MODELLING INTERACTION EFFECT OF NANOSILICA PARTICLES ON NANOSILICA/EPOXY COMPOSITE STIFFNESS

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Abstract

Tensile testing is carried out to obtain stiffness and strength properties of Nanopox F400/ Epikote 828 composite system. A graded interphase of nanosilica particles is considered for the micromechanics prediction. Ten layers of interphase on each nanosilica particle are simulated with varying material properties in each layer. A numerical methodology, implemented in Abaqus, is based on the representative volume element (RVE) concept.
The addition of 10% volume fraction of nanosilica into the resin system enhanced the elastic modulus of the neat resin by nearly 40%. Both the presence of a graded interphase around the reinforcing particles and interaction effects play an important role and need to be accounted for the prediction of the elastic modulus. Results from the simulations are compared to measured data; a good correlation is achieved provided that the properties of the interphase are accurately represented.

1. Introduction

Many efforts have been made to increase the toughness of epoxy- based composites. One of the effective techniques to improve the fracture toughness of a composite is to enhance the epoxy matrix toughness by adding fillers, such as nanosilica particles. In this work, the epoxy polymer modified with nanosilica particles is chosen. This optimized composite system is expected to be superior in stiffness, strength and toughness properties that are so much required in the design and building of aerospace and other advance engineering structures.

Predictive modelling based on the physics of composite material behaviour is wealth generating; by guiding material system selection and process choices, by cutting down on experimentation and associated high costs; and by speeding up the time frame from the research stage to market [1]. The prediction of the macroscopic stress-strain response of composite materials related to the description of their complex micro-structural behaviour exemplified by the interaction between the constituents and the overall property of a composite material is governed by the properties of its constituents and microstructures [2]. Many models have been formulated to predict the effective (overall) mechanical properties of particulate composite materials. The so-called "effective" properties of a particulate composite material are achieved by homogenization process over a “representative volume element” (RVE).

Many micromechanics approaches have been proposed for the evaluation of the overall elastic property of heterogeneous materials and their dependence on the properties of constituents and the microstructure of materials. Conventional micromechanical analytical models such as Mori-Tanaka [3], Halpin and Kardos [4] widely used for usual composites with micro-sized reinforcements, were recently used to predict the overall stiffness of nanocomposites [5].

Boutaleb et al. [5] have introduced a micromechanics-based analytical model on silica particle/polymer nanocomposite that can predict the stiffness and yield stress by taking the role of nanoparticle size, interphase and volume fraction into account regardless interaction effect of the particles. The result showed that overall nanocomposite behaviour was controlled by interphase surrounding the nanoparticle. It was concluded that due to changes in molecular structure, the interphase presents a graded modulus, ranging from that of silica to that of polymer matrix. A good agreement was found on the comparison between the model and available experimental data.

A continuum-based constitutive model for nanosilica/polyimide composites was introduced with involving four different nano-silica/polyimide interfacial treatments [6]. Molecular modelling techniques were applied to evaluate the equilibrium molecular structures of RVEs of the material system. The elastic properties and dimensions of nanoparticle and polyimide were calculated using Molecular Dynamic (MD) simulation. Mori-Tanaka method and effective interface model were applied to predict composite Young’s moduli and shear moduli. The Young’s moduli and shear moduli remained constant for all material system. The result showed that the model can be used to describe tensile stress-strain results of a metal matrix particulate composite as well as the corresponding response of particulate polymer nanocomposite.

In this work the modulus of elasticity of the RVE is predicted using finite element analysis (FEA) by incorporating graded interphase and interaction effect of the particles into account. The graded interphase was used according to the work done by Boutaleb et.al [5]. The whole thickness of the interphase applied in the FEA and micromechanics is varied according to the nanosilica volume fraction [7].

2. Micromechanics Model

Micromechanics model analysed in this paper is based on the model proposed by Boutaleb et al. [5] with single particle in a RVE. Graded interphase of the particle was assumed due to the perturbed region undergoing a gradual transition from the properties of the silica to the properties of the matrix. Constitutive equation of the equivalent homogenous medium is presented by:

$$\bar{\sigma} = \bar{C} : \bar{\varepsilon} \tag{1}$$

where “:“ signifies the multiplication between two fourth-order tensors, $\bar{C}$ is the effective linear-elastic stiffness tensor, $\bar{\sigma}$ and $\bar{\varepsilon}$ are the macroscopic (homogenised) stress and strain tensors, respectively. Macroscopic stress and strain tensor are derived by using homogenisation method as follows:

$$\bar{\sigma} = \frac{1}{V} \int_V \sigma\,(x)dx \tag{2}$$

and

$$\bar{\varepsilon} = \frac{1}{V} \int_V \varepsilon\,(x)dx \tag{3}$$

where V is the RVE volume, σ and ε are the microscopic stress and strain tensors, respectively. Overall elastic stiffness tensor of the nanocomposite is expressed by:

$$\bar{C} = C^0.\left[I - \phi^{\Sigma}T^{\Sigma}.\ (\phi^{\Sigma}S.T^{\Sigma} + I)^{-1}\right] \tag{4}$$

