Computational Materials Science 96 (2015) 520–535

Contents lists available at ScienceDirect

Computational Materials Science

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

A unified framework for stochastic predictions of mechanical properties of polymeric nanocomposites

N. Vu-Bac [a], M. Silani [a,b], T. Lahmer [a], X. Zhuang [c,*], T. Rabczuk [a,*]

[a] Institute of Structural Mechanics, Bauhaus-Universität Weimar, Marienstr. 15, D-99423 Weimar, Germany
[b] Department of Mechanical Engineering, Isfahan University of Technology, Isfahan 84156-83111, Iran
[c] Department of Geotechnical Engineering, Tongji University, Shanghai 200092, China

ARTICLE INFO

Article history:
Received 7 March 2014
Accepted 11 April 2014
Available online 10 July 2014

Keywords:
Polymer clay nanocomposites (PCNs)
Computational modeling
Micromechanical modeling
Mechanical properties
Stochastic predictions

ABSTRACT

We propose a stochastic framework based on sensitivity analysis (SA) methods to quantify the key-input parameters influencing the Young's modulus of polymer (epoxy) clay nanocomposites (PCNs). The input parameters include the clay volume fraction, clay aspect ratio, clay curvature, clay stiffness and epoxy stiffness. All stochastic methods predict that the key parameters for the Young's modulus are the epoxy stiffness followed by the clay volume fraction. On the other hand, the clay aspect ratio, clay curvature and the clay stiffness have an insignificant effect on the Young's modulus of PCNs. Besides the results on the sensitivity of the input parameters, this work includes a comparative study of a series of stochastic methods to predict mechanical properties of PCNs with respect to their performance.

1. Introduction

Polymer (epoxy) clay nanocomposites (PCNs) have been studied extensively as a new generation of polymeric materials and received wide interests in the research community of material sciences and engineering due to their exceptional thermal and mechanical properties. The clay particle structure is either exfoliated or intercalated. For enhanced functional properties of nanocomposites at the same clay concentration the former is preferred [1]. An exfoliated PCN was developed by the Toyota group by synthesizing a nylon 6/clay nanocomposite [2,3]. They reported that the mechanical properties (tensile modulus and strength) of nylon 6/clay nanocomposites were significantly improved even at low clay concentrations. It is believed that the enhanced properties of PCNs strongly depend on the clay aspect ratio and the mechanical properties of the nanoclay. After pioneering success in the nylon 6/clay system, the nanocomposite technology has been extended to other polymeric systems, including elastomers [4,5] and epoxies [6,7]. It was shown that enhancement of the thermal/mechanical properties in the polymeric nanocomposites is sensitive to the particular polymer chosen.

Analytical and numerical predictions of the overall composite stiffness have been extensively studied [8]. A comprehensive review of micromechanical models fiber-reinforced polymers, such as Halpin–Tsai [9] and Mori and Tanaka [10], was outlined by Tucker and Liang [11]. Analytical studies [12,13] suggest that the high clay stiffness, high aspect ratio and volume fraction are the key parameters governing the stiffness of clay nanocomposite. Sheng et al. [14] proposed a multiscale finite element method (FEM), accounting for the hierarchical morphology of the PCNs, to predict the macroscopic properties of PCNs by using the so-called 'effective particles' in which the model parameters such as the particle volume fraction, particle aspect ratio and orientation and particle/matrix property ratios were taken into consideration. Scocchi et al. [15] developed a bottom-up approach to study the relative macroscopic properties of PCNs. Fermeglia and Pricl [16] proposed a hierarchical procedure for bridging the atomistic and macroscopic results through mesoscopic simulations.

As the mechanical properties of a certain polymer clay nanocomposite system are affected by multiple uncertain parameters such as the clay volume fraction, clay aspect ratio, clay curvature, clay stiffness and epoxy stiffness, a comprehensive study of the effects of such above-mentioned parameters on the mechanical behavior of PCNs is required. So far, the stochastic effects of these parameters on the mechanical properties have not been quantified.

In this article, we conduct a comprehensive global sensitivity analysis (SA) based on a stochastic modeling of PCNs. The finite element (FE) analysis is used to predict the stiffness of the fully

* Corresponding authors. Address: Institute of Structural Mechanics, Bauhaus-Universität Weimar, Marienstr. 15, D-99423 Weimar, Germany (T. Rabczuk). Tel.: +49 (0)3643 58 4511.

E-mail addresses: xiaoyingzhuang@tongji.edu.cn (X. Zhuang), timon.rabczuk@uni-weimar.de (T. Rabczuk).

http://dx.doi.org/10.1016/j.commatsci.2014.04.066
0927-0256/

exfoliated PCNs within the framework of the stochastic modeling. Based on a micromechanical approach, a homogenized Young's modulus is computed at the meso-scale. Subsequently, different SA methods are employed to quantify the influence of the input parameters on the PCN Young's modulus. A bootstrap technique has been performed in order to assess the robustness of different SA methods.

The article is outlined as follows. In the next section, we briefly describe the FE model and homogenization techniques. Section 3 presents the complete statistical distribution for each input parameter. Different SA methods are described in Section 4. The surrogate model is presented in Section 5. The coefficients of surrogate model, sensitivity indices and bootstrap confidence intervals for the sensitivity indices will be detailed in Section 6 before the article ends with a discussion on the numerical results and the concluding remarks.

