Composite Structures 113 (2014) 225–235

Contents lists available at ScienceDirect

Composite Structures

journal homepage: www.elsevier.com/locate/compstruct

Gaussian uncertainty in homogenization of rubber–carbon black nanocomposites

Marcin Kamiński *

Department of Structural Mechanics, Faculty of Civil Engineering, Architecture and Environmental Engineering, Technical University of Łódź, Al. Politechniki 6, 90-924 Łódź, Poland

ARTICLE INFO

Article history:
Available online 26 March 2014

Keywords:
Homogenization method
Probabilistic analysis
Weighted Least Squares Method
Particle reinforced composites
Finite Element Method
Representative Volume Element

ABSTRACT

The objective of this work is Gaussian uncertainty in material parameters of the rubber–carbon black particle reinforced nano-composite in the framework of determination of its effective elastic parameters. The analytical 3D micro-mechanical model is contrasted here with the Finite Element Method one realized with the use of the system ABAQUS® and its tetrahedral finite elements C3D4. They discretize cubic Representative Volume Element (RVE) with a centrally located spherical carbon black particle, whose deformation energies accumulated during the uniaxial and biaxial uniform tension are computed. The polynomial-based Response Function Method allows to determine analytical functions relating effective elasticity tensor with Young moduli and Poisson ratios of this composite original components via the Weighted Least Squares Method; this is done in the symbolic environment of MAPLE. These functions serve for the initial sensitivity analysis, where we detect Poisson ratio of the rubber matrix as the crucial design parameter and its Young modulus as having secondary importance, while particle Young modulus and Poisson ratio are totally irrelevant. Next, the most influential parameters are randomized according to the Gaussian distribution and are employed in the dual probabilistic scheme – by using the stochastic perturbation and, independently, semi-analytical techniques to determine up to the fourth order probabilistic characteristics of the effective tensor components. Deterministic and probabilistic hypersensitivity of the effective constitutive tensor, expected according to the rubber matrix presence, is confirmed in all computational experiments. Such a probabilistic energy-related FEM approach will allow for future applications of more advanced constitutive models for the carbon black nano-composites, for the RVEs of larger sizes – containing large agglomerations of these particles and for the imperfect interfaces simulation.

1. Introduction

Computational modeling and numerical solutions for solids and structures at the incompressibility limits are of the special importance in the area of elastomers and rubber engineering and usually bring some results qualitatively different from traditional structural materials applications [1,2]. Since the rubber has Young modulus thousand times smaller ($E = 4.0$ MPa) than the carbon reinforcements ($E = 10.0$ GPa) as well as polymeric matrices and Poisson ratio very close to 0.50, then even elastic behavior of the rubber-based composites and their design sensitivity is not trivial and may return some singularities. Homogenization analysis of the latters using both analytical expressions and numerical techniques is not free from these effects, especially in the context of an uncertainty in these elastic characteristics. Randomness in the nano-scale looks even more natural than in the macro, meso or micro-scales [3] according to the uncertain distribution of the particles, some dislocations of the molecules, their interactions and interphases formed in some specific temperatures. This randomness has been studied before in case of some effective constitutive analytical equations for the elastomers [4] as well as using computational Stochastic Finite Element Method (SFEM) approach [5] for the polymer-based matrix enriched with the rubber filling spherical particles.

Of course, the history of homogenization method is very long right now and obeys the works concerned on algebraic equations for the effective constitutive relations and their bounds [6,7], some asymptotic solutions in terms of micro-macro transition [8,9] with both lower and higher order [10] geometrical expansions. This area has been extensively developed by various probabilistic techniques applied together with classical Finite Element Method (FEM). One may recall an application of the Monte-Carlo strategy [11,12], stochastic spectral techniques [13], stochastic perturbation

* Tel./fax: +48 42 6313571.
E-mail address: Marcin.Kaminski@p.lodz.pl

http://dx.doi.org/10.1016/j.compstruct.2014.03.033
0263-8223/

method of different order and precision [14] or the semi-analytical approach [15]; some probabilistic estimates of an algebraic structure are also known [16]. Furthermore, an implementation of the Voronoi Cell Finite Element Method (VCFEM) serving for the RVEs with multiple fibers or particles is available also [17] together with deterministic homogenization of thermal stresses in specific composites [18].