Where S is the Eshelby's tensor, I the fourth-order identity tensor, ϕ^{Σ} is the volume fraction of entire inclusion including interphase and C^0 is defined as elastic stiffness of the matrix and T^{Σ} is the fourth-order tensor.

The interphase is divided into ten layers and the elastic modulus of each layer *i* of the interphase $(E_I)_i$ is defined by:

$$(E_I)_i = \frac{r_{\Sigma}}{r_i} E_M + \left(\frac{r_{\Sigma} - r_i}{e}\right)^{\beta}\left(E_P - \frac{r_{\Sigma}}{r_P} E_M\right) \tag{5}$$

Where r_{Σ}, r_P and r_i are radius of the entire inclusion (particle and interphase), particle and radius of i_{th} layer, respectively. E_M is elastic modulus of matrix material and E_P is the elastic modulus of particle and e stand for interphase thickness. β represents the interphase parameter which depends on the bonding strength between the phases. The higher the β value the weaker the bonding strength and vice versa.

3. Finite Element Model

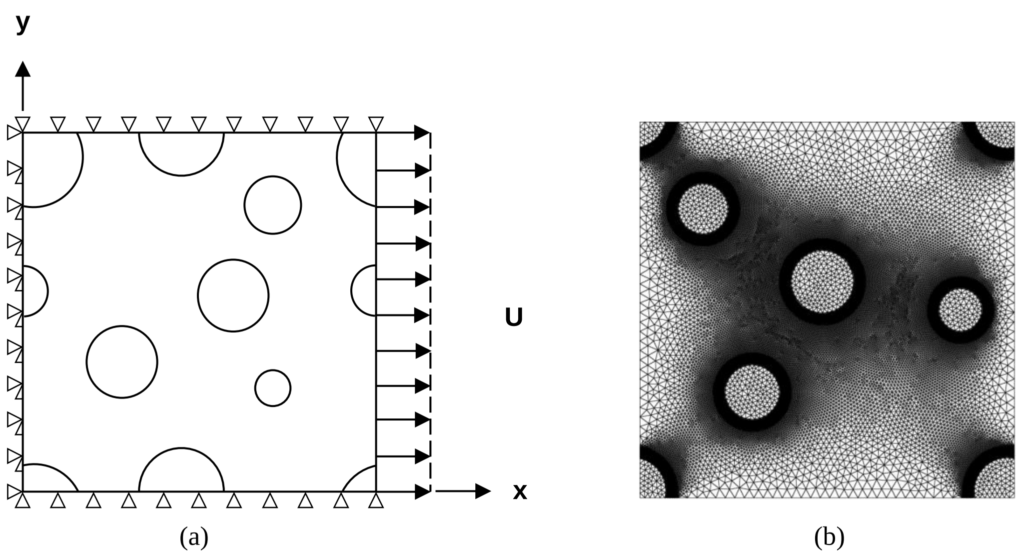


Figure 1. (a). Unit cell FE model in 2D with material and boundary condition periodicity. (b). Typical unit cell for 10% volume fraction of particles with size 160x160 nm^2.

FE analysis was performed in order to examine the role of interphase and volume fraction of the nanosilica particles embedded in epoxy polymer. Multi particles model was applied in FE model as well as single particles model. Multi particles simulation was successfully carry out by Gitman [8] to examine quasi-brittle material behaviour during tensile loading using RVE with single layer of interphase.

Multi particles were generated within rectangular matrix with random distributions and different particle size in a range between 20 to 40 nm with graded interphase around the particles. Periodicity of the material and boundary conditions were assumed for a unit cell. The idealisation of the FE problem can be seen in figure 1 (a).Volume fraction of the particles was 2, 5, 8 and 10%. The size of unit cell was 160 x160 nm with five different realisations in each volume fraction. Figure 1 (b) shows the unit cell with 10% of nanosilica volume fraction.

