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A numerical approach for determining the effective elastic symmetries of particulate-polymer composites

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ABSTRACT

In this paper we deal with the problem of determining on the one hand the effective elastic properties of particulate-polymer composite materials and on the other hand the actual degree of symmetry of the resulting homogenised material. This twofold purpose has been accomplished by building a 2D as well as a 3D finite element model of the heterogeneous material and by using the strain-energy based numerical homogenisation technique. Both finite element models are able to reproduce with a good level of accuracy the real microstructure of the composite material by considering a random distribution of both particles and air bubbles (that are generated by the fabrication process). To assess the effectiveness of the proposed models, we present a numerical study to determine the effective elastic properties of the composite along with a comparison with the existing analytical and experimental results taken from literature and a sensitivity analysis in terms of the spatial distribution of the particles of the unit cell. Numerical results show that both models are able to provide the equivalent elastic properties with a very good level of accuracy when compared to experimental results and that the particulate-reinforced polymer composite could show, depending on the particles volume fraction and arrangement, an isotropic or a cubic elastic symmetry.

1. Introduction

Particulate-polymer composites are used in a wide range of engineering applications. The addition of stiffer particles to a matrix, with easy fabrication process, can improve the mechanical properties of the matrix with a slight increase of weight.

An exhaustive review on the effects of particle size, particle/matrix interface adhesion and particle loading on the stiffness, strength and toughness of such particulate-polymer composites is given in Ref. [1]. In particular all of the reviewed papers concern experimental works and/or analytical theories developed on this subject. In Ref. [2] the authors developed an analytical formulation predicting the tensile strength of particulate-filled composites taking into account also the effect of the particle size. Ishai and Choen [3] developed a theory giving the lower and upper bounds for the Young's modulus of a two-phase particulate composite whose Representative Volume Element (RVE) was a cubic matrix with a cubic inclusion. More recently in Ref. [4] Lin and Ju presented an analytical study to predict the effective elastic moduli of three-phase composites containing randomly dispersed spherical particles. A comparison between different classical homogenisation methods is presented in Ref. [5]. Concerning the experimental-based studies on this particular class of composite materials, in Ref. [6] the effects of particle size on the mechanical and impact properties of cured epoxy resins filled with spherical silica have been analysed. Another experimental study on the effect of the size and of the volume fraction of alumina trihydrate particulate inclusions on the mechanical strength of an anhydride-cured epoxy resin was conducted in Ref. [7]. Spanoudakis and Young [8] evaluated experimentally the effect of strain rate, volume fraction and particle size upon the stability of crack propagation, the Young's modulus, the critical stress intensity factor, and the fracture energy in an epoxy resin reinforced with spherical glass particles. The tests have been performed through double-torsion tests. In a more recent work [9] both experimental and numerical analyses have been shown. The effect of concentration of Aluminium trihydrate (ATH) on the strength of polypropylene (PP) has been addressed. In the past years many others experimental works have been carried out on this subject, see for instance [10–14].

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The general conclusions of these works are the following ones:

- above a particular size of the particles it seems that there is no effect of the particles size on the composite equivalent elastic moduli, while, when the particles size is lower than this value the size has a more significant effect on these properties. However, this particular dimension is not uniquely defined but it depends upon the particles and matrix properties together with the adhesion between these two phases;
- the volume fraction of particles is the most important parameter for the evaluation of the effective properties of the equivalent homogenised material;
- the matrix–particles interface adhesion has only a slight effect on the elastic properties.

If, on the one hand, the works on the numerical homogenisation of the mechanical properties of polymeric materials reinforced with long or short fibres are numerous, on the other hand, the numerical analyses to determine the properties of particulate-polymer composites are less numerous. In Ref. [15] the authors developed a gradient-enhanced computational homogenization procedure in order to take into account the effects of the micro-structure on the macro-scale equivalent continuum. The 2D finite element (FE) model of the aluminium plate was made taking into account the voids at the micro-scale level. In Ref. [16] the authors present a numerical homogenisation of the elastic properties of a three-dimensional cubic unit cell containing non-overlapping identical spheres randomly distributed using a random sequential algorithm suitable for particle volume fractions of up to 50%. Wernik and Meguid [17] modelled the micro-structure of the RVE of carbon nanotube-reinforced structural adhesives in order to determine numerically the constitutive response of the adhesive.

