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Analysis of the effect of manufacturing imperfections in the elastic properties of platelet nanocomposites

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ABSTRACT

We have developed and validated a conceptually simple model capable of predicting the macroscale elastic properties of a platelet nanocomposite. The model allows for studying the individual and combined effect of the parameters with influence on those properties, namely nanofiller weight fraction, misalignment, dispersion quality, size distribution and nanofiller-matrix interfacial characteristics. The model shows a very good correlation with experimental results. The interfacial characteristics under different strain states are evaluated at the nanoscale by means of a cohesive model which considers out-of-plane strains and angular distortions, so that the full, strain-dependent elastic tensor can be calculated, allowing for homogenization and subsequent study of the effect of filler orientation, dispersion quality and size distribution on the elastic properties at the macroscale. The use of a low complexity nanoscale model allows us to conceptually and quantitatively explain the causes underlying the divergences between the expected and experimental macroscale material stiffness experimentally found by different researchers.

1. Introduction

Nanofillers such as nanofibers, nanocellulose, carbon nanoparticles or nanoclays, have attracted considerable attention over the last decades due to their remarkable mechanical, thermal, electrical and electronic properties [1,2]. From the mechanical point of view, notable increases in stiffness, strength, fracture toughness and fatigue life have been reported in different nanocomposites for small nanofiller contents [3–6]. However, for a given nanofiller content, the experimentally measured mechanical properties differ greatly from the theoretically expected ones. For example, Bortz et al. [3] report a notable difference between the expected (Halpin-Tsai model) and experimental results in the Young's modulus of graphene oxide (GO)-epoxy composites. Fig. 1 shows that for contents over 0.1% in weight, the stiffness increase is progressively smaller, even negative. Tang et al. [7] measured very slight increases in the Young's modulus even when improving the dispersion in a reduced graphene oxide (RGO) nanocomposite. Wan et al. (2014) [8] report scarce improvement in the Young's modulus when increasing the GO weight fraction over 0.1%, the tendency being the same even when the GO was functionalized. Wang et al. [9] found the same tendency of reduction in the marginal reinforcement effect when increasing the filler content. Wan et al. (2013) [10] also found the same results even when improving the dispersion and the interface quality. Wei et al. [11] and Ahmadi et al. [12] report comparable results with graphene nanoplatelets.

The degree of enhancement obtained is mainly dependent of four parameters: firstly, dispersion plays a fundamental role, as nanoparticles tend to agglomerate [7]. Secondly, the stress transfer capacity at the nanoparticle - matrix interface, which depends on the interfacial stiffness and strength. Thirdly, as different interfacial models [9] demonstrate, axial strain at the nanoparticle grows from the nanoparticle ends towards the center, as it is obtained due to the shear at the interface, which is maximum at the ends. Consequently, a critical length is needed for achieving a strain at the nanoparticle center which is the same as that imposed at the matrix. The nanoparticle average stress will be lower, and consequently the nanocomposite stiffness will also be reduced, if the length is comparatively smaller than this value.

Finally, nanoparticle alignment will affect the nanocomposite mechanical properties along each direction. Various studies have focused on the degree of improvement when a dominant orientation is present [13,14]. Some empirical models, such as the proposed by Krenchel [15], give approximations for aligned and misaligned nanocomposite mechanical characteristics, which are still used nowadays for the correction of the mechanical properties obtained at the nanoscale.

The Halpin-Tsai [16] is a semiempirical, approximate model which provides parametric expressions for obtaining the mechanical

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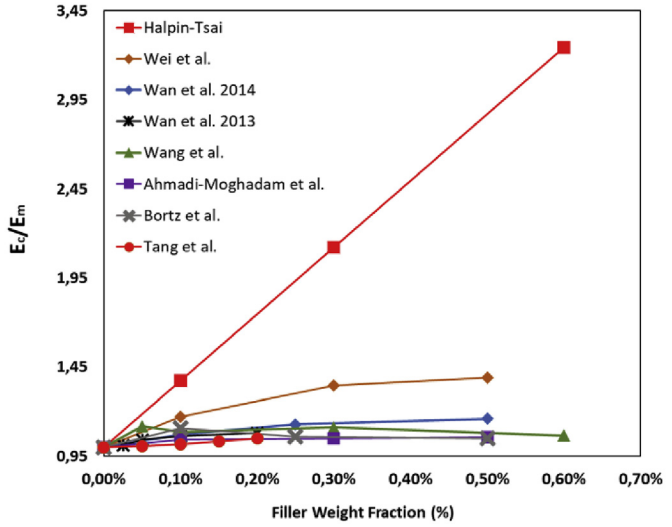

Fig. 1. Young's modulus experimental results (Bortz et al. [3], Tang et al. [7], Wan et al. (2014) [8], Wang et al. [9], Wan et al. (2013) [10], Wei et al. [11], Ahmadi et al. [12]) and Halpin-Tsai results.

characteristics of the nanocomposite. More accurate nanoscale models have been reported [17–22] for the prediction of different nanocomposite properties in particular cases. Other researchers conduct homogenization in a minimum representative nanoscale domain and extract some conclusions at the macroscale [23–26]. However, few models take into account, by means of a generalized approach, the simultaneous as well as the individual influence of all the relevant nano- and macroscale parameters in order to accurately predict the complete macroscale elastic properties.