Abaqus FEA software was used in this simulation. Graded interphase was set as a third phase in the perturbed region around nanoparticles. The whole interphase thickness was defined according to particle volume fraction [7, 9]. A perfect bonding assumption is applied between phases. Displacement load was applied on each model with the value equal to 1% strain. Single particle model was also conducted in this work as a comparison with multi particle model.

4. Experimental Details

4.1. Materials

Epikote 828 epoxy, a diglycidyl ether of bisphenol-A (DGEBA) was used and cured with an alicyclic anhydride hardener. Epikote 828 is a low viscosity resin. Nanopox F400 is nanosilica (SiO_2) reinforced bisphenol-A based epoxy resin. It was supplied as a colloidal solution with 40 wt% in epoxy resin. This nanosilica is commonly used in fibrous composites. Evonik Hanse GmbH, Geesthacht, Germany supplied the spherical silica nanoparticles with average particle size about 20 nm and maximum diameter below 50nm. The silica phase has surface-modified with an extremely narrow particle size distribution.

4.2. Nanosilica epoxy composite fabrication.

4.2.1. Pure Epikote 828 polymer

Epikote 828, 1-methyl-5-norbornene-2,3-dicarboxylic anhydride (NMA) and benzyl dimethyl amine (BDMA) with stoichiometric ratio 100:90:1 were mixed in a glass beaker. The mixture was stirred in a heated paraffin oil bath of 80°C by using mechanical stirrer for 1 hour. After stirring, Epikote 828/NMA/BDMA mixture was degassed in the vacuum oven at 80°C for 20 minutes and then poured into the preheated rubber mould. Curing process in programmable oven included pre-cure at 80°C for 2 h, cure 120°C for 3 h, post-cure at 150°C for 4 h, with ramp rates of 1°C/min. After curing has been done, all the specimens need to be ground and polished to get the required dimension with accuracy of 0.01 mm.

4.2.2. Nanopox F400 Epikote 828 composite

Epikote 828 was mechanically mixed and stirred with of Nanopox F400 in a heated paraffin oil bath of 80°C for 1 hour at 400 rpm. The Nanopox F400 was taken in specific amount with the purpose of preparing nanocomposites with 3-16 wt% nanosilica content. The mixture was completely degassed in a vacuum oven at 80°C in order to get rid of the entrapped air during stirring process. Then the curing process was performed as described in section 4.2.1. Nanosilica volume fraction was measured based on ASTM E1131 [10], ASTM D972 [11] and ASTM D3171-99 [12]. The fabrication process of the pure Epikote 828 and Nanopox F400/Epikote 828 composite referred to work done by Jumahat et al. [13].

4.3. Tensile test of the pure Epikote 828 and Nanopox F400 modified Epikote 828 composite

Five dogbone shape specimens were tested for each pure Epikote 828 and Nanopox F400/Epikote 828 composite system. Tensile tests were conducted by using a Tinius Olsen universal testing machine at crosshead speed of 1 mm/min with wedge type grips. All the data were acquisitioned using computer interface for analysis. The tensile test was conducted according to British standard BS EN ISO 527-1 and -2: 1996 [14].

5. Result and discussion

5.1. Micromechanics prediction

Interface parameter β demonstrates the effect on the variation of the elastic modulus inside interphase as depicted in figure 2 (a) according to equation 5. In relation to a surface chemical treatment or surface modification of the nanosilica, the lower β value corresponds to a stronger bonding between two phases while the higher β value correlates to a weak bonding between the two phases. As the consequence, interface parameter β also presented considerable effect on the elastic modulus of nanosilica composite as shown in figure 2 (b). Elastic modulus of the nanosilica epoxy composite increases as the β value decrease for each nanosilica volume fraction. Nevertheless, the interphase itself also has strong influence on the elastic modulus of the nanosilica composite as the elastic modulus of nanosilica composite with no interphase is lower than the one with a very low bonding strength between interphase (β = 100).

