

# ULTRA-COMPOSITES: DESIGNING STOCHASTIC COMPOSITES WITH LARGE MICROSTRUCTURAL VARIABILITY

CATALIN R. PICU<sup>1,2</sup>, VINEET NEGI<sup>1</sup>, JACOB MERSON<sup>1</sup>

*Abstract.* Regular composite materials are made from a small number of constituent phases, usually two, which are arranged spatially such to maximize the stiffness and strength of the material. In this article, we review results related to defining a new class of stochastic composite materials with large microstructural variability both in terms of the composition and spatial distribution of constituents. Further, new data on Green functions in stochastic continua is presented. The Green functions in random composites are stochastic and their mean is proportional to  $\log(|\mathbf{x}|)$ , where  $\mathbf{x}$  is the distance to the point where the force acts, just like in the homogeneous case. However, the pre-logarithmic constant and hence the mean displacement field in the stochastic composite has larger values than the equivalent field in the homogeneous material having Young's modulus equal to the mean of the distribution function of moduli of the stochastic composite. The difference increases with increasing variance of the moduli distribution.

*Key words:* stochastic composites, homogenization, particulate composites.

## 1. INTRODUCTION

Composite materials are ubiquitous. Man-made composites are combinations of two or three materials, usually with quite different properties, which are arranged spatially to increase the overall strength, stiffness and toughness of the composite. Particulate composites are typically made by dispersing a reinforcing phase in a matrix. For example, rubber particles are dispersed in epoxy to increase its toughness. The mechanism leading to this effect is cavitation in the incompressible rubber particles under the applied far-field load. Another example is that of metal-based nanocomposites in which ceramic nanoparticles are dispersed in a metal (e.g. Ni) at very small volume fractions but large number density to act as obstacles for dislocation motion and therefore to allow controlling creep under high temperature loading conditions.

Composites in which the matrix is reinforced by fibers, either chopped or continuous, are even more common [1]. Examples are epoxy reinforced with glass

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<sup>1</sup> Rensselaer Polytechnic Institute, Department of Mechanical, Aerospace and Nuclear Engineering, Troy, NY 12180, USA

<sup>2</sup> "Politehnica" University of Bucharest, Department of Strength of Materials, Romania

or carbon fibers, used for various structural applications ranging from helicopter blades to bicycle frames. A less common example from the same category is that of polymers reinforced with fibers made from the same polymer. For instance, polypropylene can be reinforced with polypropylene fibers which have much higher stiffness and strength than the bulk material. The enhanced mechanical properties of the fibers are due to the preferential alignment of polymeric chains during fiber drawing. Since the fibers and matrix are chemically identical, the interface between them is perfectly bonded and load transfer between matrix and fiber is optimal.

Most biological materials are composites. Wood has a layered structure on the millimeter scale and is a cellular cellulosic material on smaller scales. Bone is a composite of (soft) collagen and (hard) hydroxyapatite exquisitely structured on all scales from the nanoscale to the scale of the skeleton [2]. The cornea or the human eye is made from layers of collagen fibers which are perfectly aligned parallel to each other in every given layer [3]. Collagen is oriented differently in different layers. This structure provides stiffness and strength while insuring transparency.

It is interesting to observe that biological and man-made composites are different in fundamental ways. One such difference is the fact that most natural composites are structured on multiple scales. Another difference is that nature may use multiple constituents to construct a composite, while artificial composites are made from a small number of constituents – usually just a matrix and a reinforcing phase. Interestingly, biological composites are quite insensitive to variability, i.e. their properties do not change much if the properties of their constituents fluctuate. In fact, it may be thought that such fluctuations are actually allowed in order to improve the overall composite behavior and to render it less sensitive to the accumulation of microstructural damage. Furthermore, biological composites (e.g. bone) are continuously repaired and renewed. This removes the damage, but increases the structural variability of the material.

The distribution of the reinforcing phase in man-made composites is either random or regular. Regular microstructures are only a design concept since variability introduced during manufacturing unavoidably leads to some degree of randomness of the structure even in the most controlled cases. For example, in woven composites fiber bundles do not contain exactly the same number of fibers and the weaves are not identical down to the micrometer scale.

Composite design usually does not account for variability. In fact, in composites with periodic microstructure, such as all woven pre-pregs, variability is undesirable and is considered to lead to premature damage nucleation and failure. Therefore, standard composite design and manufacturing aims to minimize structural variability such to, presumably, maximize macroscopic properties. In manufacturing of composites with random distribution of reinforcements, it is usually sought to create truly random microstructures and any clustering is considered undesirable. For example, poor dispersion and/or distribution of fillers is considered the key reason for poor performance of nanocomposite materials [4]. Few example of the

opposite trend exist in which clustering and/or the formation of a percolated structure of fillers is thought to be desirable. Rubber used in automobile tires is a molecular network reinforced with carbon black (graphitic) nanoparticles. In order to obtain good toughness and wear resistance, fillers should form fractal clusters which percolate through the network [5].

