order statistics) of local descriptors was the traditional way for characterizing and quantifying microstructure (Fullwood et al. 2010). Such metrics are often introduced as distribution functions which deliver probability of presence of a particular local state in materials point. The simplest characteristic, which is volume fraction of each instance, can be calculated as proportion of the area or volume covered by one of the binary values. Another example could be the orientation distribution function (ODF) for polycrystalline microstructures.

More complicated descriptors originate from random field theory and are introduced with the functions applied over the random sets of data. Among the most commonly applied statistical descriptor functions are n-point probability functions, surface correlation functions, pore-size functions, cluster functions, nearest neighbor functions, linear path functions and others (Matheron 1975; Torquato 2002).

One of the most widely used statistical instrument is correlation functions. These functions show correlation between random values, which, for instance, are spatial positions of heterogeneities in matrix. They are one of the simplest statistical measures that contain spatial information and yet are much more informative than one-point statistics (Fullwood et al. 2010). The correlation functions themselves can be constructed for any random field inherent to heterogeneous media. In general, geometrical characterization is performed with random indicator function field, for which n-point spatial correlation function for the RVE can be defined as:

$$S_{\lambda_\alpha{}^{(n)}}\left(\overrightarrow{r}, \overrightarrow{r}_1, \ldots, , \overrightarrow{r}_n\right) = \left\langle \lambda_\alpha\left(\overrightarrow{r}_1\right)\lambda_\alpha\left(\overrightarrow{r}_2\right)\ldots\lambda_\alpha\left(\overrightarrow{r}_n\right)\right\rangle \tag{1}$$

where $\lambda_\alpha\left(\overrightarrow{r}\right)$ is the indicator function, which is equal 1 if the position of the radius-vector $\overrightarrow{r}$ indicates phase α and 0 in any other case; $\lambda_\alpha\left(\overrightarrow{r}\right)$ is the mean value of the function averaged over the RVE, which is equal to the volume fraction of phase α. The latter representation applied to other random fields will be discussed in section 3 within the perturbation approach. The higher-order correlation functions for the indicator function of two-phase RVE with polydisperse spherical inclusions are presented on Fig. 2.

The other definition of the correlation functions is suggested by (Torquato 2002) who introduced the n-point correlation function as:

$$P_{S^{(n)}}\left(\overrightarrow{r}_1, , \overrightarrow{r}_2, , \overrightarrow{r}_3, \ldots, , \overrightarrow{r}_n\right) = p_S \tag{2}$$

where p_S is probability that n points represented by radius-vector are in local state S. These functions were used by many authors in expressions for determination of effective properties, such as conductivity, elastic moduli, fluid permeability.

There are several methods of obtaining values of correlation functions for plain images or digitized three-dimensional models. The traditional way is sampling the image via set of randomly placed vectors and calculating local state at the points (Yeong and Torquato 1998a). The specific image processing algorithms were applied by Berryman (Berryman 1985). Cule and Torquato as well as Fullwood (Cule and Torquato 1999; Fullwood et al. 2008) used fast Fourier transform techniques. The rigorous description of the procedures and algorithms of obtaining values and applications of correlation functions can be found, for example, in works of S. Torquato (Torquato 2002) and D.T. Fullwood (Fullwood et al. 2010). Layered fast spherical harmonics expansion were proposed for representation of the correlation functions in (Li et al. 2010).

Application of correlation functions in engineering analysis had started with development of X-ray

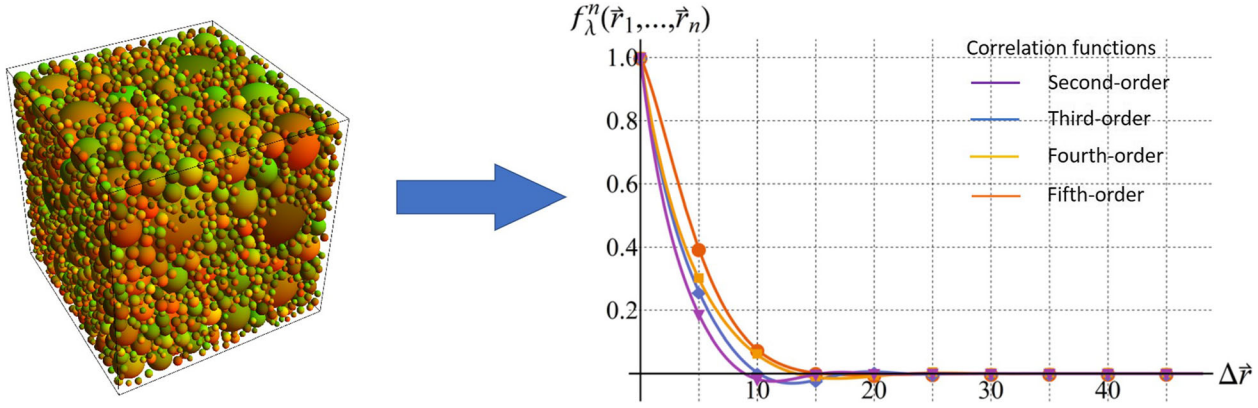

Fig. 2 High-order correlation functions for RVE of media with polydisperse particles

scattering and diffraction. One of the fundamental works (Debye et al. 1957) studied application of correlation functions to characterization of X-ray scattering data from porous materials with random distribution of microstructural properties. The paper (Corson 1974) delivered methodologies that link properties of two-phase structures to the experimentally calculated two-, and three-point probability functions. Small angle X-ray scattering technique has been used to get information on the distribution of inclusions and dispersion of particles (Walenta 1985). Being used in the analysis of effective properties or microstructural stress and strain fields, correlation functions can sense the influence of microstructural morphology parameters on the studied values (Saheli et al. 2005; Fullwood et al. 2010). A number of works were devoted to implementation of high-order multipoint correlation functions in characterization of the microstructure. High-order correlation functions can be approximated using lower order functions. Such approach was implemented in (Remond et al. 2016).

The technique based on n-point correlation functions is applicable not only to randomly reinforced types of materials. The concept of n-point orientation correlation function applied to polycrystals is presented in (Huang 2005). N-point correlations for multi-phase materials and polycrystals are also discussed in (Groeber et al. 2008). Spatial clustering of 3D microstructures obtained from the vertical metallographic planes was analyzed by means of two-point correlation functions in (Tewari et al. 2004). The paper (Hansen et al. 2003) incorporates results of the exhaustive branching technique, which is an alternative approach for reconstructing discrete microstructures from two-point correlation functions, applied to polycrystalline microstructures. This technique showed some advantages before annealing methods. Gaussian random fields were used by Roberts (Roberts and Teubner 1995; Roberts 1999). Principles of genetic algorithms for media reconstruction with correlation functions were applied by Basanta and Kumar (Basanta et al. 2003; Kumar et al. 2008). Research presented in paper (Fullwood et al. 2008) proposes novel algorithms based on phase-recovery methods used in signal processing. More case studies were discussed in (Jeulin 2000; Bochenek and Pyrz 2004; Al-Ostaz et al. 2007). The authors (Baniassadi et al. 2012) investigated the minimum required size of RVE basing on convergence of the independent correlation functions.