An application of the generalized stochastic perturbation technique [19] that allows for relatively large random fluctuations of the given input random variable is explored in this work together with some concurrent semi-analytical probabilistic technique. The homogenization-based numerical analysis is conducted via calculation of a deformation energy during uniform extension of the RVE [20]. It was undertaken to verify an effect of the Young moduli uncertainty on the basic probabilistic characteristics of such a material, where spherical particles have this modulus about thousand times smaller than the surrounding polymeric matrix. Now, very similar computational methodology is engaged for a homogenization of the composite with carbon black particles [21] having Young modulus more than thousand times larger than the rubber matrix and, further, randomization procedure is applied to all material characteristics of the constituents. The main benefit is in probabilistic analysis of the homogenization procedure when one of the components approaches to the compressibility physical limit. It is interesting indeed how Gaussian randomness in the Poisson ratio of the rubber affects probabilistic characteristics of the effective tensor for various levels of an input coefficient of variation. Computational procedure is provided with the system ABAQUS, where the FEM test are all carried out and, further, with the use of the symbolic environment MAPLE, where the Weighted Least Squares Method (WLSM) approximation [22] and probabilistic calculus are done both. A deterministic comparison with analytical bounds available in the literature [6] shows a perfect agreement of these methods, while probabilistic moments of the homogenized tensor during randomization of the rubber Poisson ratio shows probabilistic analysis from totally different perspective.

2. Randomized homogenization method

Let us consider a bounded continuum $\Omega \subset \Re^3$ with no initial stresses and strains (cf. Fig. 1 including also its further computational FEM discretization) consisting of two disjoint elastic constituents. The elastic characteristics of $\Omega = \Omega_m \cup \Omega_p$ are treated as design random parameters and they result in random displacement field $u_i(\mathbf{x};\omega)$ and random stress tensor $\sigma_{ij}(\mathbf{x};\omega)$ satisfying a linear elasticity elliptic boundary value problem. The particle volume Ω_p is uniquely defined with its deterministic radius, while the matrix Ω_m surrounds it in the RVE with a perfect interface. Let us assume that there are non-empty subsets of external boundaries of the domain Ω (with the dimensions $2l_1 \times 2l_2 \times 2l_3$), namely $\partial\,\Omega_\sigma$ and $\partial\,\Omega_u$, where the additional boundary conditions are defined.

Contrary to the deterministic case study, now we solve the entire set of the boundary value problems with the same boundary conditions and with additionally modified given input random parameter $b \equiv b^{(\alpha)}$, $\alpha = 1,\ldots,n$. The set of solutions to the boundary differential equation systems describing static equilibrium around the mean value of this parameter indexed by α is determined.

$$\sigma_{ij}^{(\alpha)}(\mathbf{x}) = C_{ijkl}^{(\alpha)}(\mathbf{x})\varepsilon_{kl}^{(\alpha)}(\mathbf{x}), \tag{1}$$

$$\varepsilon_{ij}^{(\alpha)}(\mathbf{x}) = \frac{1}{2}\left(\frac{\partial u_i^{(\alpha)}(\mathbf{x})}{\partial x_j} + \frac{\partial u_j^{(\alpha)}(\mathbf{x})}{\partial x_i}\right), \tag{2}$$

$$\sigma_{ij,j}^{(\alpha)}(\mathbf{x}) = 0, \tag{3}$$

$$u_i^{(\alpha)}(\mathbf{x}) = \hat{u}_i(\mathbf{x}); \quad \mathbf{x} \in \partial\Omega_u, \tag{4}$$

$$\sigma_{ij}^{(\alpha)}(\mathbf{x})n_j = \tilde{t}_i; \quad \mathbf{x} \in \partial\Omega_\sigma, \tag{5}$$

The corresponding variational formulations indexed also by α are introduced to get the amount of strain energy stored in the RVE and this is done via application of the Finite Element Method. It yields [20]

$$\int_\Omega C_{ijkl}^{(\alpha)}\varepsilon_{ij}^{(\alpha)}\delta\varepsilon_{kl}^{(\alpha)}d\Omega = \int_{\partial\Omega_\sigma}\tilde{t}_i\delta u_i^{(\alpha)}d(\partial\Omega), \tag{6}$$

where the left hand side (LHS) of Eq. (6) corresponds to the elastic energy of this structure and its right hand side (RHS) is equivalent to the stress boundary conditions applied. A determination of the effective tensor uses the strain energy of the heterogeneous medium, i.e.