This paper presents a multiscale model capable of taking into account all the nano- and macroscale parameters explained, requiring a considerably low computational effort, which shows a very good correlation with experimental results. In addition, the simplicity of the nanoscale model allows for the derivation of conceptual and quantitative conclusions regarding the influence of each parameter and also its combined effect on the macroscale elastic behavior. The multiscale methodology we propose in this work can be adapted to any kind of nanofiller. However, the particular examples we provide correspond to an epoxy/graphene nanocomposite. As the stress state and damage mechanisms arising at the nanoscale will strongly depend on the nanofiller shape, size, aspect ratio and mechanical properties, the results would differ to those obtained if other kind of nanofiller had been used.

2. Methodology

When a uniaxial strain state is imposed in a macroscale domain, local strain states at the nanoscale will not be uniaxial if, for instance, misalignment exists. A correct representation of the macroscale stress tensor implies the consideration of the full strain state (1), where subindex i represents a generic nanoscale domain.

$$[E_x,\ E_y,\ G_{xy}]_i = f(\varepsilon_x,\ \varepsilon_y,\ \gamma_{xy})_i \tag{1}$$

However, constitutive Eq. (1) are governed by the mechanisms previously cited in section 2. The resulting nanoscale behavior will be considered by means of a cohesive finite element model able to allow for an accurate homogenization (2) (Fig. 2).

$$[E_x,\ E_y,\ G_{xy}]_i = f(\overline{\sigma_x},\ \overline{\sigma_y},\ \overline{\tau_{xy}}) = f(\varepsilon_{xi},\ \varepsilon_{yi},\ \gamma_{xyi},\ l_F,\ t_F,\ \upsilon_F,\ E_F,\ G_F,\ E_M,\ G_M,\ k_I,\ \tau_a,\ k_{II},\ \sigma_a,\ G_{II}) \tag{2}$$

l_F being the nanofiller length, t_F the nanofiller thickness, υ_F the nanofiller volumetric fraction, E_F, E_M, G_F and G_M the nanofiller and matrix Young's and shear modulus, respectively; k_I and k_{II} the in-plane and out-of-plane interface stiffnesses, τ_a and σ_a the in-plane and out-of-plane interface strengths, and G_{II} the interfacial fracture energy.

To obtain a simple yet accurate model, at the nanoscale we only consider the nanofiller and interfacial properties. The remaining relevant parameters -misalignment, dispersion and length distribution-are considered at the macroscale, after homogenization is carried out at the nanoscale model. This approach does not imply a substantial error, given the quite good correlation of both nanoscale and macroscale models with experimental results (see sections 2.1. and 3.3.).

Homogenization is numerically carried out through application of the generalized Hooke's elasticity equations using the calculated mean stresses $\overline{\sigma_x}$, $\overline{\sigma_y}$, $\overline{\tau_{xy}}$. Homogenization results for different imposed strain states, volumetric fractions, nanofiller lengths and interfacial characteristics can then be stored as lookup tables for subsequent use in the macroscale model.

The macroscale model is also solved by means of the finite element method. Element strains are calculated and element constitutive equations are consequently updated at each load step by means of the lookup homogenization maps. With this methodology, misalignment state, dispersion quality and length distribution can be easily considered, by assigning different local coordinate system orientations and constitutive parameters at each element. Finally, macroscale stiffness can be calculated by application of the generalized Hooke's law at the macroscale domain.

It is important to point out that the proposed model is intended for the calculation of the nominal elastic properties of nanocomposites, that is, in the small strain range, in the same fashion as other researchers, whose results have been used for validation [27,28]. This allows for assuming that debonding is the main nanoscale damage mechanism, as experimentally validated by other authors [29,30]. Consequently, we do not include other damage mechanisms, as crack tipping, bridging or shear banding, only observable at crack surfaces, where bigger strains have occurred.

2.1. Nanoscale model

We have used a Cohesive Zone Model (CZM) with mixed mode debonding for calculating the resulting stresses required for homogenization. As misalignment level and dispersion quality will be considered at the macroscale, a Minimum Representative Domain (MRD) was defined by assuming a perfectly dispersed and aligned nanocomposite (Fig. 2), in the same fashion as other researchers [31].Where

$$t_m = t_F \frac{1-\upsilon_F}{\upsilon_F} \tag{3}$$

Different combinations of imposed longitudinal and transverse deflections Δ_X, and Δ_Y, were imposed at the MRD matrix free edges (Fig. 2), as a function of the desired strain state (ε_X, ε_Y, γ_{XY}) to be imposed, according to (4), where x and y are coordinates of each edge position at which a displacement is imposed.

$$\begin{pmatrix}\Delta x\\ \Delta y\end{pmatrix} = \begin{bmatrix}\varepsilon_X & \frac{\gamma_{xy}}{2}\\ \frac{\gamma_{xy}}{2} & \varepsilon_Y\end{bmatrix}\begin{pmatrix}x\\ y\end{pmatrix} \tag{4}$$

FEM discretization and model resolution were performed with the software *Comsol*®. CZM interfacial elements and a mapped, shell element mesh with adequate aspect ratio and size were used. Symmetry boundary conditions were not considered in the MRD due to the antisymmetry associated to shear.

We validated the nanoscale cohesive model by comparison of the results obtained by imposing a uniaxial strain state with the experimental results obtained by Guo et al. [27] and Lee et al. [32]. As shown in Fig. 3, a remarkable correlation is obtained when using the same stiffnesses, nanofiller length and volume fraction and interfacial parameters as those found in the experiments (k_I = 74 TPa/m, τ_a = 0.5 MPa, G_F = 0.08 J/m).