In general, several authors have mentioned limitations of the lower-order correlation functions that led, for example, to unallowable error estimate for the original and reconstructed media. It was also shown that utilizing 2D sections as a reference for 3D reconstruction puts limits on possibilities to consider certain morphological features intrinsic specifically to 3D structures. This has required introducing the supplement functions, capable to capture specifics of the internal geometry in order to perform more sophisticated analysis. Some of these functions are introduced below.

Surface correlation functions capture information about the spatial distribution of interfaces in the material and are based upon the surface indicator function $\chi_\alpha(\overrightarrow{r}_1)$, which is nonzero only at the interface of phase α (Torquato 2002; Fullwood et al. 2010). Such two-point functions can contain both surface and phases indicator functions:

$$F(\overrightarrow{r}_1,,\overrightarrow{r}_2) = \langle \chi_\alpha(\overrightarrow{r}_1)\lambda_\beta(\overrightarrow{r}_2)\rangle \tag{3}$$

Some functions were introduced due to needs of taking into account the features of the specific types of media. Thus, pore size distribution function $P(\delta)d\delta$ was introduced to characterize porous media and shows probability that a randomly chosen point in RVE lies at the distance interval $[\delta, \delta + d\delta]$ to the nearest point belonging to pore-solid interface.

In case when the microstructure of heterogeneous materials is not statistically homogeneous, the particles in RVE may form groups and clusters. The cluster function $C^{(\alpha)}$

$\left(\overrightarrow{r}_1, , \overrightarrow{r}_2\right)$ gives the probability that two points $\overrightarrow{r}_1$ and $\overrightarrow{r}_2$ are in the same cluster of the phase α. This function was described and used in (Torquato 2002; Jiao et al. 2009; Liu et al. 2013).

The lineal path function $L^{(\alpha)}(z)$ defines probability that a randomly sampled line segment of the specified length lies inside the particular phase. The same approach is used to introduce chords - all of the line segments which are formed by intersections of an infinitely long line with the two-phase interface. The chord length distribution function $C^{(\alpha)}(z)dz$ outputs probability of finding a chord of length between z and $z + dz$ in phase α. The chord length distribution and the lineal path function are two common measures that contain information on particle (or grain) shape that is independent from spatial distribution. The applications examples of these two measures are transport problems, fluid permeability (including fluid flow through porous media), stereology.

The group of the nearest neighbor functions is associated with particle-reinforced media and analyses probability of finding the nearest neighbor at some given distance from a reference particle. A number of variations of these functions that differ by the way of definition of the sampled distance and reference points, which can be distance between centers of the particles, between boundaries of the particles, between arbitrary points in RVE and particle's center. Reverse functions may be introduced, for instance, to estimate spaces in matrix which are free of the particles.

Lineal path function and nearest-neighbor function were used for accurate statistical media reconstruction (Quintanilla and Torquato 1997; Yeong and Torquato 1998a; Hahn et al. 1999; Davis et al. 2011).

As was mentioned above, some specific statistical descriptors can be introduced for characterization of polycrystalline materials (Groeber et al. 2008). Thus, the orientation distribution function is a measure of the macroscopic texture of a material. The misorientation distribution function provides information on the local arrangement of grains. The micro-texture function is a measure of the amount of clustering of orientations and misorientations.

The summary of major morphological descriptors is presented in Table 1. Order of the function determines whether the function is basic for the microstructural property or is a derivative of basic descriptor. For instance, the indicator function is lower order since it extracts particular feature of the microstructure, while the correlation functions are of higher order since they are constructed from the lower-order indicator functions. The «Measure» column gives brief description of main characteristic that the function delivers. There is also information about the type of material for RVE of which the function can be applied as well as the dimension of sample required to experimentally obtain the values of the function (three-dimensional data or plain two-dimensional image).

The mathematical properties and characteristics of descriptors that have been introduced for microstructure of heterogeneous media and materials are comprehensively studied in (Serra 1982; Sobczyk and Kirkner 2001; Torquato 2002; Fullwood et al. 2010).

Application of the mathematical morphology instruments for microstructural characterization of heterogeneous media allows to establish quantitative microstructure-property link and evaluate impact of important inherent features such as clustering, percolation, dispersion, orientation on overall mechanical properties using the specific type sensitive descriptors. This theoretically underpins the concept of design of materials with prescribed properties by controlling microstructure during manufacturing and processing.

For the materials with predominantly periodic microstructure, such as textile composites with woven and braided microstructure, some of the above described statistical correlation instruments can be used to evaluate deviations from the periodic trends (Cox et al. 2014).

Table 1 Main morphological descriptors and their properties

Function	Order	Measure	Material	Sample dimensions
Indicator function	Lower	Presence of phase	Any random media	2D and 3D
Correlation function	Higher	Spatial correlation	Any random media	2D and 3D
Surface correlation functions	Higher	Interfaces distribution	Any random media	2D and 3D
Pore size distribution function	Lower	Level of porosity	Porous media	Only 3D
Cluster function	Higher	Particles clustering	Any random media	Only 3D
Lineal path function	Lower	Connectedness of microstructure	Any random media	2D and 3D
Chord length distribution function	Lower	Connectedness of microstructure and particles shape	Any random media	2D and 3D
Nearest neighbor functions	Lower	Particles distribution	Particle-reinforced media	2D and 3D
Orientation distribution function	Lower	Grain orientation, macroscopic texture	Polycrystals	Only 3D
Misorientation distribution function	Lower	Orientation of local arrangement of grains	Polycrystals	Only 3D
Micro-texture function	Lower	Amount of clustering of orientations and misorientations	Polycrystals	Only 3D

They also help to understand how randomness in microstructure affects the properties of composites.

Heterogeneous media reconstruction

An important practical application of the statistical descriptors is mathematical reconstruction of RVEs based on the reference sample experimental data, formalized in a form of statistical descriptors. Such procedure helps to avoid repeating of imaging techniques each time when the microstructure model is required. Created microstructure is assumed to correspond to all relevant microstructural features.