$$U^{(\alpha)} = \frac{1}{2}\int_\Omega C_{ijkl}^{(\alpha)}\varepsilon_{ij}^{(\alpha)}\varepsilon_{kl}^{(\alpha)}d\Omega. \tag{7}$$

The homogenized medium accumulates the same amount of elastic energy having effective elastic characteristics series $C_{ijkl}^{(eff)(\alpha)}$. A comparison of these two energies leads to

$$\frac{1}{2}\int_\Omega C_{ijkl}^{(\alpha)}\varepsilon_{ij}^{(\alpha)}\varepsilon_{kl}^{(\alpha)}d\Omega = \frac{1}{2}\int_\Omega C_{ijkl}^{(eff)(\alpha)}\varepsilon_{ij}^{h(\alpha)}\varepsilon_{kl}^{h(\alpha)}d\Omega. \tag{8}$$

where $\varepsilon_{ij}^{h(\alpha)}$ denotes the strain tensor associated with the homogenized equivalent medium. This relation may serve directly for a calculation of the homogenized elastic characteristics if only the strain tensor has unit value; we assume additionally that the geometrical dimensions of this RVE are equal in all directions (that means $l_1 = l_2 = l_3 = \delta$) to focus numerical experiments on an influence of the random fluctuations of material parameters of both components by only. The boundary conditions are specified as

$$\begin{aligned}\varepsilon_{ij}^{x_1}: &u_1(\delta,x_2,x_3) = \Delta_1, \quad u_2(x_1,\delta,x_3) = 0, \quad u_3(x_1,x_2,\delta) = 0,\\ &u_1(-\delta,x_2,x_3) = -\Delta_1, \quad u_2(x_1,-\delta,x_3) = 0, \quad u_3(x_1,x_2,-\delta) = 0,\end{aligned} \tag{9}$$

as well as

$$\begin{aligned}\varepsilon_{ij}^{x_2}: &u_1(\delta,x_2,x_3) = 0, \quad u_2(x_1,\delta,x_3) = \Delta_2, \quad u_3(x_1,x_2,\delta) = 0,\\ &u_1(-\delta,x_2,x_3) = 0, \quad u_2(x_1,-\delta,x_3) = -\Delta_2, \quad u_3(x_1,x_2,-\delta) = 0.\end{aligned} \tag{10}$$

due to the symmetry of the problem. One writes after the strain tensor definition

$$\varepsilon_{ij}^{x_1} = \frac{\delta_1}{\delta}, \quad \varepsilon_{ij}^{x_2} = \frac{\delta_2}{\delta}. \tag{11}$$

Therefore, we obtain the following system of linear algebraic equations describing effective characteristics as linear functions of the deformation energies of the RVE under uniform strains:

$$\begin{cases} \frac{1}{2}C_{1111}^{(eff)(\alpha)}(\varepsilon_{11}^{x_1})^2 = U_1^{(\alpha)} \\ \frac{1}{2}C_{2222}^{(eff)(\alpha)}(\varepsilon_{22}^{x_2})^2 = U_2^{(\alpha)} \\ \frac{1}{2}\left\{C_{1111}^{(eff)(\alpha)}(\varepsilon_{11}^{x_1})^2 + 2C_{1122}^{(eff)(\alpha)}\varepsilon_{11}^{x_1}\varepsilon_{22}^{x_2} + C_{2222}^{(eff)(\alpha)}(\varepsilon_{22}^{x_2})^2\right\} = U_{12}^{(\alpha)} \end{cases} \tag{12}$$

where the last RHS equals to

$$U_{12}^{(\alpha)} = U_1^{(\alpha)} + U_2^{(\alpha)} + \frac{1}{2}\int_\Omega \sigma_{ij}^{x_1(\alpha)}\varepsilon_{ij}^{x_2}d\Omega + \frac{1}{2}\int_\Omega \sigma_{ij}^{x_2(\alpha)}\varepsilon_{ij}^{x_1}d\Omega \tag{13}$$

in case of any combination of the boundary conditions imposed. Numerical solution to Eq. (6) proceeds after spatial discretization of the displacements fields series typical for the FEM using classical