



Nonlinear multiscale modelling of quasi-solid state behaviour of PET/MWCNT nanocomposites: 3D RVE-based approach

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ABSTRACT

Nonlinear 3D RVE-based computational multiscale model for prediction of the quasi-solid state behaviour of PET/MWCNT nanocomposites is proposed in this work. The model links the time and length scale of representative morphology of the nanocomposite with the corresponding macroscopic scales via the numerical homogenisation. Finite Element simulations of the uniaxial extension of the nanocomposite near the glass transition show that the addition of MWCNTs leads to significant stiffening of PET. It is caused by an excellent reinforcing ability of the nanotubes, which on average are found to carry the load around two orders of magnitude larger than the matrix. A significant decrease of nanocomposite stresses with increasing processing temperature in its narrow range near the glass transition is also predicted by the model.

1. Introduction

Subjecting entangled polymers to fast and large deformations near their glass transition is a common approach for manufacturing polymeric products. That approach can also be relatively easily adopted for polymers reinforced with nanoparticles [1–3]. During those processes polymers are in the semi-solid state and encounter complex nonlinear viscoelastic behaviour within, or near the temperature/time (rate) window between the glass transition and relaxation of entangled network (characterized by the time of tube disengagement by reptation). Thus, the polymer rheology is highly complex under these conditions, and for some entangled polymers it is further affected by structural instability under large deformations, which can cause stress-induced crystallization. Presence of nanoparticles in the polymer adds further complexity in disturbing the rheological behaviour under processing conditions. Thus, advanced multiscale modelling of polymer nanocomposites can assist in providing more insight into the effects of nanoparticles on the polymer behaviour in the processing regime of interest. Then, it can also be used in simulations to find optimum material composition and optimum quasi-solid state processing conditions for polymer nanocomposites.

The rheological behaviour of entangled polymers and their nanocomposites under those conditions is challenging to capture with a mathematical model that can further be used in numerical simulations and optimization of the process. Several attempts have been made over the last two decades in developing models that capture the large deformation behaviour of unfilled entangled polymers under the conditions of interest [4–6]. However, there is only few attempts in

describing the behaviour of nanofilled polymers in the same temperature and strain rate window. Particularly, the research has exclusively focused on two-dimensional (2D) multiscale RVE-based modelling polymer nanocomposites with nanoplatelet-like reinforcement (i.e. nanoclay-filled polymers) [7–10]. However, the 2D RVE approach is unable to capture accurately the nanocomposite morphology and interactions between the nanotube-like nanoparticles and polymeric matrix, and thus it cannot describe correctly the stress transfer from the matrix to the nanotube across the interface.

Therefore, this work aims at developing a 3D multiscale nanocomposite model for predicting the effects of multi-walled carbon nanotubes (MWCNTs) on macroscopic behaviour of poly(ethylene terephthalate) (PET)/MWCNT nanocomposites just above the glass transition. It is achieved here by combining a 3D Representative Volume Element (RVE) concept with nonlinear homogenisation, and physically-based constitutive model for PET.

This communication starts with a short description of the multiscale model for the nanocomposite. It is then followed by the results and their discussion.

2. 3D RVE-based multiscale nanocomposite model

2.1. Multiscale modelling framework

An ultimate multiscale framework for a nanocomposite should link interactively length and time scales related to molecular interactions between the nanofiller and polymer matrix, nanofiller dispersion and distribution that define nanocomposite morphology, and macroscopic

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response, to provide accurate predictions of processing-structure-property relationships for nanocomposites. However, the time scale of the current problem, which considers relaxation processes in the polymer matrix near the glass transition, extends well above nanoseconds. This is currently beyond capabilities of molecular dynamics (MD)-like simulations even when one considers a small nanoscale domain consisting of a single nanoparticle and a surrounding polymer. Thus, a two-scale approach of linking nanocomposite morphology and macro scales, enhanced with a physically-based model for the polymeric matrix to capture the aforementioned time-dependent processes, was pursued in this work. The approach used in this work is based on the Representative Volume Element (RVE) concept and the nonlinear homogenisation (coarse-graining) that generally enables the macro-to-RVE and RVE-to-macro transitions. For that it is assumed that the scale separation holds, and that the RVE is statistically representative of the entire nanocomposite morphology. Moreover, it is assumed that the macroscopic deformations are uniform, which justifies the use of the first-order homogenisation. Each material point at the macroscopic scale is assumed to be linked with an RVE, which represents explicitly the nanocomposite morphology i.e. distribution and dispersion of MWCNTs. Every material point is linked here to RVEs of the same morphology, which implies global periodicity.

The deformation at each macroscopic material point, which results from the macroscopic boundary conditions, is transferred onto a relevant RVE using the macroscopic deformation gradient F_M . Hence, current position of a point on the RVE boundary is represented by the position vector in the deformed configuration x_{RVE} , and it is driven by the macroscopic deformation gradient F_M as follows:

$$x_{RVE} = F_M X_{RVE} + w, \tag{1}$$

where X_{RVE} is the position vector of the point in the undeformed configuration, while w is the fluctuation field arising from the nanocomposite morphology. Eq. (1) represents *macro-to-RVE* scale transition. Then, using $x_{RVE} = X_{RVE}^+ + u_{RVE}^-$, Eq. (1) can be expressed in terms of displacements of the RVE faces

$$u_{RVE} = (F_M - I)X_{RVE} + w, \tag{2}$$

where I stands for the second-order identity matrix. The assumed periodicity of nanocomposite morphology leads to the following periodic boundary conditions

$$u_{RVE}^+ - u_{RVE}^- = (F_M - I)(X_{RVE}^+ - X_{RVE}^-), \text{ where } w^+ = w^-, \tag{3}$$

where the subscripts '+' and '-' denote opposite faces of the RVE.

