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Modeling of the effect of particles size, particles distribution and particles number on mechanical properties of polymer-clay nano-composites: Numerical homogenization versus experimental results

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ABSTRACT

The main goal of this paper is to predict the elastic modulus of partially intercalated and exfoliated polymer-clay nano-composites using numerical homogenization techniques based on the finite element method. The representative volume element was employed here to capture nano-composites microstructure, where both intercalated exfoliated and clay platelets coexisted together. The effective macroscopic properties of the studied microstructure are obtained with two boundary conditions: periodic boundary conditions and kinematic uniform boundary conditions. The effect of particle volume fractions, aspect ratio, number and distribution of particles and the type of boundary conditions are numerically studied for different configurations. This paper investigate also the performance of several classical analytical models as Mori and Tanaka model, Halpin and Tsai model, generalized self consistent model through their ability to estimate the mechanical properties of nano-composites. A comparison between simulation results of polypropylene clay nano-composites, analytical methods and experimental data has confirmed the validity of the set results.

1. Introduction

Polymeric composites reinforced with nanoscale reinforcements such as nanotube-reinforced, silica nanoparticle-reinforced and nanoclay-reinforced have recently attracted a tremendous attention in researchers and industrials, since they exhibit enhanced mechanical properties. According to Kojima et al. [17]; clay nanoparticles are classified best candidates to strengthen polymers materials, due to their mechanical and physical properties, their high aspect ratio, their high availability in nature and production low cost.

Generally, there are three different techniques to characterize the behavior of nanocomposites: experimental approaches, analytical methods based on the theories of bounds and models and numerical methods based on the representative volume element (RVE) coupled with finite element methods (FEM). It should be mention that in experimental works it is very difficult to control the influence of the particle size, particles shape and its distributions on the macroscopic behavior of polymer clay nanocomposites (PCN). For that, some works confront the experimental data with numerical and analytical methods to determine the effect of morphological parameters. The elastic properties are then determined by applying analytical or numerical methods. The most important ones are: Mori and Tanaka [21] (MT), Halpin and Tsai [11] (HT) and general self consistent (SC) method. For analytical bounds, the used micromechanical methods are: the first order bounds of Voigt [24]; the second order bounds of Hashin and Shtrikman [10] and the third order bounds of Beran and Molyneux [1]. For numerical characterization, the technique of the homogenization based on RVE and FEM is introduced in many situations in order to estimate the effective properties of nanocomposites. For example, Fornes and Paul [9], Sheng et al. [23], Hbaieb et al. [12], Dong and Bhattacharyya [3], Figiel and Buckley [8] and Pahlavanpour et al. [22].

Fornes and Paul [9] proposed an experimental work to understand the origin of the superior reinforcing efficiency observed in well exfoliated polymer clay nanocomposites compared to conventional reinforcements using composite theory. They found that composite theories of HT and MT were employed to better understanding of the superior reinforcement observed for well-exfoliated

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Nomenclature

Geometric parameters

ζ	aspect ratio of clay particles
P	volume fraction
w_f	weight fraction
ρ_f	clay particle density
ρ_m	matrix density

Effective properties

E_{ij} and $\sum_{ij}$	macroscopic strain and macroscopic stress tensors
$\langle\varepsilon_{ij}\rangle$ and $\langle\sigma_{ij}\rangle$	the average value of strain and average value stress
σ_{ij}	second rang stress tensor
U_i	displacement field
υ	periodic fluctuation
K^{app} and μ^{app}	apparent elastic properties of bulk and shear moduli
E_c	effective elastic modulus
ν_c	effective Poisson coefficient
k_f μ_f E_f	bulk, shear and Young's moduli of clay particles
k_m μ_m E_m	bulk, shear and Young's moduli of matrix
ε_r	relative error

