Eshelby continuum modeling of CNT-reinforced composites in free vibrations

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SUMMARY. The present work is a first step towards a refined modeling and simulation of the dissipative phenomena characterizing carbon nanotube composites. In the past decade, nano-structured materials have gained significant importance from an engineering point of view for the wide range of applications that require high levels of structural performance and multifunctionality. We propose a numerical investigation of the vibratory behavior of a suitable equivalent continuum model within a linearized elastodynamic context. The continuum model is based on the homogenization procedure of Mori and Tanaka which derives the elastic properties of the composite material by means of the Eshelby theory for elastic inclusions embedded into an elastic hosting material. Within this framework, we study the modal properties of the equivalent continuum in terms of vibration modes and frequencies, which clearly highlight the enhanced elastic properties due to the presence of CNT inclusions. We also pause to elaborate on the homogenization procedure by itself since several ways can be explored to obtain the elastic tensor of the equivalent continuum. While such approaches are completely equivalent in providing the linear elastic response, they may suggest different ways of setting up a nonlinear procedure that describes effectively the internal interfacial dissipation.

1 INTRODUCTION

Carbon nanotube-reinforced composites exhibit significant dissipative phenomena which are strictly related to the processes of interfacial damping between nanotubes and hosting matrix. The real physical process develops in a highly complex fashion and leads to a wide scattering of the mechanical performance of the nano-composites. The interfacial areas between nanotubes and polymeric matrices turn out to be order-of-magnitude greater than those of traditional composites. Free and forced vibration tests were conducted in [1] on samples in the form of cantilevered beams to characterize the natural frequencies and the modal damping ratios. They observed that the enhancement in damping ratio (up to 700% increase for a multi-walled nanotube–epoxy beam as compared to the plain epoxy beam) is more important than enhancement in stiffness by using carbon nanotube reinforcement.

Besides the aforementioned works, a few studies have addressed some aspects of the vibratory behavior of carbon nanotubes and their composites. Among these aspects, it is worth mentioning modeling approaches and simulation of vibrating nanotubes, studies of nanomechanical resonators and oscillators, the use of vibration measurements to characterize the nanotube mechanical properties, nanotube augmentation of dynamic structural properties of composites, vibrations of nanotube-based sensors and actuators [2].

Nonetheless, even in the context of simplified models according to which the dissipation takes place only along the axial direction of the nanotubes (see, for instance, [3]), a suitable equivalent continuum representation needs to be validated through different and complementary approaches, unless

one aims to rely on highly sophisticated multiscale computations which are hard to be handled and very costly. Most of the equivalent-continuum approaches are based on the Eshelby theory [4] and its generalizations to heterogeneous bodies made of a hosting material with embedded inhomogeneities (fibers, particles, carbon nanotubes, voids, and so on). Among various methods that have attempted to generalize the Eshelby theory, we employ one of the most widely used approaches, namely, that due to Mori and Tanaka [5]. From a physical and mathematical point of view, the interfacial conditions between hosting and embedded materials play the most crucial role [6]; these conditions can be treated in several ways within the homogenization procedure. An attempt of making order out of the multitude of seemingly different approaches can be found in Benveniste [7].

We hence start from considering the results presented in [7] with the aim of highlighting some general aspects and laying down alternative procedures to determine the equivalent elastic properties of the composite materials. This paves the way towards a suitable framework for improving the model in view of future works, by directly accounting for the nonlinearities.

Along these lines, we perform some numerical tests within the context of linearized elasticity, by comparing the proposed model with both numerical and experimental results from the literature. We show that the theoretically obtained bounds on the elastic properties prove to be very close to those measured through careful experiments reported in the literature. In agreement with previous theoretical studies, reinforcement with carbon nanotubes is clearly capable of enhancing the longitudinal Young's modulus of nano-composites. The extent of enhancement is a function of alignment, volume fraction of nanotubes, and the type of matrix material. The improvement of mechanical performance, relative to the pure uniform matrix, achieves a maximum when the carbon nanotubes are uniformly aligned with the loading direction and the difference in the elastic modulus of the matrix and the carbon nanotubes is the highest. These fundamental elastic properties are shown to reflect themselves into the modal vibrational signatures of the nano-composites.