This discussion, and in particular the comparison between biological and artificial composites, indicates the need to reconsider the role of structural and compositional variability in the design of such materials. Clearly, accounting for variability greatly broadens the design space and it is not immediately clear whether significant property enhancement may or may not be obtained in regions of this space which have not been explored to date. This consideration motivated research on the effect of stochasticity on the overall material properties [6–10,11]. The objective of the present article is to review some of these results and to add new data on the dependence of the Green functions on material composition in a stochastic composite. The importance of this new result is related to the classical use of Green functions in linear elasticity to construct solutions for various boundary value problems.

Before proceeding with this discussion it should be observed that the current use of a small number of components in man-made composites is mostly conditioned technologically. It is easy to mix two constituents, but considerably harder to work with a large number of materials. In addition, without a theoretical basis, it is not immediately obvious that such complex compositions offer functional benefits. Recent technological advances, in particular the large growth of additive manufacturing, make now possible going beyond the traditional technological limits. These new techniques have to be guided by relevant theoretical developments. This provides motivation for the type of studies discussed in this article.

## 2. PROBLEM SET-UP

Let us consider a composite domain  $\Omega$ , of boundary  $\Gamma$ , over which a boundary value problem is defined. In general, we are interested in the effective properties of the composite, so the boundary value problem is defined such to allow for the identification of specific “composite properties.” For example, if one is interested in the effective stiffness,  $E_e$ , the boundary conditions applied on  $\Gamma$  would represent a uniaxial tension test.

Stochastic microstructures on  $\Omega$  can be generated by considering that one (or multiple) of the constituent properties, say Young’s modulus, are defined as functions of position and of a stochastic variable,  $\xi$ , i.e.  $E(\mathbf{x},\xi)$ . The associated distribution function of the fluctuating property,  $p(E)$ , can be specified in terms of a set of parameters such as a finite number of its moments. Let us denote these moments as  $m_E$ . In the case of a standard two-phase composites,  $p(E)$  is composed from two delta functions located at the values of Young’s modulus of phases A and B, respectively. In general, the distribution  $p(E)$  may have any functional form.

This entails, of course, that the composite is made from a combination of a range of materials or that the entire domain  $\Omega$  has graded composition.

Using the concept of a composite made from distinct inclusions (as opposed to that of a composite with graded properties), it is apparent that the size and shape of inclusions may be also considered stochastic variables and likewise may be defined by distribution functions described by a finite set of moments,  $m_S$ . Particles may be randomly distributed in the matrix, or distributed in a spatially correlated way. This property can be described by using the two-point autocorrelation function  $ACF(\mathbf{y}) = \langle E(\mathbf{x} + \mathbf{y})E(\mathbf{x}) \rangle_{\mathbf{x}, \xi}$ , where the average  $\langle \rangle$  is taken over multiple origins and replicas,  $\xi$ . Multipoint correlation functions may be also used for this purpose.  $ACF$  includes the information about the size and shape of inclusions in an average sense. If vectors  $\mathbf{x}$  and  $\mathbf{y}$  are sampled on a scale comparable with or larger than the inclusion size,  $ACF$  represents only the spatial distribution of inclusions and contains the information about the particle size and shape only in an average sense. For a random particulate composite, the  $ACF$  is a delta function. Most real microstructures are not perfectly random and have some degree of clustering. Then, the  $ACF$  function has a finite range and may be exponential or power law. The characteristic correlation length  $\lambda$  associated with  $ACF$  defines the mean cluster size. Such internal length scale can be defined in the case of exponential spatial correlations, but not in the case of power law correlated fields.

Let us collect all variables defining these distributions and correlation functions into a vector,  $\mathbf{v} = \{m_E, m_S, \lambda\}$ . These variables define the space of stochastic variables over which composite properties can be explored and optimized.

### 3. BRIEF BACKGROUND ON HOMOGENIZATION

Finding the effective properties of a composite, such as the effective elastic-plastic behavior or effective moduli, is the subject of homogenization theory. Reviews on the homogenization of random composites are presented in [12, 13, 14]. Remarkable results have been obtained regarding the bounds of the elastic moduli of such composites. These expressions are generally given in terms of the volume fraction of the constituents. The closest bounds for the bulk and shear moduli which take into account only the volume fraction have been derived by Hashin and Shtrikman [15]. The bounds apply equally to two and multi-phase composites.

