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Non-linear elastic moduli of Graphene sheet-reinforced polymer composites

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ABSTRACT

The non-linear elastic moduli of the Graphene sheet-reinforced polymer composite are investigated using a combined molecular mechanics theory and continuum homogenisation tools. Under uni-axial loading, the linear and non-linear constitutive equations of the Graphene sheet are derived from a Taylor series expansion in powers of strains. Based on the modified Morse potential, the elastic moduli and Poisson's ratio are obtained for the Graphene sheet leading to the derivation of the non-linear stiffness tensor. For homogenisation purpose, the strain concentration tensor is computed by the means of the irreducible decomposition of the Eshelby's tensor for an arbitrary domain. Therefore, a mathematical expression of the averaged Eshelby's tensor for a rectangular shape is obtained for the Graphene sheet. Under the Mori–Tanaka micro-mechanics scheme, the effective non-linear behaviour is predicted for various micro-parameters such as the aspect ratio and mass fractions. Numerical results highlight the effect of such micro-parameters on the anisotropic degree of the composite.

1. Introduction

Thanks to its remarkable physical and mechanical properties, Graphene has attracted extensive research investigations since its discovery in 2004 as reported by Cao (2014). Graphene is usually studied as two-dimensional structure because of its nano-scale thickness. For understanding the mechanical properties of Graphene, several attempts have been employed among which experimental measurements and theoretical developments as well as numerical modelling. The firsts i.e experiments provide the most effective way to measure the elastic modulus of Graphene. Different values of Young modulus are presented in the open literature. These values are essentially derived from the free standing indentation based on the Atomic Force Microscope (AFM). Works by Lee et al. (2008) and Frank et al. (2007) as well as Zhang and Pan (2012) should be cited. Moreover, much high elastic modulus has been estimated by Lee et al. (2012). Indeed, using a Raman Spectroscopy method they find values of 2.4 TPa and 2.0 TPa for a mono-layer and bilayer Graphene respectively. However, Cao (2014) highlights that the value of the Poisson's ratio cannot be directly measured by experiments. Therefore, theoretical and numerical studies have been developed based on the atomistic simulation at nano-scale and continuum/structural mechanics modelling. These studies deal essentially with quantum mechanics (QM) calculations for instance in Wei et al. (2009) and semi-empirical methods like thigh-biding used in Cadelano et al. (2009); Zhao et al. (2009) as well as molecular dynamics (MD) with empirical inter-atomic potentials studied by Lu et al. (2011); Lu and Huang (2009); Sakhaee-Pour (2009, 2009); Sakhaee-Pour et al. (2008); Wang and Zhang (2012); Wang (2010); Zhang et al. (2012); Zhao et al. (2009); Zhou et al. (2013a, 2013b). Under large deformations, the elastic behaviour of the Graphene sheet must be considered non-linear. This implies the existence of an energy potential that is function of the strain which can be expressed as a Taylor series in powers of strain as presented by Lee et al. (2008). Therefore, the stress-strain relationship is described by two parameters: the linear elastic modulus E and the non-linear elastic modulus D. This relationship has been used by Cadelano et al. (2009) to derive the constitutive law and all non-linear moduli for the Graphene stretching elasticity. Works by Wei et al. (2009) should be cited.

Based on the above mentioned derivations, the Graphene sheet represents an interesting reinforcement for designing multifunctional polymer composites. Graphene-based polymer composites (Ji et al. (2010)) are widely studied using micro-mechanics tools like the scheme by Mori and Tanaka (1973). However, to derive the effective properties of such composite materials, the Eshelby's tensor for the Graphene sheet accounting for its real geometrical

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Nomenclature

α_1, α_2	angle of the carbon bonds
$\Delta\alpha_1$, $\Delta\alpha_2$	angle variations of the bonds
$\Delta\theta$	angle variation for three neighbouring atoms
Δr	aariation of bonding length
ϵ	applied strain
η	aspect ratio
ν	Poisson's ratio
$\rho_{c,\,g,\,m}$	density of the composite, Graphene, matrix
σ	observed stress
σ_x, σ_y	axial stress
τ_{xy}	shear stress
$\mathbf{A}_g$	localisation tensor
$\mathbf{I}$	identity tensor
$\mathbf{L}_g$	linear stiffness tensor
$\mathbf{L}$	effective linear stiffness tensor
$\mathbf{N}_g$	non-linear stiffness tensor
$\mathbf{N}$	effective non-linear stiffness tensor
$\mathbf{S}_0$	isotropic part of the Eshelby's tensor
$\mathbf{S}_\omega$	anisotropic part of the Eshelby's tensor
θ_i	interior points angles
D_g	non-linear modulus of the Graphene sheet
E_g	linear modulus of the Graphene sheet
i_1, i_2	orthonormal basis vectors
M_g	mass fraction of the Graphene sheet
M_m	mass fraction of the matrix
p_2, q_2, p_4, q_4	complex-variables of boundary integrals
r	bond length
t	thickness of the Graphene sheet
$U_{in-plane}$	modified Morse potential
V_g	volume fraction of the Graphene sheet
V_m	volume fraction of the matrix
x, y	position vectors
z	relative position vectors
$\mathbf{i}$	unit imaginary number