S. Torquato offered the method which involved lattice-type representation of the discretized structure and application of correlation functions (Yeong and Torquato 1998a). It is aimed on minimizing difference between actual and target correlation functions by simulated annealing. In order to tackle high computational demands of the method, the discrete fast Fourier transform is used (Fullwood et al. 2008). The other reconstruction scheme relies on correlated random Gaussian field and assigns a region the material phase property if the value of a Gaussian random variable is within the specified range. This approached was mathematically developed in further works. Its limitations relate to inconsistency when processing more than two-phase media and Gaussian character of field requirement (Rozman and Utz 2001).

Correlation functions, obtained from the experimentally gathered data, allows to create geometrical models of microstructure that are statistically corresponding to the reference real ones. The layout of this procedure is presented on Fig. 3.

The goal of the reconstruction algorithms is to minimize functional:

$$E = \sum_{\vec{r}} \left[S_2^{\alpha}(\vec{r}) - S_2^{\alpha}(\vec{r}) \right] \tag{4}$$

where $S_2^{\alpha}(\vec{r})$ is target two-point correlation function, $S_2^{\alpha}(\vec{r})$ is two-point correlation function of the reconstructed microstructure.

Some techniques allow to restore 3D structure from information extracted from 2D sections (Liu et al. 2013), which is helpful when three-dimensional imaging is unavailable.

S. Torquato investigated the problem of reconstruction of one- and two-dimensional microstructures based on stochastic optimization involving lower order correlation functions (Yeong and Torquato 1998a; Torquato 2002). Particularly, annealing and Monte-Carlo techniques are used to minimize an error between

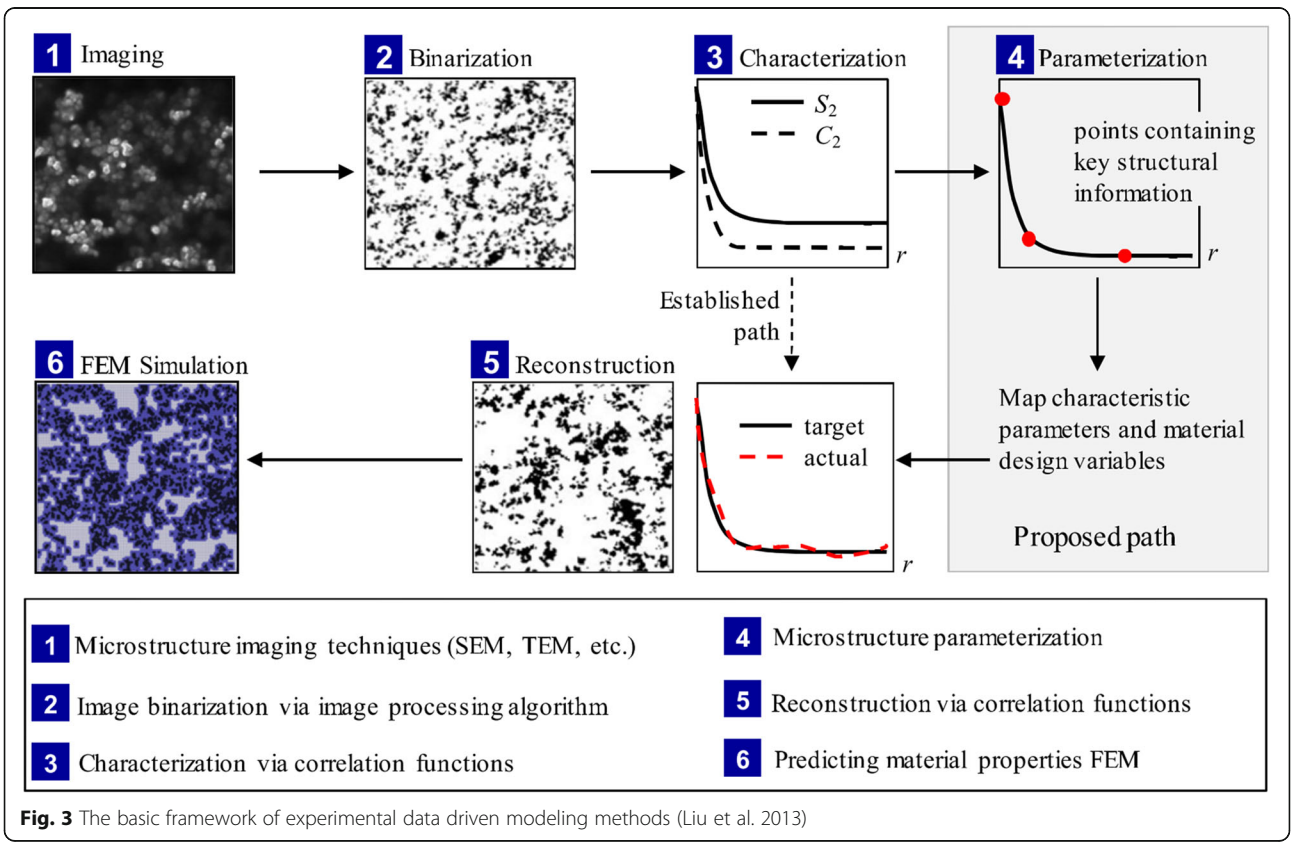

Fig. 3 The basic framework of experimental data driven modeling methods (Liu et al. 2013)

correlation functions of model and sample. C. Yeong and S. Torqauto offered a procedure that incorporates a set of statistical correlation functions obtained from a two-dimensional image of the heterogeneous medium to reconstruct the medium in 3D (Yeong and Torquato 1998b). The artificial anisotropic effect which appears in this case and the ways of its consideration were discussed in (Sheehan and Torquato 2001). Reconstruction of random media using lower order correlation functions was later improved by Y. Jiao and S. Torquato in (Jiao et al. 2007, 2008), where the lattice-point procedure for sampling the digitized reference data as well as the modification of the optimization algorithm was described. Such approach was also implemented and investigated in (Manwart and Hilfer 1999; Rozman and Utz 2001; Talukdar et al. 2002). The other suggested variations of the reconstruction procedure included introducing different optimization techniques such as simulated annealing and maximum entropy. Necessary conditions on the two-point correlation functions were derived in (Torquato 2006).

Further developments of the above described method in the part of Monte-Carlo procedure are presented in works of Baniassadi et al. (Garmestani et al. 2009; Li et al. 2010). The latter study was particularly applied to reconstruction of waviness, geometry and preferential distribution of carbon nanotubes (CNTs). These authors also investigated solid oxide fuel cell materials, for which three-dimensional reconstruction was performed using planar section of multi-phase composites (Baniassadi et al. 2011). 2D realization of the microstructure obtained from SEM images of carbon black particle fillers dispersed in rubber was studied in (Deng et al. 2012). The work (Sheidaei et al. 2013) was devoted to 3D reconstruction of nanotube polypropylene composite from focused ion beam (FIB) and SEM images. It was shown that two-point correlation functions can only produce an approximate reconstruction while the other groups of statistical descriptors are required to obtain better results. The problems of experimental measurement of the correlation functions are also discussed and studied in (Remond et al. 2016). Three-dimensional reconstruction technique using three-point third order correlation functions was introduced in (Baniassadi et al. 2012).

