HOMOGENIZATION METHOD FOR 2-D NANO-STRUCTURE REINFORCED POLYMER MATRIX

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Introduction

Graphene possesses an outstanding array of properties including electrical, thermal, mechanical, barrier, high surface area and others, making it an important candidate as an additive for polymers [1]. The properties of graphene based polymer nanocomposites are strongly dependent on graphene preparation techniques, method of incorporation into polymers and finally morphological control during processing. One important additive property is an increase in stiffness. A predictive model is needed to understand the increased stiffness of the reinforced matrix of these composite materials. Using a 2D image and FE representation of micrographs of polyethylene (PE) embedded with (2-D) graphite nanoplatelets obtained via melt extrusion, the mechanical properties are assessed based on computational homogenization.

Highly Structured Graphene-polymer nanocomposites

The graphene based nanocomposites properties are dependent on several of the above mention processing considerations. In particular, flow field - matrix - filler interactions are crucial for de-aggregating the particles, dispersing the filler into the matrix and ensuring the desired orientation in graphene polymer nanocomposites during processing. In the absence of major dispersion issues, Graphene/GnP particles orient in the flow direction at low shear rates (as low as 10^{-2} s^{-1}), thus enabling the creation of highly structured (thermoplastic in this case) polymer nanocomposites [2-4]. Such polymer nanocomposites exhibit anisotropic electric and thermal properties with potential applications to field grading materials [2] and heat management components [3]. A basic general requirement for nanocomposites is the mechanical reinforcing effect of the fillers. In the highly structured GnP-polymer nanocomposites, moderate improvements in young's modulus, the yield strength and tensile strength were recorded with increasing filler concentration [2-3]. Higher values were recorded along the extrusion flow direction, compared to the perpendicular direction, due to the flow-induced molecular orientation [3]. A downside of improved mechanical properties is the change from ductile to brittle fracture with increasing filler concentration [2-3].

Computational Homogenization Graphene-polymer nanocomposites

To predict and to complement the experimentally observed increased stiffness of the reinforced matrix, a model based method has been developed to assess the effective elastic representation of the graphene enhanced polymer composite based on computational homogenization. The effective elastic stiffness of the matrix can then be used for component design of e.g. CFRPs, via upscaling the matrix to the ply level response using standard technologies.

The approach presented here uses experimental through-thickness SEM images of nanocomposites to create a virtual representation of the experimental microstructure. The SEM images are analysed systematically through a series of image analysis techniques that reduce errors in flake extraction, increase repeatability and would greatly reduce the time require to analyse large sets of samples. There are five image analysis steps. First, the raw image is cropped to 50x50 µm randomly. Then Fiji's Auto Threshold is used with the Shanbhag Method [5]. Following that, the flakes shorter than 5 µm are removed. To properly represent the microscopic fluctuation using finite elements, the Feret's Diameter Method is used to approximate the remaining flakes as line elements. For the assessment of the effective elasticity, a representative volume element (RVE) of the nano-structure reinforced polymer matrix was established. In the homogenization procedure, the 2-D flakes are considered as internal 2D elements that store energy for membrane actions, cf. ref [6], whereas the PE bulk is considered isotropic elastic.

Results

Preliminary results are currently available and further results will be included in the full paper. Representative images were used for computational homogenization at graphene concentrations of 1, 3, and 5% by weight. Macroscopic stiffness properties were simulated and compared to the experimental of Gaska et al. [3] with respect to increasing volume concentrations, orientation and distribution of the graphene. These images were used to create a 2D representation of the nanocomposite material. Where the graphene flakes are represented as 1D line features with an extremely high stiffness. Currently linear representations of the flakes are used for simplicity, but the analysis can be used for curved or spline representations. The homogenized stiffness output from the computational models correlate well with the experimental stiffness values. For the three cases a single sample was analysed with this homogenization method. The resulting average error was 8% when compared to the experimental results for stiffness in the direction of extrusion. Using this method, the homogenized stiffness will be computed for each image sample and then averaged over the 5 samples. The error is expected to decrease as the number of samples analysed increases.

3%

a)   b)   c)
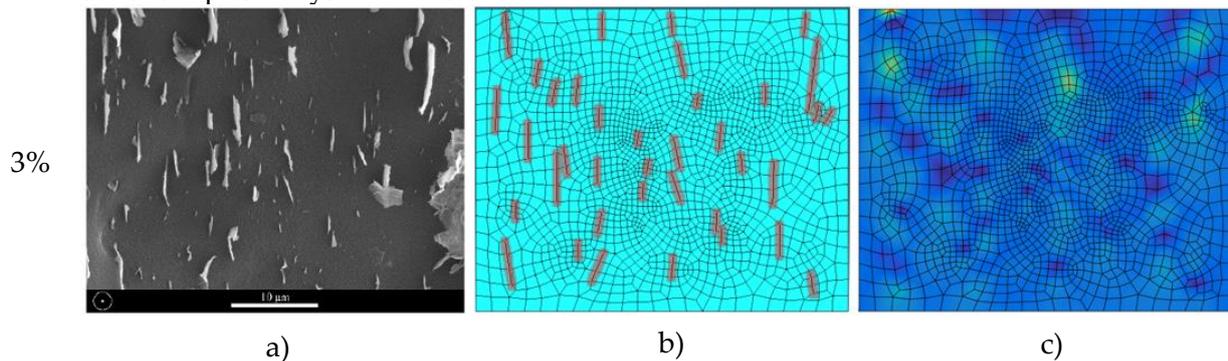
Figure 1: *Homogenization of E11 for Graphene-LDPE Nanocomposites: a) experimental image [3], b) mesh with 1D graphene representations, c) deformation in the vertical (11) direction*

Limitations of this method include correlation between the 2D and true 3D properties, image variation throughout the material, and variation of the surface normal in the samples. The current homogenization approach uses a 2D representation which artificially creates infinite plate structures (of the 1D flakes) that run out of plane. This assumption could yield an overly stiff homogenization prediction. There are some methods to alleviate this issue that will be discussed in the full paper. Image variation can have a significant impact on this method. Nano composites with significant variation in graphene particle concentration, particle size, and particle direction will require more images to develop representative stochastic homogenization results. Fractured experimental samples, such as those used in this paper, can have high and low areas along the fracture boundary. These undulations can create issues when using automated image analysis and must be considered. In addition, these fracture boundaries may tend to propagate through weaker (resin rich) regions of the nanocomposite material. This, in turn, could lead to an under prediction of stiffness from this homogenization method.

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