morphology is less discussed and remain a challenging task. Herein, we suggest that the Graphene sheet is not an elliptical inclusion. It is therefore approximated by a rectangular shape. Relevant researches that derive the Eshelby's tensor for an arbitrary inclusion's shape are due to Rodin (1996). He overcomes the resolution of tricky integral equations due by non-uniformity of the strain within non-ellipsoidal inclusions. He therefore derives an algorithmic closed-form solutions of the Eshelby's tensors for arbitrary polygonal and polyhedral inclusions. Moreover, Nozaki and Taya (1997, 2000) highlight that the Eshelby's tensor at the centre and the averaged Eshelby's tensor over a polygonal inclusion are equal to that of a circular inclusion whatever the orientation of the inclusion. Using the irreducible decomposition of the Eshelby's tensor by Zheng et al. (2006), Zou et al. (2010) derive explicit expressions of the Eshelby's Tensor Field (ETF) and its average for a wide variety of non-elliptical inclusions. They formulate some remarks about the elliptical approximation to the average of ETF which is valid for a convex non-elliptical inclusion but becomes unacceptable for a non-convex non-elliptical inclusion. Based on the results of Zou et al. (2010) mainly the averaged Eshelby's tensor, Klusemann et al. (2012) has investigated the effective responses of composites consisting of non-elliptical shape in the context of several homogenisation methods.

The goal of this work is to consider a rectangular inclusion shape for deriving the non-linear elastic effective properties the Graphene sheet-reinforced polymer composite. For such a purpose, the Graphene sheet is considered undergoing non-linear deformations. Therefore, a Taylor series expansion combined with the non-linear stress- strain relationship used in Lee et al. (2008), establishes the expressions of the second order linear elastic and third order non-linear elastic moduli. This enables the derivation of a non-linear constitutive behaviour based on the Modified Morse potential for the Graphene sheet. The irreducible decomposition of the Eshelby's tensor by Zou et al. (2010) and Klusemann et al. (2012) is combined with a rectangular aspect ratio to provide the Graphene sheet with an averaged Eshelby's tensor for homogenisation purposes.

The paper is organised as follows: Section 2 establishes the theoretical framework for deriving the non-linear elastic stiffness tensor of the Graphene sheet. In Section 3, the procedure for obtaining the Eshelby's tensor for the Graphene sheet is recalled, some numerical calculations are also presented. The Mori–Tanaka micromechanics scheme is applied in Section 4 leading to the computation of the effective moduli of the composite. Numerical results obtained for different mass fractions are presented and discussed versus the anisotropic degree of the composite.

2. Non-linear stiffness tensor of the Graphene sheet

2.1. Preliminaries on Taylor series expansion

Let us consider a real value function $g(x)$ which is n times differentiable at a real value point x_0 with n being an integer. The Taylor series expansion applied to the function $g(x)$ is given such as:

$$g(x) = \sum_{n=0}^{\infty} \frac{g^{(n)}(x_0)}{n!}(x - x_0)^n \tag{1}$$

Now consider the function $g(x)$ defined as:

$$g(x) = \sqrt{a + bx} \tag{2}$$

where a and b are real constants. The derivatives of the function $g(x)$ for a quadratic truncation are given by:

$$\begin{cases} g(x) = \sqrt{a+bx} \\ g^{'}(x) = \frac{b}{2\sqrt{a+bx}} \\ g^{''}(x) = \frac{b^2}{4(a+bx)\sqrt{a+bx}} \end{cases} \tag{3}$$

Let us $x_0 = 0$, then Eq. (3) can be rewritten as follows:

$$\begin{cases} g(0) = \sqrt{a} \\ g^{'}(0) = \frac{b}{2\sqrt{a}} \\ g^{''}(0) = \frac{b^2}{4a\sqrt{a}} \end{cases} \tag{4}$$

This finally leads to:

$$g(x) = \sqrt{a + bx} \approx \sqrt{a} + \frac{b}{2\sqrt{a}}x - \frac{b^2}{4a\sqrt{a}}x^2 \tag{5}$$

Eq. (5) will be used in Section 2.2 to derive the non-linear stiffness tensor for the Graphene sheet.

2.2. Theoretical framework

For the Graphene sheet, the experimental force-deformation relation can be expressed as a phenomenological non-linear scalar relation between the applied strain ϵ and the observed stress σ:

$$\sigma = E\epsilon + D\epsilon^2 \tag{6}$$

where E denotes the Young's modulus. It is determined from components of the second-order fourth rank stiffness tensor. D stands for the non-linear (third order) elastic modulus. It is determined from components of both the second-order fourth rank stiffness