Generation of random microstructures

Types of microstructure of heterogeneous materials can be grouped into two main classes. First of them can be defined as a statistical mixture and is represented by complex interpenetrating frameworks of constituents, each of which has its own individual bearing capacity. The second group consists of materials that are formed by matrix reinforced with inclusions of spherical, lamellar, fibrous, elliptic and other forms. Some multicomponent composites may also have mixed types of structures. Parameters influencing the RVE geometry are the shape and size of the inclusions, the volume fraction and the morphological details like the spatial orientation and spatial distribution of the inclusions (Bailakanavar et al. 2012). To be statistically representative, RVEs should contain sufficient information about these features.

While now modern simulation methods allow to create geometrical models of RVEs with wide range of forms of phases and overall complexity, the simulation of random microstructures had begun with modelling of random arrangement of spheres (Fig. 4). Such microstructures initially were used in models for simple liquids, concentrated suspensions, amorphous and powder materials. Further a brief description for development of models for RVE random packing is given.

The advances in modeling of dense disordered structures are mostly connected with the two groups of approaches: random sequential adsorption (RSA) and dynamic (or concurrent construction) methods.

Sequential approaches presume determined or random generation of microstructural particles. The first inclusion is placed with its center at the chosen position inside a RVE. Next inclusion and center point is randomly chosen from the diminished volume in the RVE. Likewise, the process of sequentially and randomly positioning an inclusion is continued till the desired volume fraction is achieved or till the jamming limit is encountered. This method generates unit cells with non-intersecting inclusions wherein the gap between the inclusions is user-defined, typically of the order of inclusion size (Bailakanavar et al. 2012).

Sequential placement was studied by Bennett (Bennett 1972), who describes a consistent method when each new inclusion is placed at the point closest to the first so that it comes into contact with the existing inclusions. Intersection is controlled by testing the distance between the centers. Later, this method was modified by introduction of a parameter characterizing the filling of the area bounded by the tetrahedron (Lu et al. 1994). The algorithm that allows to control clustering of inclusions was proposed in (Kansal et al. 2000). The variation when boundary point of each new inclusion coincide with an arbitrary point on the surface of existing inclusions located at the bottom of the virtual box is used in (Nolan and Kavanagh 1992; He et al. 1999; Yang et al. 2000; Furukawa et al. 2000).

Typical particles in heterogeneous materials are often have different shape and size (see Figs. 4, 5 and 6). The RSA algorithm was used to create geometry of RVE with disks (Hinrichsen et al. 1986), spheres and spheroids (Sherwood 1999; Segurado and Llorca 2002; Ghossein and Levesque 2012), cylinders and sphero-cylinders

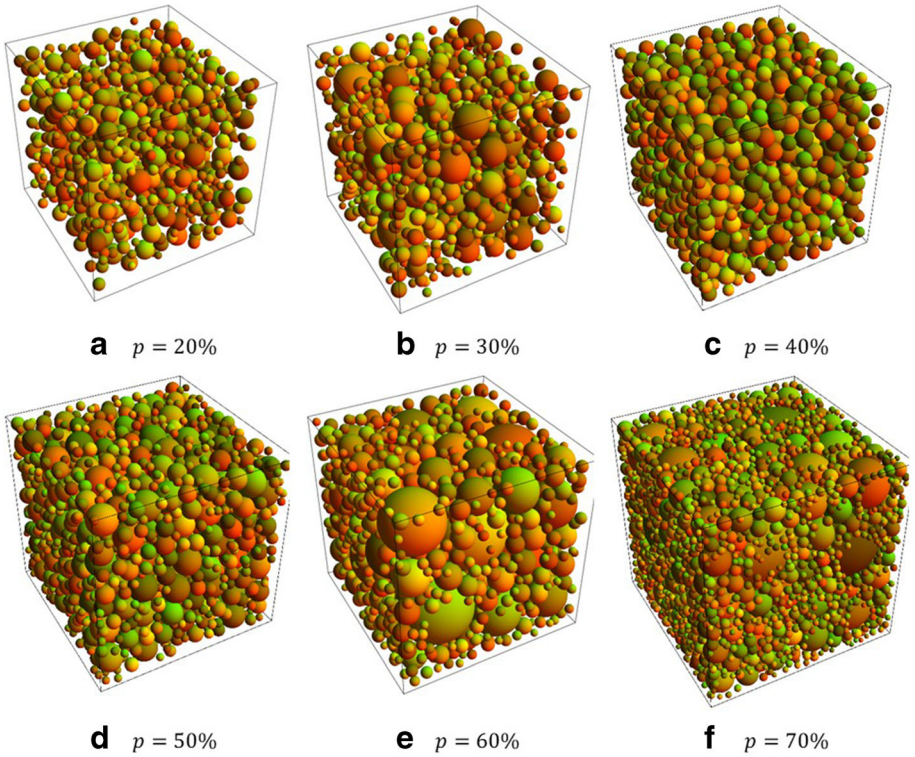

Fig. 4 RVEs of polydisperse random structures with spherical inclusion of various volume fraction *p: (a) 20%, (b) 30%, (c) 40%, (d) 50%, (e) 60%, (f) 70%*

(Böhm et al. 2002; Williams and Philipse 2003; Iorga et al. 2008; Redenbach and Vecchio 2011; Zhao et al. 2012), thick fibers and ellipsoids (Evans and Ferrar 1989; Parkhouse and Kelly 1995; Man et al. 2005; Bezrukov and Stoyan 2007).

The RSA algorithm is generally limited by the volume fraction it can reach (Evans and Gibson 1986; Parkhouse and Kelly 1995; Toll 1998; Williams and Philipse 2003). Methods aimed on increasing this limit up to 35% volume fraction were proposed by (Pan et al. 2008). The hierarchical random sequential adsorption introduced by (Bailakanavar et al. 2012) allowed to reach 45%. The cylindrical inclusions can be packed with the volume of 50% using the approach that is described in (Islam et al. 2016), while the same volume fraction can be obtained for spherical inclusions with the denser packing algorithm proposed by (Segurado and Llorca 2002).