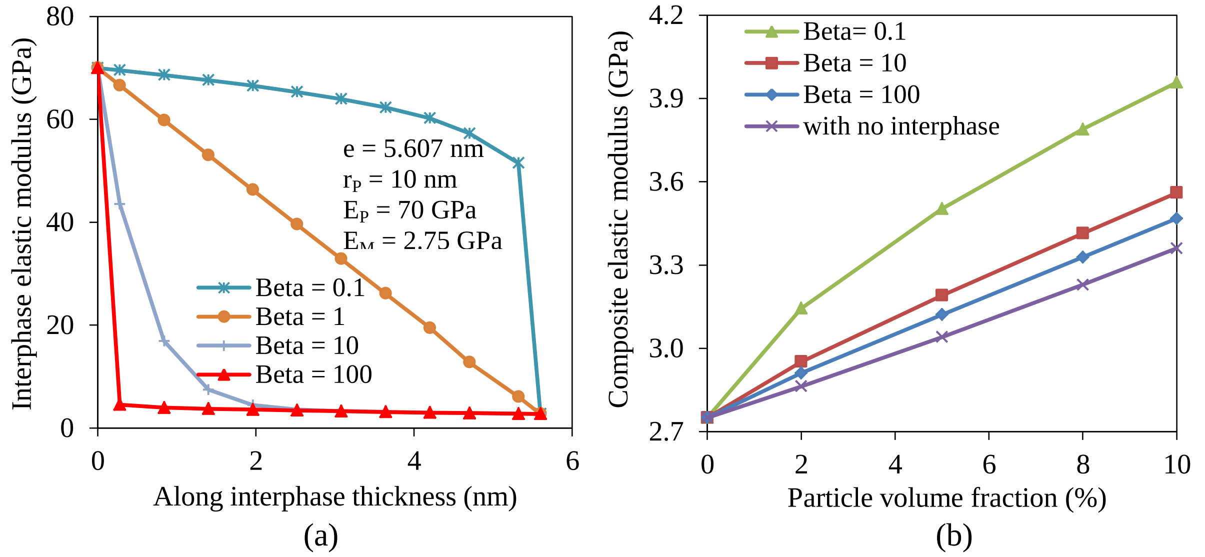
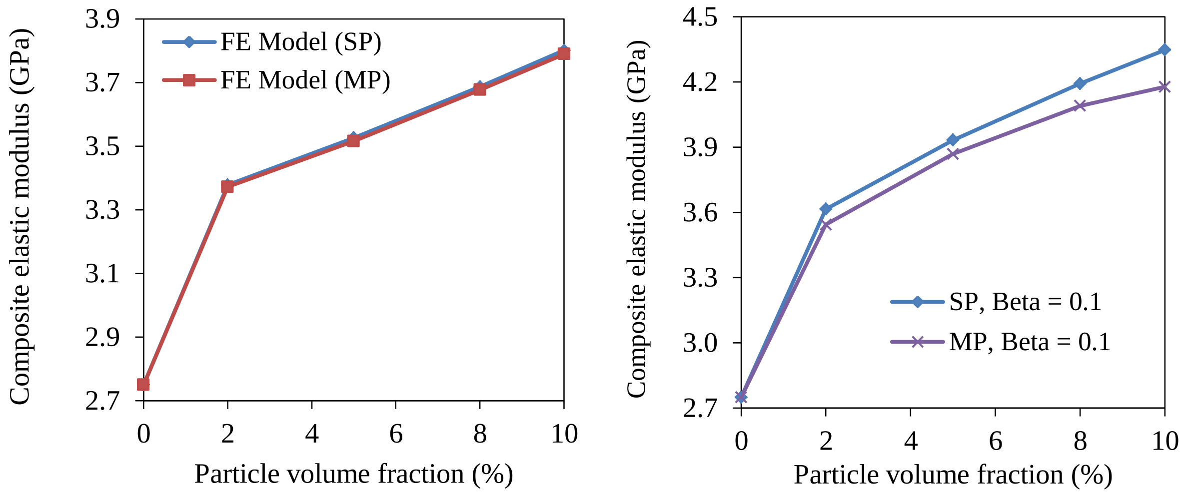


Figure 2. Influence of β parameter on the interphase elastic modulus along interphase thickness (a). Micromechanics prediction of elastic modulus of nanocomposites with graded interphase (b).