Analytical models

R and V	Reuss and Voigt bounds
HS^+ and HS^-	upper and lower Hashin-Shtrikman bounds
GSC and HT	generalized self consistent and Halpin-Tsai models

nanocomposites relative to conventional glass fibers composites. Sheng et al. [23] employed 2D aligned microstructures combined with micromechanical models of MT, HT and FEM to predict the stiffness of polymer clay nanocomposites. The particles were assumed to be all aligned and isotropic. Hbaieb et al. [12] have used 2D and 3D FEM models of the polymer/clay nanocomposites with aligned and randomly oriented particles to determine the elastic properties of this material. They calculated the effective Young's modulus for random and aligned particles and confront the results to MT model. The authors concluded that the MT model did not predict accurately the stiffness of the composites. Dong and Bhattacharyya [3] predicted the elastic moduli of polymer clay nanocomposites using numerical technique based on mapping the real 2D micro-nanostructures of clay platelets. The results were verified by the comparison of numerical results to experimental data and the conventional composites theories as HT and Hui and Shia [13] models. The results show that the numerical simulations provide the great insight to well predict the elastic modulus of Polypropylene (PP)/clay nanocomposites in comparison to conventional composites theories.

Recently, Pahlavanpour et al. [22] evaluated the performance of commonly used analytical micromechanical models to predict the elastic properties of polymer/clay nanocomposites with the help of numerical simulations based FEM. The results show that the comparison between analytical and simulations revealed that the MT model is the most reliable method to be used for the possible ranges of modulus contrast, aspect ratio and volume fraction. Lielens et al. [18] give a best prediction compared to MT model at high volume fractions when the rigidity contrast between effective particle and polymer is also high. The SC scheme overestimates the axial Young's modulus for all studied cases of PCN (polymer clay nanocomposites).

The majority of micromechanical analytical models do not take into account the influence of the particle shape on the effective properties of nanocomposites. The classical models, their accuracy and their range of applicability, based on more or less suitable hypotheses, cannot be established in the absence of an exact solution, see El Moumen et al. [7]. This can only be obtained by solving numerically the boundary value problem for a RVE of nanocomposites.

In the present paper, the main goal is to predict the effect of particle size, particles number and particles distribution on mechanical properties of randomly partially intercalated and exfoliated polymer clay nanocomposites using numerical homogenization techniques. Several microstructures with different volume fractions and aspect ratio ranging from 5% to 40% are generated. FEM simulations of detailed microstructures are performed with different boundary conditions. The effect of boundary conditions in mechanical properties of clay nanocomposites is also investigated. The RVE size of microstructures and their effective elastic properties are compared and related with number of particles. The results are then compared with both of experimental data of polypropylene montmorillonite nanocomposites (PP/MMT) and conventional composites theories of analytical model.

2. Generation of microstructures and finite element mesh

2.1. Morphology of microstructures

Nanocomposites morphologies were reconstructed digitally using Poisson process. Randomly distributed particles were generated in RVE with an algorithm implemented in MATLAB software. The algorithm is based on composite microstructures and is elaborated by El Moumen et al. [7] for the case of microstructures with ellipsoidal particles. The algorithm was adapted to intercalated and exfoliated polymer clay particles. This process is widely used for generating of composites reinforced with spherical or cylindrical particles, see El Moumen et al. [4]. The idea is to embed points randomly in a 2D plane according to a Poisson law. These points represent the center of each inclusion and then a straight line with a random orientation is generated from each of these points.

In this study, we have considered a 2D microstructure of partially intercalated and exfoliated polymer clay nanocomposites. The morphology of clay platelets (MMT) were carried with different values of aspect ratios: $\zeta = 5$, $\zeta = 10$, $\zeta = 20$, $\zeta = 40$, embedded and randomly oriented in a PP matrix with various volume fractions of 4.5%, 6% and 10%. Fig. 1 shows some examples of the generated microstructures of clay platelets in the matrix including exfoliated and intercalated distribution. The physical and mechanical properties affected to each phase are given by Sheng et al. [23] and Kim et al. [16] and listed in Table 1. It should mention that both of the matrix and clay particles are isotropic and the particles are assumed to be perfectly bound to the matrix.

2.2. Finite element meshing

Once the geometry of the microstructure is performed, a mesh can be generated. The regular finite element mesh is superimposed on the image of the microstructure using the so-called multiphase element technique. This technique was developed by Lippmann et al. [19] and extensively used by El Moumen et al. [5] and El Moumen et al. [7] for homogenization of real and virtual composite microstructures respectively. Indeed, the image of the microstructure is used to attribute the proper phase property to each