Computational Materials Science 82 (2014) 483–496

Contents lists available at ScienceDirect

Computational Materials Science

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

Probabilistic homogenization of polymers filled with rubber particles

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ARTICLE INFO

Article history:
Received 3 July 2013
Received in revised form 23 August 2013
Accepted 4 October 2013
Available online 12 November 2013

Keywords:
Homogenization method
Probabilistic analysis
Weighted Least Squares Method
Particle filled polymers
Finite Element Method
Representative Volume Element

ABSTRACT

The main purpose of this work is computational simulation of the basic probabilistic characteristics of the homogenized tensor for polymer filled with rubber particles. The Representative Volume Element (RVE) of this composite contains a single spherical particle and composite components are treated as statistically homogeneous and isotropic uniquely defined by the Gaussian elastic moduli. Probabilistic approach is based upon the generalized stochastic perturbation technique allowing for large random dispersions for the input random variables and is implemented using the polynomial response functions recovered via the Least Squares Method. Homogenization technique consists in equating of deformation energies for the real composite and artificial isotropic material characterized by the effective elasticity tensor. The cell problem is solved using ABAQUS® by an application of uniform deformations on specific outer surfaces of the composite cell and using tetrahedral finite elements C3D4, while probabilistic part is carried out in the symbolic computations package MAPLE®. The main conclusion coming from the performed numerical analysis is almost perfect agreement of our mean values with the analytical calculations, dominating role of the Young modulus of the polymeric matrix and negligible role of the elastic modulus for rubber. We also notice that the Gaussian character is transferred from the first modulus on the homogenized tensor, while randomization of the rubber Young's modulus is negligible. The energy approach will allow for future applications of more realistic constitutive models of rubber-filled polymers as well as for the RVEs of larger size – containing an agglomeration of the rubber particles, for instance.

1. Introduction

There is a variety of homogenization techniques leading to a determination of the effective tensors characterizing the homogeneous medium equivalent to the original composite structure [1–3]. They are based on some analytical bounds or exact approximations based upon the deformation criteria or the deformation energy. Analogously, we have numerical approaches to the homogenization, where we usually solve the so-called cell problem on the Representative Volume Element (RVE) to predict the homogenized behavior of the entire composite structure. It is usually carried out using the Finite Element Method by the straightforward spatial discretization of this RVE and the relevant solution via the displacement or stress-based formulations. Different typical stress boundary conditions in-between the composite components and periodicity conditions on external edges of the RVE [4–6] can be applied in this case. Alternatively, constant deformations on the RVE's external boundaries are imposed and we assure their continuity at the interface [7] to calculate effective tensor's components. The first approach needs additional spatial averaging of the induced stresses fields, while the second allows for a direct computation of the homogenized tensor's components from the internal energy accumulated in the RVE as the result of the applied uniform deformation. Composites consisting of polymeric matrix and rubber particles are a very specific area of engineering, because the particles do not play the traditional role of reinforcement for the composite's specimen. Such composites are frequently called elastomers and a lot of research attention is focused on their analysis and development recently [8–10]; the main propose of rubber filled polymers is to improve the toughness of the material. Considering further described probabilistic methodology one needs to notice a large variety of the uncertainty sources in elastomeric structures [6].

The uncertainty in the composite's parameters [4,11,12] essentially does not change the homogenization methods in the sense that some probabilistic technique needs to be added to the deterministic apparatus to perform randomization of the model. Traditionally Monte-Carlo simulation in its various implementations can be used for this purpose to get the statistical estimators of the effective tensor. Alternatively, the direct integration technique can be applied, when the analytical approximations for this tensor

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http://dx.doi.org/10.1016/j.commatsci.2013.10.006

are taken into account as well as using the perturbation or the expansion-based methodologies. The stochastic perturbation technique is sufficient to carry out all computations here, because we study the Gaussian random field of Young's modulus with no spatial correlations inside the statistically homogeneous components. However, taking into account possible large random deviations of these parameters its generalized version is preferred, where any order of probabilistic Taylor expansion is admissible. Further, we choose the Response Function Method (RFM) to omit straightforward partial differentiation of all orders' equilibrium equations and we provide the response polynomials relating the internal energy of the composite with random parameters separately. The Direct Differentiation Method (DDM) applied frequently in lower order stochastic techniques is left here to develop the basic equations of the probabilistic homogenization technique. We employ directly in our study the deformation energy of polymer–rubber composite specimen and its probabilistic moments to calculate probabilistic characteristics of the effective elasticity tensor components assuming that the homogenized medium is also isotropic. It should be underlined that, according to the best knowledge of the authors, this homogenization method was never tested before in case of any uncertainty in the composite being homogenized. Basic deterministic series of calculations are performed thanks to the FEM system ABAQUS, where cubic RVE with centrally located spherical rubber particle is subjected to uniform deformations in the directions parallel to the external edges of this specimen. An isotropy assumption is well justified here by the cubic shape of the entire cell, spherical shape of the reinforcing particle and central location of the particle's center in this RVE. An assumption of the effective macro-isotropy may be relatively easy replaced in further numerical studies with the orthotropic homogenized constitutive equation, for instance, as for the fiber-reinforced structures or, alternatively, to model analogous RVE with the elliptic reinforcing particles (after significant uniaxial deformation, for instance). We compare first the effective tensor components with its other approximations [1] to validate ABAQUS model of the RVE itself, our homogenization theory and to analyze applicability of the simple algebraic formulas in further engineering calculations.