A family of higher order bounds, that take into account statistical measures of the microstructure geometry, have been proposed more recently with the purpose of reducing the separation between the upper and lower bounds [e.g. 16, 17–20, 21]. The  $n$ -point bounds are written in terms of  $n$ -point microstructural correlation functions which define the probability that  $n$  points with specified relative positions are all located in a certain phase of the composite. A review of the higher order

bounds and the geometric parameters required for their evaluation is provided in [13]. As these formulations account for more details of the microstructure geometry and composition, the bounds become closer and hence are more useful in design.

Except in few specific periodic cases in which the filling fraction is small, the exact values of the homogenized material parameters (e.g. linear elastic or tangent moduli) cannot be found analytically and a numerical procedure is required. In the case of random composites several methods can be used to find the homogenized behavior. The most basic one is based on Monte Carlo sampling of the phase space of the composite structure, i.e. constructing replicas of the composite microstructure by sampling the relevant distribution functions, followed by numerical homogenization of each such replica. The statistics of the resulting fields are obtained in an approximated way by considering a sufficiently large number of replicas, or equivalently by sufficient sampling of the composite configuration space. This method has been used to obtain all results presented in this article [6, 11].

Another method that produces the first two moments of the distribution function of the solution fields is the Stochastic Finite Element method (SFEM) [22]. In this method the classical finite element formulation is used to write the balance equations in the weak form and in terms of the (unknown) displacement field,  $\mathbf{u}$ . The constitutive behavior of the composite continuum is expressed in terms of a deterministic position variable,  $\mathbf{x}$ , and a stochastic variable,  $\xi$ . Therefore, the solution,  $\mathbf{u}$ , is also a function of  $\mathbf{x}$ , and  $\xi$ . The constitutive parameters and the solution are written as products of deterministic functions of  $\mathbf{x}$  and stochastic functions of  $\xi$ . The deterministic functions are expressed in terms of the shape functions used in the finite element representation. The stochastic functions are expanded in a chaos polynomials series. The chaos polynomials (Hermite polynomials of Gaussian variable  $\xi$ ) can be used to approximate any arbitrary stochastic process of finite second moment. Then, finding the solution amounts to finding the coefficients of this expansion. The constitutive parameters are also expanded in a series which can be of the same type, i.e. a polynomial chaos series or of Karhunen-Loeve (KL) type [23, 24]. The KL expansion allows accounting for spatial correlations of the respective field and should be used in all cases in which the material parameters are spatially correlated. If the material parameters field is uncorrelated, using the KL expansion is precluded by the requirement to consider an infinite number of terms in the series in order to properly capture the delta function correlation of the respective process. With these two stochastic fields expressed as series expansions, the stochastic weak form is transformed (by pre-averaging over all terms that contain Gaussian stochastic variables) in a deterministic form with unknowns the coefficients of the polynomial chaos expansion of  $\mathbf{u}$ . The solution leads to the first two moments of  $\mathbf{u}$  at all points of the domain. This method is much more efficient than the Monte Carlo method because a single solve produces the entire statistical information about the fields. The method has its own limitations, mainly emerging from the approximations made when truncating the KL and chaos polynomials expansions.

Before closing this short overview, let us note that while finding the homogenized properties of the composite amounts to solving for the mean field, predicting the fracture and fatigue behavior of the material is more challenging as these aspects depend on the maxima of field fluctuations which are more difficult to capture accurately with the above methods.

## 4. RESULTS

This section presents results on the elastic-plastic behavior of two types of “ultra-composites”. Composites of the first type are made from two phases - matrix and inclusions – with inclusions being distributed either randomly or in a spatially correlated way. Composites of the second type are made from multiple phases of properties sampled from distribution functions. We are interested primarily in the effect of variability on the effective material behavior of the composite, in particular on the effective elastic modulus, yield stress and strain hardening. To isolate this effect, the mean of the distribution functions of local material properties is kept constant and the coefficient of variation of the distribution is kept as a parameter.

### 4.1. EFFECT OF SPATIAL CORRELATIONS

The results reviewed in this section have been published in [6]. Consider composites of the first type described above, i.e. they are composed from two constituents (matrix and inclusions), all inclusions have same size, shape and constitutive parameters, but the spatial distribution of inclusions is either random or spatially correlated. The matrix constitutive behavior is bilinear, with Young’s modulus  $E_1$  and constant tangent stiffness in the plastic range equal to  $E_1/10$ . Inclusions are linear elastic and have Young’s modulus six times larger than that of the matrix  $E_2/E_1 = 6$ . For simplicity, but without limiting generality, the matrix and inclusions are considered to have the same Poisson ratio, equal to 0.3. Inclusions are distributed in an uncorrelated (random) or in a correlated way, with the spatial correlation function  $ACF(r)$  being exponential.