2 EQUIVALENT ELASTIC MODEL

The equivalent continuum model considers a low volume-fraction dispersion of cylindrical inhomogeneities, representative of the carbon nanotubes, embedded into a linearly elastic isotropic matrix. There are different ways of constructing such a model, as shown in several works dealing with the general problems of equivalent continuum characterization of a composite material where the inhomogeneities, treated as inclusions, can be representative of fibers or particles or defects [6]. Broadly speaking, two approaches may be distinguished. One approach formulates the configurational forces acting at the interface between the two materials. The other approach, widely used in computational schemes, carries out directly a continuum (macroscopic) elastic characterization on the basis of suitably assumed stress and deformation patterns. The second approach can be *direct* or be grounded on energy considerations [7].

The nano-structured composite material is made of two domains denoted \mathcal{B}_{M} (hosting matrix) and \mathcal{B}_{C} (carbon nanotubes), such that $\mathcal{B} = \mathcal{B}_{M} \cup \mathcal{B}_{C}$ is the reference configuration of the composite material. A representative quantity of the relative sizes of the two domains is the volume fraction $n_{C} = V_{C}/V$, V_{C} and V being the volumes of the carbon nanotubes and of the overall composite material, respectively.

The proposed model is framed within the Eshelby theory for elastic inclusions. The original theory [4] is restricted to one single inclusion in a semi-infinite elastic, homogeneous and isotropic medium. The theory, generalized by Mori and Tanaka [5], allows to extend the original approach to the practical case of multiple inhomogeneities and a finite domain for the composite material [8]. In both cases there is an apparently similar but different definition of the Eshelby tensor, which depends

on how the stress distribution is described in the hosting matrix material caused by the eigenstrain originated through the inclusion (see [9] for more details). When the equivalent elastic properties are considered to be independent of such eigenstrain field, the most straightforward way to account for the presence of the inhomogeneities is to consider averaged strains and stresses.

Within this framework, let $L_{\rm M}$ and $L_{\rm c}$ be the elastic tensor of the matrix and of the carbon nanotubes, respectively. The constructed theory leads to a macroscopic equivalent elastic constitutive equation for the composite material in the form

$$\hat{T}(E) = L : E(x), \quad x \in \mathcal{B}$$
(1)

where the stress tensor T and the infinitesimal strain tensor E have to be interpreted as the tensorial quantities describing the equivalent elastic continuum. The Gibbs notation is adopted throughout this manuscript; (:) indicates the standard inner product between tensors, and (·) will henceforth denote the standard dot product in Euclidean space \mathbb{E}^3 .

We employ either Voigt's uniform strain assumption ($E = E^0$ with E^0 =uniform) or Reuss's uniform stress ($T = T^0$ with T^0 =uniform) depending on the type of chosen approach, the stiffness or the flexibility approach. The main ansatz for the development of the equivalent continuum theory are the following:

$$\boldsymbol{E} = n_{\rm M} \boldsymbol{E}_{\rm M} + n_{\rm C} \langle \boldsymbol{E}_{\rm C} \rangle = \boldsymbol{E}^0 \tag{2a}$$

$$\boldsymbol{T} = n_{\rm M} \boldsymbol{T}_{\rm M} + n_{\rm C} \langle \boldsymbol{T}_{\rm C} \rangle = \boldsymbol{T}^0 \tag{2b}$$

where (n_M, n_c) denote the volume fractions of the two phases; E_c and T_c are the orientation-dependent average strain and stress tensors, respectively, in a typical individual inclusion, and the brackets denote an average over all possible orientations of the inhomogeneities; E_M and T_M are the average quantities in the matrix; E and T denote the overall average strain and stress tensors in the composite material.