2. Model for PCN

We focus on PCNs that are built using the slurry compounding process. The epoxy resin is diglycidyl ether of bisphenol A (D.E.R.TM 332) and the curing agent is diethyltoluenediamine (Ethacur 100-LC, Albemarle). The pristine clay is sodium montmorillonite (MMT) [17–20].

Models of various periodic representative volume elements (RVEs) of the epoxy matrix filled with exfoliated clay platelets (particles) that are randomly oriented and dispersed are constructed. Fig. 1 shows a detailed view of the FE mesh inside the RVE of the PCN. The micromechanical FE model was implemented in ABAQUS as detailed in [21].

In this work, we assume a simplified case of *perfectly bonded* particles in an isotropic matrix.

2.1. Homogenization

2.1.1. RVE definition

The RVE approach uses the solution of the homogenization for an original heterogeneous medium to obtain an equivalent homogeneous one that is able to substitute the heterogeneous (polymer/platelet) material. Hence, the RVE size should be defined so that the RVE contains enough information to reasonably simulate an infinite medium.

Since the RVE is randomly generated, the ensemble of many realizations (RVEs) needs to be generated to create a good statistical representation of the PCN. In other words, we replace a large RVE that reasonably simulates an infinite medium by a statistical ensemble of RVEs for sake of computational efficiency. The number of realizations is determined such that the entire ensemble contains the same amount of information as a large one.

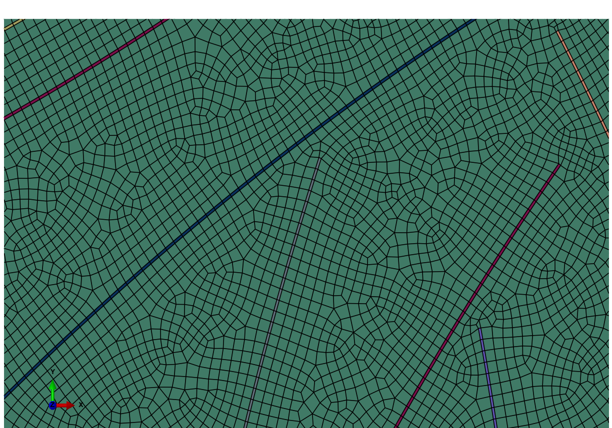

Fig. 1. A detailed view of the mesh.

The ensemble average is given by Spencer and Sweeney [22]

$$\langle R \rangle = \frac{1}{M}\sum_{k=1}^{M} R^{(k)}, \tag{1}$$

where $R^{(k)}$ is a response measured in the k-th RVE ($k = 1, 2, \ldots, M$); M is the number of RVE realizations in the ensemble. In order to estimate convergence of the statistical ensemble, the saturation criterion was used

$$\left| \frac{\langle R^{(k+1)} \rangle - \langle R^{(k)} \rangle}{\langle R^{(k)} \rangle} \right| < Tol = 1\%, \tag{2}$$

where $R^{(k)}$ denotes an expected value of k realisations, and $R^{(k+1)}$ is averaged over $k+1$ realizations. Eq. (2) allows to recognize whether the result reaches a reasonable accuracy.

2.1.2. Boundary conditions

Usually, three types of BCs can be applied to the RVE, namely linear displacement, uniform traction and mixed type BCs (which includes periodic BCs). For linear elasticity, there is an ordering relationship among the results which are obtained from those three kinds of BCs [23]. This ordering relationship finally motivates the use of periodic BCs in case of periodic structures [24,25].

In practical applications, periodic BCs are also used in case of non-periodic microstructures. It has been shown in numerous numerical studies that periodic BCs yield a fast convergence rate of the effective properties with respect to (w.r.t.) the size of the RVE [26–28]. The periodic BCs are imposed on the RVE as shown in Fig. 2 by applying a displacement on a rigid reference node that is kinematically coupled with the RVE edges in the axial direction so that the displacements of all boundary nodes are identical to those of the equivalent node on the opposite edge.

2.1.3. RVE generation algorithm

The characteristics of the clay particles (their aspect ratio (length), curvature (radius) and stiffness) are randomly generated according to predefined sets of statistical distributions [22], e.g. all clay particles have the same aspect ratio, curvature radius, stiffness, and the clay particles are randomly dispersed in the epoxy matrix. The resulting clay particle configurations are ideal for estimating the influence of each particle-characteristic on the mechanical response. A scheme of the RVE generation algorithm is shown in Fig. 3.

Firstly, the control parameters including the number of RVEs for each value of clay concentrations (volume fraction), the size of the RVE, the clay length, curvature and orientation distribution are set.

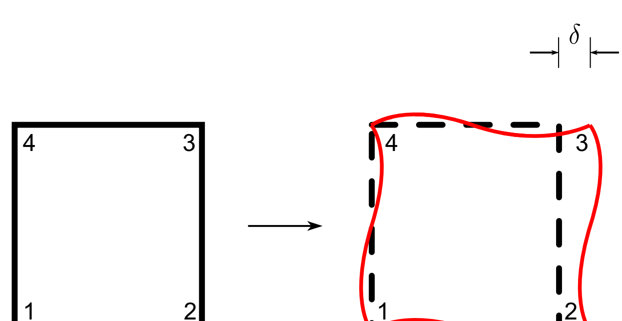

Fig. 2. Boundary conditions imposed on the RVE: (left) an undeformed RVE and (right) a deformed RVE.