Figure 1 shows two realizations of the uncorrelated (a) and correlated (b) filler distributions for composites with filling fraction  $f = 0.13$ . With the inclusion size denoted by  $d$ , the model size is more than two order of magnitude larger than  $d$ , i.e.  $L = 243d$ . The distribution of fillers in Fig.1(b) is exponential and the exponential function has a characteristic length of  $4d$ , i.e.  $ACF(r) \sim \exp\left(-\frac{r}{4d}\right)$ .

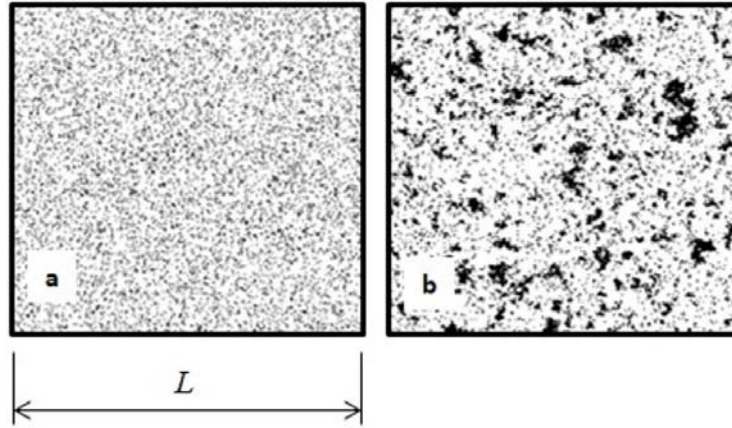


Fig. 1 – Two types of composite microstructures studied:  
 a) random distribution of inclusions; b) microstructure with an exponential correlation of inclusion positions having the same volume fraction and filler size  $d$  as that in a).

The computed effective stress-strain curve of the composite is bilinear. This feature is inherited from the fact that the matrix is bilinear. Let us denote the effective composite stiffness by  $E_e$  and the strain-independent strain hardening (slope of the stress-strain curve of the composite beyond the yield point) by  $E_p$ . Then, the homogenized behavior can be characterized using  $E_e/E_1$  and  $E_p/E_1$ , where  $E_1$  is the elastic modulus of the matrix. Figure 2 shows the variation of these quantities with the volume fraction of fillers,  $f$ . At least 50 realizations are used for each set of conditions. Since the composite is stochastic and models of finite size are considered, the values of these parameters corresponding to multiple realizations form a distribution. Figure 2 reports the mean of this distribution. The error bars indicating the standard deviation are smaller than the size of the symbols.

The continuous line in Fig.2 corresponds to random composites similar to that shown in Fig. 1a. The dashed line and cross symbols correspond to composites with spatial correlations having microstructure similar to that in Fig.1b. The two thick orange lines in Fig. 2a represent the 2D Hashin-Shtrikman bounds. The bounds are functions of the filling fraction  $f$  exclusively. It is seen that spatial correlations lead to modest increases in both parameters shown in Fig.2. The difference between the random and correlated cases is visible and larger than the sum of the standard deviations of the distribution functions corresponding to the two cases, but remains rather small. It increases with increasing filling fraction. In [6] it was shown that the weak stiffening effect observed here (Fig. 2a) is also reproduced by the higher order bounds [13] that take into account, beyond  $f$ , the presence of correlations. The bounds for the microstructure with correlations are both shifted up relative to the same bounds for the random microstructure. The effect is associated with the stronger interaction of inclusions in the case of spatially correlated filler distributions.