Together with assumption (2), we express the linearly elastic constitutive laws regarding the two materials separately as:

$$\boldsymbol{T}_{\mathrm{M}} = \boldsymbol{L}_{\mathrm{M}} : \boldsymbol{E}_{\mathrm{M}}$$
(3a)

$$\boldsymbol{T}_{\mathrm{C}} = \boldsymbol{L}_{\mathrm{C}} : \boldsymbol{E}_{\mathrm{C}} \tag{3b}$$

The Eshelby equivalence allows us to relate the stress in the matrix with the stress in the inclusion. This is possible by introducing the eigenstrain E^* acting in the inclusion:

$$\boldsymbol{T}_{\mathrm{C}} = \boldsymbol{L}_{\mathrm{M}} : (\boldsymbol{E}_{\mathrm{C}} - \boldsymbol{E}^{*})$$
(4a)

This equation states that, to within a suitable eigen-strain in the inclusion, the same constitutive equation can be used to express the stresses in the two materials. In the same way, in terms of the strains in the two materials, we can consider E_c equivalent to E_M to within a perturbation strain \tilde{E}_c , and thus rewrite (4a) as

$$\boldsymbol{T}_{\mathrm{C}} = \boldsymbol{T}_{\mathrm{M}} + \boldsymbol{L}_{\mathrm{M}} : (\boldsymbol{\tilde{E}}_{\mathrm{C}} - \boldsymbol{E}^{*})$$
(4b)

Sophisticated methods can be conceived depending on how strong the influence of the material phases is considered in terms of the inhomogeneity volume fraction with respect to the volume matrix (for a deeper insight see [10]). The Mori-Tanaka method moves along these lines [5] aiming at a proper account of higher volume fractions of interacting particles.

The elastic constitutive law of the equivalent continuum is obtained starting from a few assumptions regarding the relationships between the average strain and stress tensors. These relationships are given by (2) and by the following equation:

$$\boldsymbol{L}_{\mathrm{M}}: (\boldsymbol{E}_{\mathrm{C}} - \boldsymbol{E}^{*}) = \boldsymbol{L}_{\mathrm{C}}: \boldsymbol{E}_{\mathrm{C}}$$
⁽⁵⁾

obtained by combining (3b) and (4a). In particular, by recasting (3) and (4), the strain fields can be expressed as perturbations of the average strain according to

$$\boldsymbol{E}_{\mathrm{M}} = \boldsymbol{E} + \tilde{\boldsymbol{E}}_{\mathrm{M}} \tag{6a}$$

$$\boldsymbol{E}_{\mathrm{C}} = \boldsymbol{E}_{\mathrm{M}} + \tilde{\boldsymbol{E}}_{\mathrm{C}} \tag{6b}$$

$$\tilde{\boldsymbol{E}}_{\rm C} = \boldsymbol{S} : \boldsymbol{E}^* \tag{6c}$$

where \tilde{E}_{M} denotes the average perturbed strain in the matrix due to the presence of the inclusions while \tilde{E}_{C} represents the average perturbed strain in the inclusion with respect to the matrix; *S* is the Eshelby tensor which relates the strain in the matrix to the eigen-strain in the inclusion.

There are different ways of condensing out the stresses and strains to obtain (1). For sake of simplicity, we tacitly avoid to indicate the effect of averaging (indicated by brackets) among all possible CNT orientations. Therefore, following Benveniste [7], we first express the relation

$$E_{\rm M} = B : E_{\rm C}, \quad \text{with } B := I + S : L_{\rm M}^{-1} : (L_{\rm C} - L_{\rm M})$$
 (7)

by arranging (5) as

once the ordered sequence of (6c) and (6b), respectively, is used. By exploiting the known Hill's condition [12] which states that the equivalent elastic tensor is

$$\boldsymbol{L} = n_{\rm M} \boldsymbol{L}_{\rm M} : \boldsymbol{A}_{\rm M} + n_{\rm C} \boldsymbol{L}_{\rm C} : \boldsymbol{A}_{\rm C} , \quad \text{when} \begin{cases} \boldsymbol{I} = n_{\rm M} \boldsymbol{A}_{\rm M} + n_{\rm C} \boldsymbol{A}_{\rm C} \\ \boldsymbol{E}_{\rm M} = \boldsymbol{A}_{\rm M} : \boldsymbol{E} \\ \boldsymbol{E}_{\rm C} = \boldsymbol{A}_{\rm C} : \boldsymbol{E} \end{cases}$$
(9)

we can rewrite the latter, by using (7) that regulates the relationship between E_c and E_M , and by finding a tensor A that relates E to E_c . This is done arranging (2a) by (7) as