The distinct class of the random heterogeneous materials are cellular materials with voids, such as foams, wood, bones, etc., and, consequently, low matrix volume fraction. In this case, voids can be either topologically connected or unconnected. The geometrically simplest

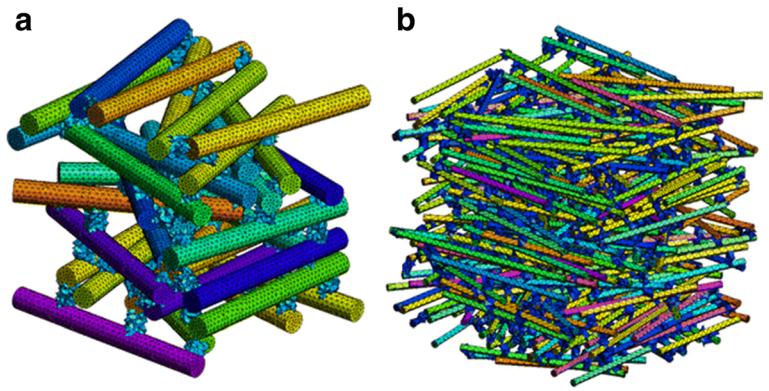

Fig. 5 (**a**) RVE with short-fiber, volume fraction 5% and cross section radius equal to 2.4, (**b**) RVE with short-fiber,volume fraction 10% and cross section radius equal to 0.6 (Bailakanavar et al. 2012)

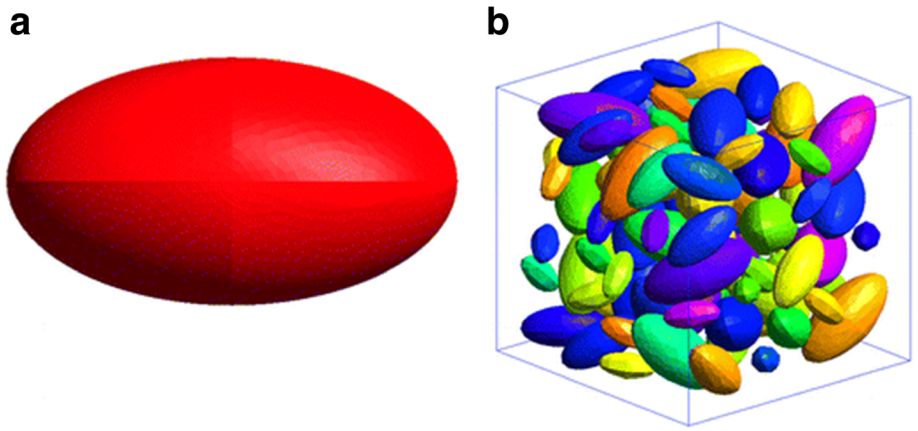

Fig. 6 Geometry of (**a**) an ellipsoidal inclusion and (**b**) an RVE with ellipsoidal inclusions (Bailakanavar et al. 2012)

topologically connected cellular materials are regular honeycombs, the in-plane behavior of which can be modeled with planar hexagonal cell models. However, unconnected voids require complex three-dimensional geometry modeling: inclusions take forms of cubes (Santosa and Wierzbicki 1998) and polyhedrons (Grenestedt 1998; Sihn and Roy 2004; Daxner et al. 2007; Su et al. 2014; Zhang et al. 2014; Böhm and Rasool 2016). Voronoi tesselations have become a common tool in modeling irregular foams, and recently random Laguerre tessellations have been proposed for generating periodic multi-cell models (Böhm 2004; Redenbach 2009).

Inclusions of complex forms are essential part of digitized geometrical representation of polycrystalline materials (see Fig. 7). Thus, for instance, within the monodispersive grain size model microstructural grains are replaced by polyhedral inclusions. Voronoi tesselations are a widespread approach for the geometrical models of polycrystals, where they are used for partitioning subregions that correspond to individual grains (Lazar et al. 2012). The grains are constructed around the centers that are distributed randomly in space. (Gross and Li 2002) combined the Monte-Carlo and the Voronoi tessellation methods for grain-growth to generate nanocrystalline structures with specified distributions of grain size. Molecular dynamics simulations and equilibration were applied within analysis of nanocrystalline materials in (Tomar and Zhou 2007).

The methods described above belong to a class of static methods and presumes that positions of inclusions are fixed after synthesis. Dynamic models allow reorganization of the structure depending on a way of interaction of inclusions with each other (Buryachenko and Pagano 2005). Various modifications of this method can be found, for example, in (Clarke and Wiley 1987; Oger et al. 1999; He and Ekere 2001; Knott et al. 2001).

Method for generation dense structures with non-intersecting inclusions is proposed in (Bennett 1972). Each consequently placed inclusion "moves" in the direction of the others along a straight line connecting their centers until the contact condition is met. In more complex models a system of differential equations is solved for specifying movements and detecting intersections (Knott et al. 2001).

There is also a method of achieving the desired density of inclusions by changing the radii of spheres (see Fig. 4d-f). In some models, each inclusion is subject to small random movements, regardless of position of the neighbor inclusions, until the best possible position for denser packing of inclusions is found (Berryman 1983; Cheng et al. 2000). The Hard-Core model, also called random sequential adsorption model, was discussed by (Lotwick 1982; Hinrichsen et al. 1986).

In addition, the models of inhomogeneous structures can be obtained by Monte Carlo methods. Some models of random structures are based on periodic lattices. Two of them are described in (Kroener 1986): according to

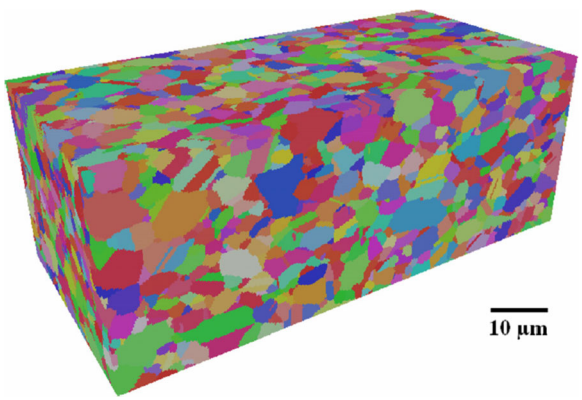

Fig. 7 3D reconstruction of polycrystalline superalloy (Groeber et al. 2008)

the first, inclusion centers are aligned with randomly selected lattice nodes, while each cell were assigned to some phase forming what is now known as voxel model.

Another type of synthesis algorithms with presumed particles movement is based on molecular dynamics (Lubachevsky and Stillinger 1990; Lubachevsky et al. 1991). Each inclusion is assigned a random velocity vector and then are set in motion. Simultaneously, their volumes gradually increase. When a binary collision occurs, the velocities of the two concerned particles are updated according to the kinetic energy conservation principle. To meet the periodicity conditions, if particle leaves the cell through a face during movement, it appears from the opposite side. This approach allows to create the densest packing structures – up to 74% volume fraction for polydisperse spheres (Ghossein and Lévesque 2013). The similar method was used for non-spherical particles as well (Donev et al. 2004).