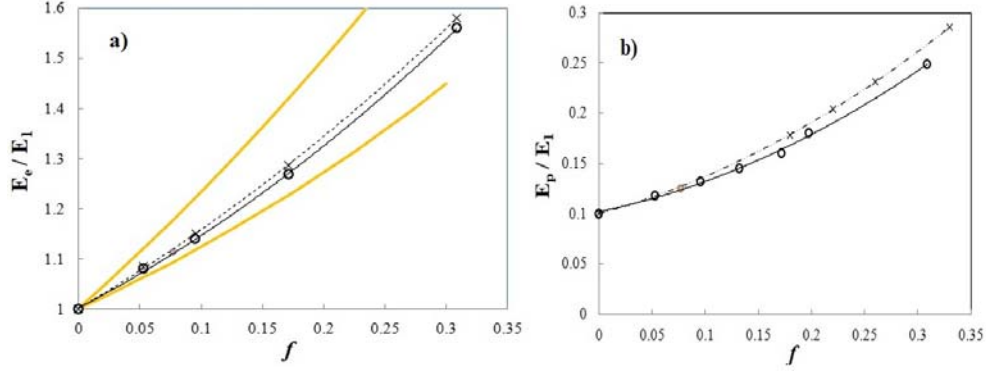


Fig. 2 – Variation of: (a) the mean elastic modulus ( $E_e/E_1$ ); (b) mean strain hardening rate ( $E_p/E_1$ ) with the volume fraction, for microstructures with randomly distributed inclusions (open symbols and continuous thin line) and microstructures with correlated distribution of inclusions (crosses and dashed line). The two thick orange lines in (a) correspond to the Hashin-Shtrikman bounds.

#### 4.2. EFFECT OF COMPOSITIONAL FLUCTUATIONS

We discuss next the effect of variability in the material parameters of the phases forming the composite. This problem is discussed in detail in [11]. Here we review only one of the three types of composites studied in this reference. Specifically, we consider composites with uncorrelated distribution of fillers (Fig. 1a). The matrix is homogeneous and elastic-plastic with a bilinear constitutive equation characterized by moduli  $E_1$  and  $10^{-2}E_1$  for the elastic and plastic branches of the constitutive law, respectively. The yield stress of the matrix is  $\sigma_{y1} = 10^{-2}E_1$ . Inclusions are linear elastic, of modulus  $E_2$  sampled from a log-normal distribution function,  $p(E_2)$ . The mean of this distribution is kept constant at  $\bar{E}_2 = 10E_1$ , while its variance is a parameter. The second moment is represented by the coefficient of variation  $c_E$  (ratio of the standard deviation to the mean) of  $p(E_2)$ . All components have the same Poisson ratio,  $\nu = 0.3$ , and plane strain conditions are considered throughout. Hence, this study outlines the effect on the overall composite mechanical behavior of allowing the filler properties to vary from filler to filler.

The effective behavior of the filled composite is approximately bilinear and hence it is characterized here using the two mean tangent moduli of the elastic and plastic parts,  $E_e/E_1$  and  $E_p/E_1$ , as well as the effective mean yield stress. Figure 3 shows the variation of  $E_e/E_1$  and  $E_p/E_1$  with  $c_E$ . These are normalized by the mean of the distribution of the corresponding quantities for cases with constant inclusion stiffness,  $c_E = 0$ , such that the plots show the effect of the variability in filler



elastic constants. The bars in the figure represent the standard deviation of the probability distribution functions  $p(E_e/E_1)$  and  $p(E_p/E_1)$  for  $c_E \neq 0$  estimated with 40 replicas for each case. It is seen that keeping the mean of the distribution function of filler moduli constant and increasing its variance, and hence  $c_E$ , leads to elastic softening, while variability in filler properties has no discernable effect on the effective strain hardening of the composite. The effective yield stress is also rather insensitive to  $c_E$  for this type of composites. The effect of filler properties variability on the elastic modulus of the composite increases with increasing the volume fraction,  $f$ . This elastic softening effect was also observed for linear elastic heterogeneous materials with fluctuating local elastic moduli in [7, 10].

It is rather straightforward to evaluate the Hashin-Shtrikman bounds for the elastic moduli using the original results [15] for multiphase composites. It is observed that, for the matrix-filler contrast considered in this study, the upper bound is essentially insensitive to  $c_E$  for the entire range  $0 < c_E < 3$  and for all  $0 < f < 1$ . The lower bound decreases as  $c_E$  increases. As also observed numerically for specific structures (Fig. 3), the dependence becomes more pronounced as  $f$  increases.

The elastic softening effect shown in Fig. 3a was also observed in networks of fibers in which each fiber has a different elastic modulus [8]. In [8] it is shown analytically that softening should always occur in both discrete structures such as fiber networks, and in continua of the type discussed here and in [6, 7, 10], and that the effect becomes more pronounced as the filling fraction,  $f$ , increases.