We thus obtain

$$\boldsymbol{A} = \left(\boldsymbol{n}_{\mathrm{M}}\boldsymbol{I} + \boldsymbol{n}_{\mathrm{C}}\boldsymbol{B}^{-1}\right)^{-1} : \boldsymbol{B}^{-1}$$
(11a)

with \boldsymbol{B} defined in (7). Consequently,

$$\boldsymbol{L} = \boldsymbol{L}_{\mathrm{M}} + \boldsymbol{n}_{\mathrm{C}}(\boldsymbol{L}_{\mathrm{C}} - \boldsymbol{L}_{\mathrm{M}}) : \boldsymbol{A}$$
(11b)

Mori and Tanaka [5] used the same assumptions, but with a different sequence of intermediate steps to obtain the equivalent elastic tensor L. They first posed the correspondence between $E_{\rm M}$ and E_* by expressing (5) as

$$L_{\rm M} : [E_{\rm M} + (S - I) : E^*] = L_{\rm C} : (E_{\rm M} + S : E^*)$$

$$\downarrow \qquad (12)$$

$$(L_{\rm C} - L_{\rm M}) : E_{\rm M} = -[L_{\rm M} + (L_{\rm C} - L_{\rm M}) : S] : E^*$$

where use of Eqs. (6) has been made. Secondly, from (2a) we can obtain the relationship between \tilde{E}_{M} and E^{*} , by using (6):

$$\boldsymbol{E} = n_{\rm M}(\boldsymbol{E} + \tilde{\boldsymbol{E}}_{\rm M}) + n_{\rm C}(\boldsymbol{E} + \tilde{\boldsymbol{E}}_{\rm M} + \boldsymbol{S} : \boldsymbol{E}^*)$$
(13)

from which

$$\tilde{\boldsymbol{E}}_{\mathrm{M}} = -\boldsymbol{n}_{\mathrm{C}}\boldsymbol{S}: \boldsymbol{E}^{*}$$
⁽¹⁴⁾

We can then rewrite (12) as

by using (14). Finally, we can express the equivalent stress tensor as

$$L: E = n_{M}L_{M}: E_{M} + n_{C}L_{C}: E_{C}$$

= $n_{M}L_{M}: (E + \tilde{E}_{M}) + n_{C}L_{M}: (E + \tilde{E}_{M} + (S - I): E^{*})$
= $L_{M}: (E - n_{C}S: E^{*} + n_{C}(S - I): E^{*}) \equiv L_{M}: (E - n_{C}E^{*})$
= $L_{M}: \{I + n_{C}[L_{M} + n_{M}(L_{C} - L_{M}): S]^{-1}: (L_{C} - L_{M})\}: E$

by using the following sequence of equations: (5), (6), (14), and (15). The latter yields the expression of the equivalent elastic tensor due to Mori and Tanaka in the form

$$L = L_{\rm M} : [I + n_{\rm C} [L_{\rm M} + n_{\rm M} (L_{\rm C} - L_{\rm M}) : S]^{-1} : (L_{\rm C} - L_{\rm M})]$$
(16)

The last expression of L can be differently employed in finite element frameworks. With respect to (11), Eq. (16) seems to be more flexibly employed in nonlinear formulations, where the nonlinearity is expected to affect the modeling of S and may possibly require re-assembling of the Jacobian matrix of the problem. However, for the linear elasto-dynamic case we have studied, the two expressions lead to the same stiffness coefficients.