Effective properties

In addition to solving the problems of morphological analysis, statistical descriptors can be used as an instrument for establishing the relationship between microstructure, local and effective properties of heterogeneous media. Finding suitable relationship between distribution, shape and properties of each constituent and macroscopic response of the heterogeneous material is one of the most investigated questions of micromechanics. Among the variety of the homogenization methods there is a broad class of those rely on statistical approach. Some advantages in these methods are introduced below.

First of all, one of the ground concepts uses perturbation (or fluctuation) representation, which presumes decomposition of the local stress and strain fields as well as local stiffness tensor into an averaged value and an addition:

$$\sigma_{ij}(\overrightarrow{r}) = \langle \sigma_{ij}(\overrightarrow{r}) \rangle + \sigma'_{ij}(\overrightarrow{r}) \tag{5}$$

$$\varepsilon_{ij}(\overrightarrow{r}) = \langle \varepsilon_{ij}(\overrightarrow{r}) \rangle + \varepsilon'_{ij}(\overrightarrow{r}) \tag{6}$$

$$C_{ijkl}(\overrightarrow{r}) = \langle C_{ijkl}(\overrightarrow{r}) \rangle + C'_{ijkl}(\overrightarrow{r}) \tag{7}$$

where $\sigma'_{ij}(\overrightarrow{r}) = \varepsilon'_{ij}(\overrightarrow{r}) = C'_{ijkl}(\overrightarrow{r}) = 0$, $\overrightarrow{r}$ is radius-vector with components (x_1, x_2, x_3). The angled brackets indicate ensemble averaging, which is equal to volume averaging when the hypothesis of ergodicity for statistically homogeneous material is assumed. The averaging procedure for any coordinate-dependent value $f(\overrightarrow{r})$ is determined as:

$$\langle f(\overrightarrow{r}) \rangle = \frac{1}{V} \int_V f(\overrightarrow{r}) dV \tag{8}$$

where integration is performed over the representative volume V.

The perturbation method is usually suitable for characterization of weakly inhomogeneous media (Lomakin 1970; Beran and McCoy 1970), which means that they are expected to give correct results in case of small differences between the components properties (Shermergor 1977) or small concentration of inhomogeneities (Jeffrey 1973; Lu and Song 1996).

The mean field approach facilitates the calculation of the averaged field quantities in the constituent phases and resultant determination of the effective properties (Budiansky 1965; Hill 1965; Willis 1992; Benveniste 2008).

Models which are built in terms of the mean field approach introduce strain concentration tensor as the functional connecting volume averaged strain for RVE and each phase:

$$\langle \boldsymbol{\varepsilon} \rangle_C = \boldsymbol{A} : \langle \boldsymbol{\varepsilon} \rangle^* \tag{9}$$

where $\boldsymbol{\varepsilon}^*$ is homogeneous strain, $\boldsymbol{\varepsilon}_C$ is averaged strain in a phase C.

One of the homogenization schemes is represented by the self-consistent methods, according to which analysis is performed for a single particle of a typical constituent embedded into the effective medium instead of studying the whole RVE embedded in a homogeneous medium.

Under the ergodic assumption, statistical measures such as N-point statistics can be used in equations of the averaging schemes (Adams et al. 2005; Fullwood et al. 2007) that combine decomposition of the perturbation method and mean field homogenization relation by means of localization tensor $\boldsymbol{a}(\overrightarrow{r})$, which can be obtained from the information regarding the local response of a material (Fullwood et al. 2010):

$$\boldsymbol{\varepsilon}'(\overrightarrow{r}) = \boldsymbol{a}(\overrightarrow{r}) \langle \boldsymbol{\varepsilon} \rangle^* \tag{10}$$

The solutions based on Green's functions, that are used in these methods, include correlation functions of different orders. Thus, localization tensor $\boldsymbol{a}(\overrightarrow{r})$ can be defined with the Green's function as follows:

$$a^{(\chi)}_{ijkl}(\overrightarrow{r}) = \frac{1}{2} \int_{V_1} \left(G_{im,j}(\overrightarrow{r}, \overrightarrow{r}_1) + G_{jm,i}(\overrightarrow{r}, \overrightarrow{r}_1) \right) \left[C'_{mnpq}(\overrightarrow{r}_1) + C'_{mnpq}(\overrightarrow{r}_1) a^{(\chi-1)}_{pqkl}(\overrightarrow{r}_1) \right]_{,n} dV_1 \tag{11}$$

where $\boldsymbol{G}$ is the Green's function, C' is fluctuation of stiffness tensor (7) that can be expressed via statistics and structural correlation functions for the RVE, χ denotes order of expansion of the series. The Green's function type is determined by the properties of medium, while the order of the series expansion χ defines the order of required statistical descriptors presumed by fluctuation tensor C'.

The Green's function in (11) has a singularity in point $\overrightarrow{r} = \overrightarrow{r}_1$. The approaches that helps to overcome it and some exact expressions are obtained in (Shermergor 1977; Kroener 1986; Torquato 1997, 2002). The results of application of the first and second derivatives of the

Green's function to resolve this issue is discussed in (Tashkinov 2015).

Series of works by S. Torquato were devoted to application of perturbation method and integral equations with Green's function to derive effective conductivity and elastic properties for two-phase media. Some exact solutions for the specific types of media are presented in (Torquato 2002).

Some other statistical models that focus on local response are based on the percolation theory and Weibull analysis (Jayatilaka and Trustrum 1977; Frary and Schuh 2005; Chen and Schuh 2006), and may also be used to connect micro- and macro- properties depending on statistical information about the microstructure. Alternative approaches rest upon neural networks, which improve fitting models for modeled and observed microstructural data (Bhadeshia 1999).

Spectral methods, incorporating microstructural correlation functions, use spectral representations of distribution functions that statistically characterize the internal structure of representative volumes and connect these representations and macroscale effective properties using existing homogenization schemes (Kalidindi and Houskamp 2007).