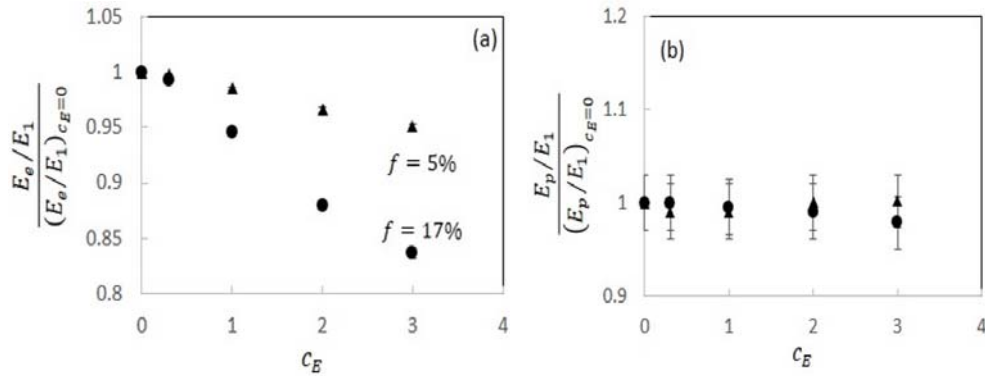


Fig. 3 – Variation of: a) the normalized mean elastic modulus ( $E_e/E_1$ );

b) normalized mean strain hardening ( $E_p/E_1$ ) with the coefficient of variation of the distribution function of inclusion moduli,  $c_E$ , for two volume fractions,  $f = 5\%$  (triangles) and  $f = 17\%$  (circles).

The standard deviation of the respective quantities is shown, but is smaller than the size of the symbols. The vertical axes are normalized with the value of the respective variable at  $c_E = 0$ .

Several other cases of composites with random spatial distribution of inclusions and with fluctuating filler and matrix properties are discussed in [6]. It is of interest to review here the general result of this study. It has been observed that:

(i) If either the matrix or inclusions have elastic moduli that fluctuate spatially (are selected from a distribution), the effective elastic modulus of the composite is smaller than that of the composites with no fluctuations and modulus equal to the mean of the respective distribution. In other words, at constant mean stiffness of constituents, fluctuations lead to overall softening. The effect is more pronounced as the variance of the distribution characterizing these fluctuations increases.

(ii) If the yield stress of the matrix is spatially variable and takes values from a distribution of given mean, the effective yield stress of the composite is smaller than that of the composite with no fluctuations and matrix yield stress equal to the mean of the distribution. Such plastic softening has been observed before in [9, 25].

(iii) If the strain hardening of the matrix fluctuates spatially, the effective strain hardening of the composite also decreases as the magnitude of these fluctuations increases. Hence, it can be generally stated that allowing a certain material property to have spatial fluctuations leads to a reduction of the corresponding effective property of the composite. This trend is opposite to that induced by the spatial correlations of filler distribution discussed in section 4.1.

### 4.3. GREEN FUNCTIONS FOR STOCHASTIC COMPOSITES

In this section we discuss the functional form of the effective Green functions for a 2D stochastic composite. Consider a two-dimensional composite material in which Young's modulus is a function of position,  $\mathbf{x}$ , and a stochastic variable,  $\xi$ , i.e.  $E(\mathbf{x}, \xi)$ . The stochastic field  $E(\mathbf{x}, \xi)$  may be spatially correlated or not. A unit force,  $\mathbf{F}$ , is applied at a point of this composite and the respective point is taken as the origin of the coordinate system (Fig. 4). The analysis seeks to determine the deformation field produced by the unit point force. Without loss of generality (due to the statistically isotropy of the distribution of heterogeneity), we consider the force to act along the  $x_1$  axis of the coordinate system in Fig.4. The displacement field,  $\mathbf{u}$ , is referred to the same coordinate system.

The problem is solved using a Monte Carlo method (section 2). To this end, the domain is discretized in  $100 \times 100$  cells of size  $\lambda$ , each cell being homogeneous and isotropic. The color in Fig.4 indicates the local Young's modulus, which is selected from a distribution of mean  $\bar{E}$  and of coefficient of variation  $c_E$ . The Poisson ratio is identical in all cells,  $\nu = 0.3$ . A point force of magnitude  $F = 1$  is placed in the center of this domain and the outer boundary is fixed. For each value of  $c_E$ , a large number of realizations (one hundred) are produced, each is subjected to the same deterministic boundary conditions and

loading and is solved using a finite element solver (Abaqus). The resulting displacement field is stochastic,  $\mathbf{u}(\mathbf{x}, \xi)$ . We are interested in the mean and the coefficient of variation of this field,  $\bar{\mathbf{u}}(\mathbf{x})$  and  $\mathbf{c}(\mathbf{x})$ . In particular, we want to compare  $\bar{\mathbf{u}}(\mathbf{x})$  with the displacement field produced by a unit point force loading a homogeneous continuum of Young's modulus equal to the mean of the distribution function of moduli in the stochastic composite problem,  $\bar{E}$ , and  $\nu = 0.3$ .