3 NUMERICAL TESTS

3.1 Experimental validation

The approach illustrated in the previous section can be used to predict the effective elastic modulus of carbon nanotube-reinforced nano-composites. To make these comparisons meaningful, it is essential that the synthesis of the nano-composites be carried out experimentally with precise control of the microstructure of the composites. In particular, special attention has to be devoted to the following aspects: homogeneous dispersion, efficient interfacial stress transfer, and good alignment. One of the few works where we found these requirements fulfilled is [13]. The single-walled carbon nanotubes (SWNTs), produced by catalytic chemical vapor deposition, had diameters of about 1-2 nm and lengths of about 5-15 μ m. Pristine SWNTs, with a weight fraction equal to 0.5 %, were dispersed into epoxy. In different tests, the nano-composites were fabricated with dispersions of functionalized SWNTs with various generations of grafted dendrimers. We did not consider the case of functionalized SWNTs since the functionalization typically alters the mechanical properties of the carbon nanotubes; however, these properties are not available. In all cases, SWNTs were uniformly dispersed and aligned by means of a reactive spinning process. The alignment, as we shall see, plays a crucial role in the enhancement of the elastic modulus of the resulting composite as well as in the enhancement of the tensile strength. The composites were characterized by optical and electron microscopy and by conducting tensile tests. The tensile tests delivered a Young modulus of 3.47 GPa.

To carry out the calculation of the effective elastic modulus within our theoretical setting, we had to compute the CNT volume fraction n_c , one of the prescribed data of our computational framework. We assumed the specific weights of $1.20 \cdot 10^3$ Kg/m³ for the epoxy resin and $7.85/6 \cdot 10^3$ Kg/m³ for the CNTs, respectively. The resulting CNT volume fraction is $n_c = 0.46\%$. In agreement with the experimentally measured characteristics, in the numerical computations we employed the following mechanical parameters: $(E, v, \lambda) = (2.35 \text{ GPa}, 0.4, 3.357 \text{ GPa})$ for the epoxy resin and (E, v) = (970 GPa, 0.28) for the CNT. The thus calculated elastic modulus is 3.27 GPa which turns out to be only 5 % lower than the experimentally measured value.

3.2 Vibrational modal properties

In order to move towards modeling and simulation of the internal dissipation mechanisms in CNT composites, we have focused on the modal vibrational properties of the nano-composites as a preliminary step of our work. We have hence conducted an eigenvalue analysis varying the CNTs alignment considering the specimen of Figure 1.



Figure 1: Variation of the CNT alignment.

The test was numerically carried out using the following mechanical parameters: $(E, \nu) = (970 \text{ GPa}, 0.28)$

mode	1	2	3	4	5	6	7	8	9	10
matrix	1.07	2.41	6.16	6.45	7.92	8.63	14.66	15.73	17.18	17.19
$\phi = 0$	0.30%	1.27%	3.32%	1.50%	11.08%	48.76%	9.66%	8.81%	1.57%	7.18%
$\pi/12$	2.12%	4.83%	3.86%	6.06%	12.03%	44.13%	9.23%	8.67%	6.46%	12.83%
$\pi/6$	10.74%	11.17%	6.43%	17.50%	10.74%	36.22%	10.38%	3.68%	8.33%	14.61%
$\pi/4$	32.97%	16.36%	14.52%	28.51%	7.98%	36.11%	5.92%	14.17%	14.27%	18.51%
$\pi/3$	60.06%	22.87%	27.22%	27.46%	11.89%	42.94%	8.21%	26.42%	23.09%	31.08%
$5\pi/12$	81.83%	26.14%	28.54%	25.72%	31.51%	45.55%	14.55%	20.77%	36.60%	59.14%
$\pi/2$	92.03%	25.88%	27.19%	25.80%	44.31%	43.54%	18.28%	17.00%	37.98%	52.99%

for the CNTs and $(E, v, \lambda) = (3.3 \text{ GPa}, 0.4, 4.7 \text{ GPa})$ for the epoxy resin, respectively, which thus represent the hosting matrix.

Table 1: Variation of the frequencies of the lowest ten vibration modes with the CNTs orientation when $n_c = 1\%$. The variations are given in % increments with respect to the frequencies of the all-matrix material.

The frequencies of the lowest ten modes of the specimen, made of pure matrix or made of the composite CNT-reinforced material, are compared in Table 1. Here, the ratios of the frequencies with respect to the lowest frequency of the equivalent homogeneous, isotropic, all-matrix, cantilevered plate are reported. When we take the specimen made of the pure matrix material, this ratio becomes 1 in the first mode. Moreover, we conveniently normalized the mode shapes according to $\int_{\mathcal{B}} \psi_k \cdot \psi_k dV = 1, \forall k = 1, 2, ...,$ where $\psi_k(\mathbf{x})$ denotes the *k*th mode shape of the plate. Note finally that, unless otherwise stated, the volume fraction n_c is assumed to be 1% in most of the reported calculations.