2. Homogenization method

Let us consider a statistically heterogeneous and bounded continuum $\Omega \subset \Re^3$ with no initial stresses and strains. Elastic properties and geometry of Ω (see Fig. 1) may be 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 linear elasticity elliptic boundary-value problem. Let us assume that there are non-empty subsets of external boundaries of the domain Ω, namely $\partial\Omega_\sigma$ and $\partial\Omega_u$, where the Dirichlet and von Neumann boundary conditions are defined.

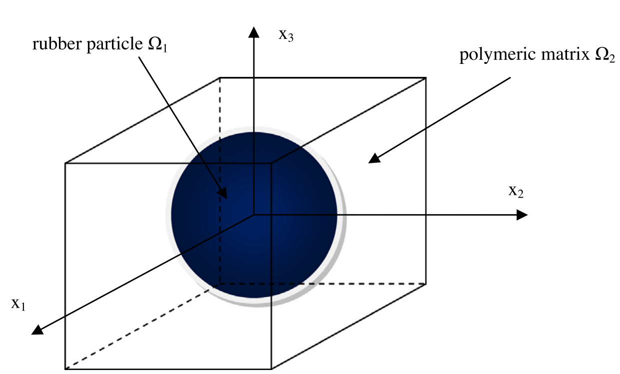

Fig. 1. Periodic two-component composite idealization.

Contrary to the deterministic case study and also random situation, where perturbation-based technique is used in its Direct Differentiation Method version, now we need to solve the whole set of the boundary value problems with the same boundary conditions and with additionally modified input parameter $b \equiv b^{(\alpha)}$, $\alpha = 1, \ldots, n$. We look for the set of solutions to the boundary-differential equation systems describing static equilibrium around the mean value of this parameter, so that

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

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

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

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

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

We follow variational formulation, also as the finite set of integral equations, to get an appropriate numerical solution for the strain energy in terms of the Finite Element Method. It yields

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

where the left hand side of Eq. (6) corresponds to elastic behavior of the structure and the RHS is equivalent to the stress boundary conditions applied. It needs to be mentioned that indexing with respect to the RFM should be added to the computational domain Ω as far as stochastic shape sensitivity is to be modeled; the corresponding extension to $\partial\Omega_u$, $\partial\Omega_\sigma$ and additional conditions may reflect an uncertainty in a structure external boundary.

Determination of the effective material tensor needs the strain energy of the heterogeneous medium

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

The homogenized medium is a linear and isotropic one, which accumulates the same amount of energy having effective elastic characteristics' series $C_{ijkl}^{(eff)(\alpha)}$, so that we compare this against the energy stored in the homogenized medium

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

where $\varepsilon_{ij}^{h(\alpha)}$ denotes the strain tensor adjacent to the homogenized equivalent medium.

Further, we set specific boundary conditions to the composite RVE to make this comparison, which correspond to the uniform expansion of this cube with the dimensions $2\delta \times 2\delta \times 2\delta$, i.e. [7]

$$\begin{aligned}\varepsilon_{ij}^{x_1}:\ & u_1(\delta,x_2,x_3)=\delta_1,\ u_2(x_1,\delta,x_3)=0,\ u_3(x_1,x_2,\delta)=0,\\ & u_1(-\delta,x_2,x_3)=-\delta_1,\ u_2(x_1,-\delta,x_3)=0,\ u_3(x_1,x_2,-\delta)=0,\end{aligned} \tag{9}$$

as well as

$$\begin{aligned}\varepsilon_{ij}^{x_2}:\ & u_1(\delta,x_2,x_3)=0,\ u_2(x_1,\delta,x_3)=\delta_2,\ u_3(x_1,x_2,\delta)=0,\\ & u_1(-\delta,x_2,x_3)=0,\ u_2(x_1,-\delta,x_3)=-\delta_2,\ u_3(x_1,x_2,-\delta)=0.\end{aligned} \tag{10}$$

According to the definition one writes

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