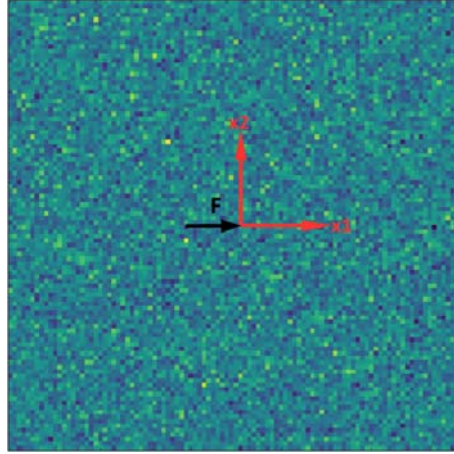


Fig. 4 – Two-dimensional domain used for the computation of Green functions for the stochastic composite. The color indicates the value of the local Young's modulus.

We note that this model is an approximation of the problem stated in the first paragraph of this sub-section. Specifically,  $E(\mathbf{x}, \xi)$  is piecewise constant and hence its correlation function is a step function equal to 1 for  $|\mathbf{x}| < \lambda$  and 0 for  $|\mathbf{x}| > \lambda$ . Taking  $\lambda \rightarrow 0$ , or conversely looking at the variation of  $\bar{\mathbf{u}}(\mathbf{x})$  and  $\mathbf{c}(\mathbf{x})$  for  $\infty \gg |\mathbf{x}| \gg \lambda$  corresponds to rendering the field uncorrelated.

The solution for the point force problem has been derived early in the history of continuum mechanics. Kelvin derived the solution for the 3D linear isotropic case, while Boussinesq [26] and Cerruti [27] derived the solution for the infinite half space loaded by a point force acting on its boundary. The similar solution for the point force acting at a point within a half space was derived by Mindlin [28]. In 2D, the displacements vary logarithmically with the distance from the point force:  $\mathbf{u} = a\mathbf{F} \log(|\mathbf{x}|) + \mathbf{b}(\mathbf{x})$ , where pre-logarithmic coefficient  $a$  is a function of the elastic constants. Let us take as reference the case of a homogeneous material with modulus  $\bar{E}$  and write the solution for this case as  $\mathbf{u} = a(\bar{E})\mathbf{F} \log(|\mathbf{x}|) + \mathbf{b}(\mathbf{x})$ , where we make explicit that the pre-logarithmic coefficient depends on  $\bar{E}$ . We observed

from the numerical solution that the functional form of the singular term of the displacement field in the stochastic composite remains unchanged:  $\mathbf{u} = a' \mathbf{F} \log(|\mathbf{x}|) + \mathbf{b}'(\mathbf{x})$ , where coefficient  $a'$  is now a function of  $\bar{E}$ ,  $c_E$  and possibly of the higher moments of the distribution function of  $E$ .

The continuous curve in Fig. 5 shows the dependence of the difference between the coefficients corresponding to the stochastic and homogeneous cases *versus* the coefficient of variation of the distribution of  $E$ , i.e.  $(a' - a(\bar{E})) / a(\bar{E})$  *versus*  $c_E$ . The coefficient  $a$  increases with  $c_E$ , indicating that the mean displacements are larger in the stochastic case compared with the homogeneous reference case. This result agrees with the overall softening of the composite associated with increasing  $c_E$  discussed in section 4.2 and shown quantitatively, although for a slightly different type of stochastic composite, in Fig. 3a).

This result may be obtained using a different method, based on SFEM. The stochastic Young's modulus field  $E(\mathbf{x}, \xi)$  was expanded in a KL expansion which was then used to formulate the weak form of the problem within the Galerkin finite element formulation. In the weak form, the problem is expressed as a linear system involving the stiffness matrix  $\mathbf{K}$ , nodal displacements and nodal forces. However in the stochastic finite element formulation, a series of stiffness matrices ( $\mathbf{K}^{(n)}$ ) corresponding to each term in the KL expansion appear. The linear system of equations becomes

$$\left( \mathbf{K}^0 - \sum_{n=1}^M \xi_n \mathbf{K}^{(n)} \right) \mathbf{u} = \mathbf{F}, \quad (1)$$

where  $\xi_n$  are uni-variate Gaussian identically distributed independent random variables (RV's), and  $\mathbf{K}^{(n)}$  are the stiffness matrices. This can be re-written:

$$\left( \mathbf{I} + \sum_{n=1}^M \xi_n \mathbf{Q}^{(n)} \right) \mathbf{u} = \mathbf{u}_0, \quad (2)$$

where  $\mathbf{Q} = (\mathbf{K}^0)^{-1} \mathbf{K}^{(n)}$  and  $\mathbf{u}_0 = (\mathbf{K}^0)^{-1} \mathbf{F}$ .