The comparison in terms of the frequencies for the case of epoxy clearly emphasizes the fact that the nano-structured composite material exhibits improved mechanical properties. In particular, for the specific conducted tests, the lowest frequency of the specimen with longitudinally oriented CNTs ($\phi = \pi/2$) is about 100 % that of the specimen with the homogeneous matrix material.

To investigate in depth the fundamental properties of the vibration modes of nano-structured composites, we observed the spatial patterns of the modal stored-energy and its distortional part in each mode. In particular, according to [14], the distortional energy interpretation for the von Mises yield criterion can also be used effectively to construct yield criterions for particle-reinforced composites that bear isotropic properties.

The analysis of the contour plots of the von Mises stress, associated with modes possessing close frequencies in epoxy-based CNT-reinforced composites, shows in Figure 2 that, in the optimal case with longitudinally aligned CNTs, the third mode would vibrate with the same frequency as that of the fifth mode of the all-matrix plate. The von Mises stress distribution would not be very different in the CNT-reinforced case although it exhibits higher values due to the higher stiffness.

4 CONCLUSIONS

In this work, we have investigated the elastic properties of carbon nanotube-reinforced composites and the linear vibrations signatures employing an equivalent continuum formulation based on the Eshelby-Mori-Tanaka approach. This approach, also known as the Equivalent Inclusion-Average Stress method, consists of the combination of the equivalent inclusion idea of Eshelby with the concept of average stress and strain in the isotropic elastic matrix of Mori and Tanaka. The elastic inclusions are assumed to be bonded to the matrix through a perfect interface. The amount of inclusions is naturally prescribed in terms of their volume fraction.

The theoretically obtained bounds on the elastic properties proved to be very close to those measured through careful experiments reported in the literature. In agreement with previous theo-

retical studies, reinforcement with carbon nanotubes is clearly capable of enhancing the longitudinal Young's modulus of nano-composites. The extent of enhancement is a function of alignment, volume fraction of nanotubes, and the type of matrix material. The improvement of performance, relative to the case of pure uniform matrix, achieves a maximum when the carbon nanotubes are uniformly aligned with the loading direction and the difference in the elastic modulus of the matrix and the carbon nanotubes is the highest.



Figure 2: Slices of the contour plots of the von Mises stress associated with modes possessing close frequencies in an epoxy-based (Epx) composite plate: (a) 5th mode of pure matrix, (b) 4th mode of CNT-reinforced with $\phi = \pi/6$, (c) 4th mode of CNT-reinforced with $\phi = \pi/4$, and (d) 3rd mode of CNT-reinforced with $\phi = \pi/2$. For the sake of clarity, the ranges in plots (b) and (c) were bounded with respect to the actual ranges of [1e⁻³, 1.153] and [5.5e⁻⁴, 0.5], respectively.

The in-depth investigations into the elastic modal vibration properties, confined to the analysis of the lowest ten modes of cantilevered composite plates, have shown that the integration of carbon nanotubes into the hosting material is indeed a powerful and useful mechanism for tuning the vibration properties of the composite plates. We obtained, in the case vulcanized rubber as hosting material, predictions of frequency increases in the lowest mode of the order of 500% without altering the mass density of the material. In particular, nano-composites plates can be designed so as to respond with a dominant selected mode to certain known excitations. Further, the selected mode can be such that the likely probability of failure may be minimized since a certain degree of control

is possible over the way the distortional modal energy is distributed within the vibrational energy patterns.

This study is the first step towards a more complex nonlinear equivalent continuum modeling capable of describing the internal interfacial dissipation at the carbon nanotubes-matrix interfaces. Towards this end, we analyzed and compared different ways of obtaining the equivalent elastic tensor of the composite material. Although the different approaches turn out to be substantially equivalent in linear elasticity, alternative procedures to the one here adopted (Benveniste [7]) are envisioned to likely lead to models of the dissipative phenomena with more efficient computational features.

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