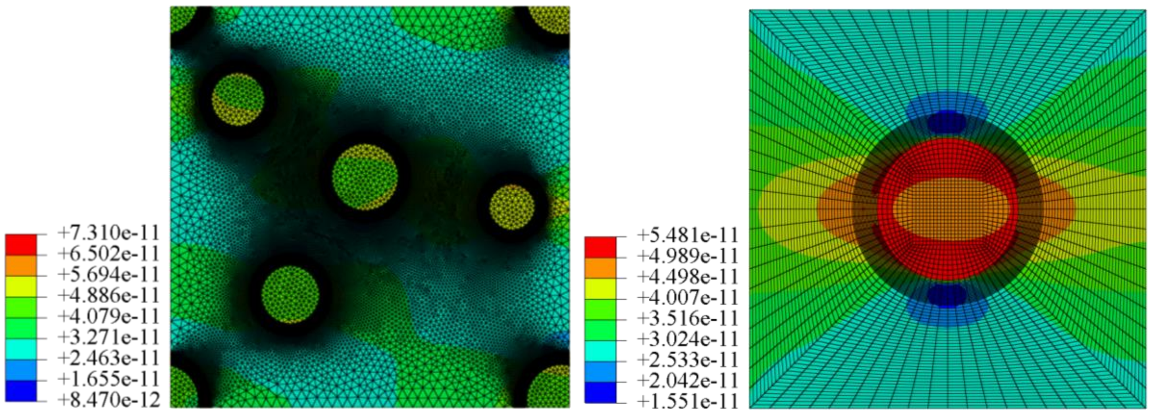
5.2. Finite Element (FE) Analysis

Finite element analyses were carried out by employing multi particles RVE with and without graded interphase. As a comparison, single particle RVE analyses were also performed. Elastic modulus of multi particles RVE increases as particle volume fraction increases. Elastic modulus of multi particles RVE with various particle volume fractions superimpose with elastic modulus of single particle RVE as illustrated in figure 3 (a). Nevertheless, if graded interphase is applied around particles then multi particle FE analyses present a decrease on the elastic modulus of nanosilica composite compared to single particle FE analyses as depicted in figure 3 (b). This is due to the interactions between particles in multi particles models with graded interphase as shown in figure 4 (a).



(a) (b)

Figure 3. Comparison of elastic modulus based on single particle (SP) and multi particles (MP) FE analysis without interphase (a) and with graded interphase (b).



(a) (b)

Figure 4. Von Misses Stress distribution of the RVE with graded interphase, 10% of volume fraction of nanosilica and beta value of 10 on multi particles (a) and single particle FE analyses (b).

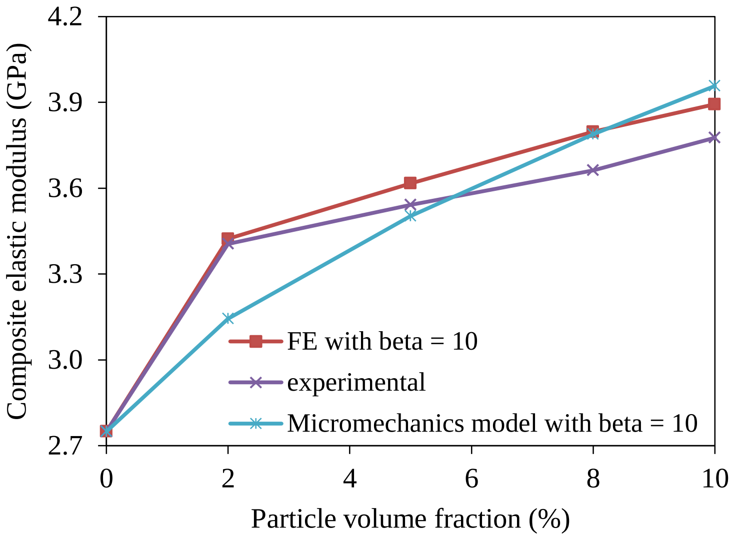


Figure 5. Composite Elastic modulus comparison of the Nanopox F400 modified Epikote 828 composite between experimental, FE modelling and micromechanics model.

5.3. Experimental Result

Tensile test has been performed to validate analytical and numerical modelling. It can be seen from figure 5 that the addition of nanosilica particles to the matrix has significant influence on the elastic modulus of the composite with an increase as the nanosilica content increases. In addition, the nanocomposite exhibits higher tensile strength and failure strain which also indicate higher fracture toughness. Fracture toughness enhancement of nanocomposites is achieved by bending the crack path around the nanosilica surface [15] which also leads to a higher failure strain of the nanocomposites system.

Figure 5 shows that the multi particles FE model with beta value of 10 exhibits good correlation with experimental results and an improved prediction compared to the micromechanical approach.

Conclusions

The addition of nanosilica particles to the matrix system significantly improves the tensile properties of the composite. Finite element results have shown the influence of incorporating the interphase and interaction effect on the prediction of the elastic modulus of nanosilica epoxy composite. The developed multi-particle FE model with graded interphase also shows a very good correlation with experimental result.

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