For the assumed kinematic scale transition and periodic boundary conditions, the deformation of an RVE leads to the stress response that is used to define the macroscopic Cauchy stress as

$$\sigma_M = \frac{1}{V_{RVE}} \int_{V_{RVE}} \sigma_{RVE}(x_{RVE}) dV_{RVE}, \tag{4}$$

where V_{RVE} denotes the volume of the deformed RVE. Eq. (4) represents the *RVE-to-macro* scale transition. The multiscale approach for the PET/MWCNT nanocomposites is summarised in Fig. 1, and was integrated with the commercial FE software ABAQUS using its Python scripting and user-defined material (UMAT) subroutine.

2.2. Constitutive behaviour of nanocomposite constituents

The nonlinear multiscale approach is complemented by relevant constitutive laws for the PET and MWCNTs. A version of the glass-rubber model [4] modified with an empirical representation of tension-thinning of the PET flow, and described in detail elsewhere (see [8]), was used here to capture highly nonlinear, strain-, temperature- and time-dependent behaviour of PET matrix in the nanocomposite. A unique feature of this model is its ability to capture the lock-up of viscous flow, which is manifested experimentally by the onset of strain hardening during stretching, and thus it is assumed here to be a precursor of

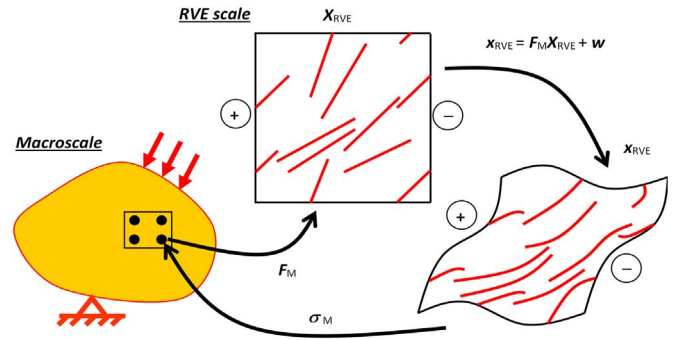


Fig. 1. Nonlinear multiscale approach for PET-MWCNT nanocomposites based on the RVE concept and first-order nonlinear homogenisation.

the stress-induced crystallisation in PET. This 3D physically-based model combines stress contributions from intermolecular interactions (*bond-stretching* part, B) and conformational changes of an entangled polymeric network (*conformational* part, C) of PET. This results in the overall Cauchy stress tensor composed of deviatoric stress components B ($\widehat{\sigma}_B$) and C ($\widehat{\sigma}_C$), and the volumetric (σ_m) stress component, as follows:

$$\sigma = \widehat{\sigma}_B + \widehat{\sigma}_C + \sigma_m I, \text{ where } \sigma_m = K \ln J \tag{5}$$

where I is the second-order identity tensor, K is the bulk modulus, and J stands for the volume ratio. Full mathematical details of the modified glass-rubber model along with a full set of parameters are described in Ref. [8] (see Section 3 and Appendix A (the model), and Appendix C (the model parameters)). The model has been implemented as a UMAT subroutine in ABAQUS.

MWCNTs were approximated here as effective nanofibres because of computational efficiency of this description compared with the discrete representation of MWCNTs. Thus, the nanotubes were modelled using the effective linear elastic and transversely isotropic properties as in [10], using relevant closed-form formulas based on the molecular mechanics approach [11], and the replacement method proposed in [12]. For that purpose, perfectly straight MWCNTs were only considered in this work, where each wall of the MWCNT was assumed to exhibit an armchair structure. Additionally, external and internal diameters were taken as 15.5 nm and 4 nm, respectively, which in combination with the interlayer distance of 0.34 nm, it provided a wall number of 18, which was then used in the calculation of effective properties of the nanotubes. The full set of elastic constants used in the current work has been reported in Table 2 in Ref. [10] – see the column related to straight (effective) fibres under tension – it was assumed that the linear elastic response of a MWCNT in tension and compression is equal.

Perfect bonding at the interface between PET and MWCNTs was assumed in this communication for computational convenience. Clearly this is a significant simplification to the real situation where non-functionalised MWCNTs (as assumed here) may be weakly bonded with the surrounding polymeric matrix *via* van der Waals interactions. A more realistic treatment of this problem would require development of an interfacial law and/or a transition layer similarly to [10,13] based on molecular mechanisms, to capture accurately polymer-nanotube interactions within the temperature range near the glass transition. Moreover, the behaviour of the polymer matrix near the nanotubes was assumed here to follow the same constitutive behaviour as the bulk polymer. Thus, the so called interphase polymer region around nanoparticles (see e.g. [14]) was not modelled in this work, due to the lack of experimental evidence.

2.3. Generation and FE meshing of RVEs

Acceptance rejection-algorithm was implemented in an in-house built Python-based script and used to populate 3D cubic RVEs with randomly distributed and randomly oriented effective nanofibres. In

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