Other woks on the determination of the mechanical behaviour at the macro-scale of heterogeneous materials are [18] and [19]. In Ref. [18] the authors studied the influence of different micro-structures and the grain size effect on the mechanical behaviour at the macroscopic scales of zirconium alloys. In Ref. [19] an analysis on the effect of the strain distribution at the micro-scale on the mechanical properties at the macro-scale was conducted. Finally, in Ref. [20] the authors showed an FE-based homogenisation of SiC/SiC composites using a Statistical Volume Element (SVE) to take into account the microstructure of the material.

Very often, lately, the works on the numerical determination of mechanical properties of such a kind of composites, are focused on a given field of applications: the particulate reinforced adhesives. Indeed, epoxy adhesives are brittle and need to be toughened by adding elastomeric particles, hard particles (silica, metal, etc.) or nano-components. The studies on this special class of materials aim to determine the strength properties, the fracture energy [8] or the whole interface decohesion behaviour [21,22]. However, regarding the elastic behaviour of the material, all of the works do a very strong hypothesis: the material is assumed to be isotropic despite the inclusions can introduce a certain degree of anisotropy. The aim of the present work is to develop numerical FE-based models capable of reproducing a composite microstructure which is representative of the one observed on real materials. Subsequently these models will be used to perform numerical analyses in order to calculate the full set of elastic properties of the material. With such protocol, we will be able to determine the degree of anisotropy of the equivalent homogeneous material at the macro-scale. In addition, to prove the effectiveness of the proposed FE models the sensitivity to particles size, distribution, and particles volume fraction is studied and the results are compared with the analytical and experimental ones taken from literature. The paper is organised as follows: the description of the particle-filled matrix homogenisation problem is introduced in Section 2. The 2D FE model along with the related sensitivity analysis and the comparison and validation with analytical and experimental results is presented in Section 3. The 3D FE model together with the related numerical study to determine the full set of elastic properties and its validation with other existing analytical results are detailed in Section 4. Finally, Section 5 ends the paper with some concluding remarks.

2. Homogenisation of the elastic properties: problem description

In the last decades, several analytical, numerical and experimental techniques have been developed in order to determine the effective properties of composite materials as a function of geometric and material properties of both the inclusions and the matrix. Each method presents a certain level of sophistication. Concerning the analytical solutions, the majority of theories developed to determine the Young modulus of particulate-polymer composites are based on empirical or semi-empirical equations. For example, the Einstein's equation [23,24] for the prediction of the Young's modulus of particle-filled polymers is based on the assumptions of perfect bonding between particles and matrix and perfect uniform distribution of inclusions. This theory does not take into account the effect of particles size: the Young's modulus is a linear function of the particles volume fraction V_p. Moreover it does not take into account the interaction between the adhesive and the particles in terms of stress and strain field, hence, it is valid only for low values of V_p. Successively others more elaborated theories have been developed to improve the Young's modulus estimation, see for example [25–29]. Since the analytical techniques make use of some simplifying assumptions to obtain the elasticity solution of the composite material, some theories, rather than defining a single value of the Young's modulus, determine the bounds of the modulus itself. Concerning the case of particulate micro and nano-composites we can cite the Reuss-Voigt's Lower (RV-LB) and Upper (RV-UB) Bounds [30,31] and the Hashin-Shtrikman Lower (HS-LB) and Upper (HS-UB) Bounds [32] which are more restrictive than those of Reuss-Voigt.

To improve the proposed theories several studies have been conducted to develop new experimental-based analytical techniques [33] for determining the effective properties of the composite material. However, experimental-based methods, see for instance [34–36], require a standardised procedure for the measurements and the main drawbacks of these procedures consist in the fact that they are very expensive in terms of both time and money.

In this regard, the numerical methods can present a valid support for the experimental tests. Contrary to analytical theories, numerical-based techniques such as FE methods, do not make use of simplifying assumptions and are not expensive as the experimental tests. In addition, depending on the level of refinement of the model, they can lead to realistic solutions in terms of the elastic response of the structure. Therefore, the use of valuable models can indeed support a campaign of numerical tests in two main ways:

- orient in a focused manner the types of experimental tests to be conducted and, then, back to validate and confirm the numerical results;
- if the numerical model is validated the number of tests to be performed can be considerably reduced therefore leading to a reduction of costs and time.

As a consequence, in this work we have chosen a FE-based approach as a numerical homogenisation technique to determine