System (2) is formally inverted and the inverse matrix is written as a von Neumann series expansion, leading to an approximation for the unknown field  $\mathbf{u}$ :

$$\mathbf{u} = \left( \sum_{k=0}^{\infty} (-1)^k \left( \sum_{n=1}^M \xi_n \mathbf{Q}^{(n)} \right)^k \right) \mathbf{u}_0. \quad (3)$$

This expression is re-written symbolically as:

$$\mathbf{u}_{\text{mean}} = \mathbf{u}_0 + \mathbf{u}_2 + \mathbf{u}_4 + \dots + \mathbf{u}_k + \dots, \quad (4)$$

$$\text{where } \mathbf{u}_k = \sum_{i1=1}^M \sum_{i2=1}^M \dots \sum_{ik=1}^M \langle \xi_{i1} \xi_{i2} \dots \xi_{ik} \rangle \mathbf{Q}^{(i1)} \mathbf{Q}^{(i2)} \dots \mathbf{Q}^{(ik)} \mathbf{u}_0.$$

The magnitude of the  $\mathbf{u}_k$  terms was found to decrease rapidly with index  $k$ . Moreover, accurate calculation of higher  $\mathbf{u}_k$  terms is computationally very expensive. As a result, in this analysis only the  $\mathbf{u}_2$  term is considered. Note that the first term in the expansion,  $\mathbf{u}_0$ , corresponds to the homogeneous case. The mean displacement response so obtained was compared with the displacement field computed using the Monte-Carlo approach for various values of the coefficient of variation,  $c_E$ . The result is shown in Fig. 5 with dashed line. It is observed that the approximation of eq.(4) is very good up to fairly large values of  $c_E$ .

## 5. CONCLUSIONS

Several results pertaining to the elastic-plastic response of stochastic composites are discussed in this article. It is shown that in composites made from two materials, matrix and inclusions, the spatial distribution of inclusions influences the overall elastic modulus and strain hardening of the composite. Specifically, in presence of spatial correlations, or inclusion clustering, both the elastic modulus and the strain hardening of the composite increase relative to the respective parameters of the composite of same volume fraction and composition but without inclusions.

In composites with randomly distributed inclusions in which the stiffness of the inclusion material is allowed to vary from inclusion to inclusion we observe that the overall composite stiffness decreases with increasing the degree of variability. The strain hardening parameter is largely insensitive to these fluctuations.

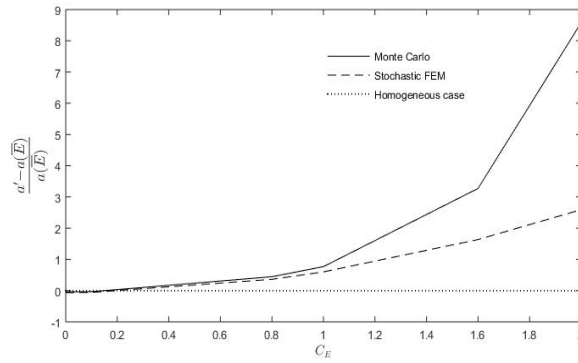


Fig. 5 – Variation of the difference between the pre-logarithmic coefficients  $a$  of the Green's function for the stochastic composite and for the homogeneous case, with the coefficient of variation of the stochastic field  $E(\mathbf{x}, \xi)$ . The continuous line corresponds to the Monte Carlo result, while the dashed line was obtained with the approximate stochastic finite element formulation described in text.

The displacement field associated with a point force acting on a 2D continuum – the Green functions – has been evaluated with two methods: a Monte Carlo technique and a procedure based on the stochastic finite element method. We observe again that the spatial variability of the composite elasticity leads, in average, to softening. Specifically, the displacement field in the stochastic composite has larger values than the corresponding field in the homogeneous material having modulus equal to the mean of the distribution of moduli in the stochastic composite case. The difference increases with increasing the degree of variability and can become important at values of  $c_E$  close to 1 or larger.

These results indicate that stochasticity in composition may lead to unexpected global composite behavior and these effects may be exploited in material optimization.

**Acknowledgements.** This work has been supported in part by the Romanian National Authority for Scientific Research, CNCS-UEFISCDI, project number PN-II-ID-PCE-2011-3-0120, under contract 293/2011.

*Received on March 15, 2016*

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