The known fact is that it is impossible to achieve exact prediction for effective properties of random media, so rigorous assessment can be done in the form of bounds. One of the most commonly used bounds are based on variational principle according to which the effective parameter is expressed in terms of functional for which the variational (extremum) principle is being formulated. Primarily, variational methods for homogenization purposes analyze approximate fields substituted into energy bounds by averaging the energy density (Hashin and Shtrikman 1963), while classical variational methods employ minimum energy principles to generate bounds on effective properties. The microstructural information is considered via different types of correlation functions (Willis 1981; Torquato 1991; Ponte Castaneda and Suquet 2001; Milton 2002). The correlations which are required for calculation of the bounds of effective values using variational methods express, for instance, probability of finding n points with position $r^n = \left\{ \overrightarrow{r}_1, \ldots, , \overrightarrow{r}_n \right\}$ in phase i for statistically inhomogeneous media, finding a point with position $\overrightarrow{r}_1$ in the exterior to the phase particles and any n particles with coordinates r^n, finding points at the interface, finding a nearest neighbor at radial distance r from the origin. Each type of these functions depends upon the relative position of the n points inside the RVE. The bounds of the effective properties are usually characterized by the number of points involved in the correlation functions (Torquato 1991).

Improved bounds, which were developed later and are tighter than the traditional Hashin-Shtrikman bounds, depend on high order correlation functions rather than volume fractions. Three-point bounds were developed in (Beran 1971; Torquato 1980, 1991; Milton 1982; Castaneda 1998; Agoras and Ponte Castañeda 2011; Ponte Castañeda 2012). Mikdam and Baniassadi (Mikdam et al. 2009, 2010, Baniassadi et al. 2011, a) have applied the strong-contrast formulation to predict the effective electrical and thermal conductivities of a two-phase composites using two-point and three-point correlation functions. The current limitation is that in the case of strong inhomogeneity of microstructure these bounds might not be helpful because they tend to diverge with increasing contrast between the properties of the constituents (Torquato 2000; Buryachenko 2007).

Large amount of work on developing of the variational estimates of effective properties of heterogeneous media has been done by P.P. Castaneda, the recent developments in that area as well as review of the previous research can be found in (Ponte Castaneda 2016).

In order to simplify modeling, the real stochastic structure can be replaced by some specific one, which is then being analyzed. Among such substituends are fractal-type media based on spherical or cylindrical coated inclusions (Hashin and Shtrikman 1962; Benveniste and Milton 2003), two-phase composites with statistically equivalent phases (Dykhne 1970), regular structures (Keller 1963; McKenzie et al. 1978; McPhedran and McKenzie 1978), unit cell with randomly dispersed inclusions (Kushch 1997; Guseva and Lusti, 2004) or percolation models (Sahimi 1998).

There are many other homogenization techniques exist that utilize different instruments for characterization of specifics of microstructure as well as linear and nonlinear behavior of materials. For instance, Rasool and Boehm (Rasool and Böhm 2012) compared inhomogeneity shape effects on the linear elastic, thermoelastic and thermal conduction effective responses of composites. Klusemann (Klusemann et al. 2012) studied three strategies for dealing with inhomogeneities of non-elliptical shape in the context of homogenization method: mean-field methods used in combination with analytical expressions for the Eshelby tensor, Mori-Tanaka method in combination with the replacement tensor approach and direct Finite Element discretization of microstructures. The review of multiscale methods for modelling mechanical and thermomechanical responses of composites can be found, for example, in (Kanouté et al. 2009).

Statistical characterization of local stress and strain fields

The problem of prediction of effective properties in some approximation can be solved involving only the averaging statistical descriptors. However, models of failure and nonlinear analysis are tied on the specific features of local stress and strain fields where microscale fluctuations must be taken into account. Special attention should be given to the interfaces between the

constituents and their vicinities as those are the most common zones of failure initiation. The random structure of heterogeneous materials leads to necessity of analysis of large number of system's realizations (Buryachenko 2007). To eliminate that, distribution of fields in random structures can be estimated using the statistical moments of stress and strain fields that can be obtained either for homogeneous RVE or for its constituents separately as well as for the interface between the matrix and inhomogeneities. At this case, the same techniques as was developed for analysis of effective properties can be applied. The importance of the stress fluctuations is discussed in details in (Buryachenko 1996; Ponte Castaneda and Suquet 2001) for a wide class of nonlinear problems of micromechanics such as plasticity, damage, viscosity, or creep. The multipoint statistical moments of the stochastic stress and strain fields are used as the characteristics of the deformation processes in the components of the material.

The above described perturbation approach as well as method of integral equations can be applied for calculation of statistical descriptors for the local stress and strain fields in microstructure.

A general scheme for calculating the second-order moments of the random elastic fields in the case of a composite of inclusion-matrix type using perturbation approach is presented, for example, in (Buryachenko 2007). Kroener (Kroener 1986) and Beran (Beran 1965) developed statistical mathematical formulations to link correlation functions to properties in multiphase materials.

The exact estimation of all components of the second moment tensor of the pure elastic and internal residual stresses using perturbation approach is given in (Buryachenko and Kreher 1995), where it was shown that the second moment of the stress field is constant within the inclusions if a homogeneity of random effective stress fields in the neighborhood of each ellipsoidal inclusion was assumed. Explicit relations for second moments of stresses were obtained in (Buryachenko and Rammerstorfer 1998) and utilized second and third order interactions between inclusions.

Combination of perturbation method and variational principle (Bergman 1978), which is based on the estimation of the perturbation of an energetic function due to a variation of the material properties, was developed for estimation of stress fluctuations for RVEs with isotropic components (Bobeth and Diener 1986; Kreher 1990). Xu (Xu et al. 2009) employed the generalized variational principles to decompose a boundary value problem with random microstructure into a slow scale deterministic problem and a fast scale stochastic one using a Green's function based multiscale method.

Recursive approach for computing a probability density function of stress fields is described in (Hori and Kubo 1998). Numerical statistical analysis at the inclusion scale level was performed by (Babuška et al. 1999).

Several exact solutions for second order moment of stress fields were offered for some cases of deterministic structures. One of such models is media with regular structure (Evans 1978; Fu and Evans 1985; TVERGAARD and HUTCHINSON 1988). However, their limitations are connected with lack of consideration of spatial distribution of constituents which may have a significant effect on the local stresses.

Method of integral equations allows estimation of second moments of stresses in the components. Considering both binary and triple interaction of the inclusions, explicit relations for second moments of stresses can be obtained (Buryachenko 2011). The method implies that description of heterogeneous medium can be implemented via structural-phenomenological approach when mechanical properties of microstructural components are defined with conventional phenomenological equations and criteria while microscopic strain and stress fields are computed using the solutions of stochastic boundary value problems (SBVPs) with rapidly oscillating coefficients. Fluctuations of displacements can be obtained by integration with the Green's function method:

$$u_i'(\overrightarrow{r}) = \int_{V_1} G_{ij}(\overrightarrow{r}, \overrightarrow{r}_1) \frac{\partial P_{jn}(\overrightarrow{r}_1)}{\partial x_{1n}} dV_1 \quad (12)$$

Here, $u_i'(\overrightarrow{r})$ is fluctuation of displacements, $G_{ij}(\overrightarrow{r}, \overrightarrow{r}_1)$ is the Green's function. $P_{jn}(\overrightarrow{r}_1)$ is a functional, containing constants from boundary conditions as well as stiffness tensor. The structure of the functional is different depending on the ways of solution.

The study of behavior of composites with random structures in these frameworks was established by works of Lifshitz and Rosenzweig (Lifshitz and Rosenzweig 1946) which were devoted to SBVPs of elasticity theory for polycrystalline media. Depending on the SBVP solution different statistical models can be distinguished. The numerous ways of closing the integral equations were offered – the method of effective medium (Hashin 1968; Buyevich 1992; Koelman and de Kuijper 1997), differential method (Milton 1985; Zimmerman 1996; Phan-Thien and Pham 2000), Mori-Tanaka-Eshelby method of the average fields (Hatta and Taya 1985; Benveniste 1986; Chen and Wang 1996; Weber et al. 2003), the singular approximation method (Shermergor 1977; Shvidler 1985), the strong isotropy hypothesis and method of conditional moments (Khoroshun et al. 1993), correlation approximation and multipoint approximation (Volkov and Stavrov 1978; Tashkinov et al. 2012; Tashkinov 2015). Some of the techniques are described below.

If composite's constituents are isotropic and RVE can be macroscopically considered an isotropic medium, the strong isotropy hypothesis can be used (Khoroshun et al. 1993). It assumes that for calculation of two-point moments the components that depend on a choice of direction between two points of can be neglected.

Method of conditional moments for random composites is based on an assumption that the fluctuations of random fields within a component are quite small. This allows to reduce the problem to a system of linear algebraic equations for one-point moments. Such modified problem is solved in two-point approximation using the statistical information and a number of simplifying hypotheses concerning the nature of the distribution of inclusions in the matrix volume (Khoroshun et al. 1993).

For a wide class of stochastic heterogeneous media models, periodic structure can be regarded as a realization of a random structure. The method of periodic components suggests that averaged parts of fields decomposition correspond to the periodic structure (Sokolkin and Tashkinov 1985; Pan'kov et al. 1997). Such representation allows taking into account fractional content, connectivity and geometric shape of components which are common both to the random and periodic structures.

Local approximation method is based on the specifics of short-range interactions of inclusions in matrix composites, according to which the problem of deformation of heterogeneous media is reduced to the simpler problem of deformation of an unbounded domain with an ensemble of a small number of inclusions. Feature of local interactions is not related to the specific nature of the mutual arrangement of the inclusions as well as to their shape, so the method has been applied for composites with random structures (Anoshkin et al. 1991). The hypothesis of limiting locality of correlation functions allows obtaining a single-point approximation of SBVPs and avoiding computation of the integrals over the field of statistical dependence of the correlation functions.

Many stochastic methods are based on the assumption of statistical independence of random fields of physical and mechanical properties of composites, which means that each geometric point is identified with a grain of heterogeneity and the fluctuations of physical and mechanical properties in the neighboring grains are not correlated, and, thus, only the one-point statistical characteristics of a random structure can be taken into account. For the composite with deterministic properties of the structural elements all one-point structural statistical characteristics are determined by relative volume concentration of elements in the assumption of homogeneity and ergodicity of the random fields.

The fundamental work by T.D. Shermergor (Shermergor 1977) offered a number of techniques for the weak contrast heterogeneous media. In the singular approximation (Shermergor 1977) only the formal component of the second derivatives of the Green's function for a homogeneous unbounded medium is retained. The correlation approximation also uses the formal component, however, it takes into account only pair interactions, so the correlation functions of a higher order than binary are disregarded. In general, the correlation theory can be applied when the standard deviations of the structural elastic moduli are small in relation to their mathematical expectation. Correlation approximation was developed in (Shermergor 1977; Volkov and Stavrov 1978). Stochastic methods in correlation approximation lead to good results for a small difference in the elastic modulus of the composite or weak anisotropy. The full correlation approximation assumes that all the terms, obtained in solution of the SBVP in the first approximation, are being considered, including those containing correlation functions of order higher than the second. The statistical characteristics in the full correlation approximation and second approximation of the solution of the SBVP were calculated in (Sokolkin and Volkova 1992; Tashkinov et al. 2012; Tashkinov 2014, 2016).

The second order moments of stress can be used in statistical models for nonlinear mechanical behavior and fracture of heterogeneous media. The crucial role of these moments in nonlinear analysis is explained by the fact that the yield surface, inclusions interface failure criterion and the energy release rate are the quadratic functions of the local stress distributions (Buryachenko 2011). Thus, Sakata (Sakata et al. 2012) discussed prediction of microstructural failure probability using Monte-Carlo simulation, the perturbation-based stochastic homogenization method and a stochastic multiscale stress analysis. Mishnaevsky (Mishnaevsky et al. 2004) studied the effect of particle clustering on the effective response and damage evolution in particle reinforced Al/SiC composites and used probabilistic analysis to determine failure of matrix and materials.

Conclusions

While comprehensive analysis of interconnection of multiscale parameters of heterogeneous materials remains a challenging problem of micromechanics, in some instances microstructure can be described statistically. This work reviews the main techniques and methods for multiscale mechanical analysis of random heterogeneous materials by means of statistical descriptors and instruments. Experimental and numerical reconstruction based on statistical descriptors to obtain an accurate structure can be used for optimization of heterogeneous materials. Statistical characterization of local microstructural fields of stress and strain is one of the instruments for estimation of damage nucleation and

understanding of nonlinear phenomena. The same numerical techniques and theoretical approaches allow to obtain fast numerical assessment for homogenized properties taking into account multipoint microscale interactions.

Some drawbacks of the statistical methods are that in most cases they are not customized for calculation of precise results. In order to receive more rigorous estimates, the introduced approaches can be used in combination with or within the other analytical and numerical methods of mechanics of composites. Besides, the microstructure of heterogeneous materials in the frameworks of statistical characterization is considered static, although some approaches can be generalized to dynamical problems.

Statistical descriptors and measures are important elements of microstructure sensitive design methods which facilitate inverse design for optimized performance of heterogeneous materials. Effectiveness of a particular statistical metric is connected with ability to capture the most influential features of microstructure. With extension of accessibility of computational capacities, statistical approaches allow to consider high-order interactions and open possibility to enhance the existing mechanical theories with more precise microstructural statistical instruments.

Abbreviations

2D: Two-dimensional; 3D: Three-dimensional; CNTs: Carbon Nanotubes; FIB: Focused Ion Beam; RSA: Random Sequential Algorithm; RVE: Representative Volume Element; SBVPs: Stochastic Boundary Value Problems; SEM: Scanning Electron Microscopy; TEM: Transmission Electron Microscopy

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