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Damage model of carbon nanotubes debonding in nanocomposites

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ABSTRACT

The progressive interfacial debonding between aligned carbon nanotubes and the hosting matrix of a nanocomposite in the direction normal to the CNTs axis is described by means of an equivalent constitutive model with evolutionary damage. The Eshelby–Mori–Tanaka theory is used to describe the macroscopic mechanical response of the nanocomposite for a given volume fraction of the different phases (i.e., perfectly bonded and fully debonded CNTs). The novelty of this work is the proposition of a new thermodynamically consistent phase flow law that describes the cumulative progression of debonding derived from the Weibull statistics. Monotonic and cyclic uniform strain histories are considered to investigate the nanocomposite response features such as the stress–strain softening hysteretic cycles, the progressive degradation of the elastic moduli, and the dissipated energy.

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1. Introduction

Nanostructured materials based on carbon nanotubes are used in a variety of applications, ranging from nanocomposites with enhanced mechanical performance or multifunctional capabilities, composites for energy storage, etc. The rapid and continuous growth of advanced aerospace, automotive, and military applications requires use of these special materials with high-performance characteristics that enable structures to achieve challenging target properties. One of the open problems is an accurate mechanical modeling of such multi-phase nanostructured materials that can account for nonlinear phenomena such as the cumulative debonding between the matrix and the CNTs up to the ultimate state of failure. This is important for example toward the assessment of the fatigue life of materials or devices based on dispersion of CNTs as mechanical reinforcing agents or enhanced electrodes such as is the case for supercapacitors for energy storage.

In the literature, various theories have been developed to describe the progressive debonding of composites embedding different types of inclusions. Recently, in the context of nonlinear finite element modeling supported by experimental data [1,2], the investigation of the molecular interaction between polymer matrices and CNTs highlighted the influence of local properties of the CNTs, such as chirality and aspect ratio, on the constitutive softening response under tensile conditions in terms of Young's modulus. Such results reveal the need for a continuum modeling of the damage behavior. However, it is worth mentioning that the conducted tests do not consider pure hydrostatic stress states which drive the debonding phenomenon, which is the focus of the present work.

In the past, one of the first micromechanical theories was developed in [3], in order to examine the debonding process of a brittle matrix composite containing aligned oblate inclusions under a high triaxial state of stress in which the debonding rate was assumed to be governed by a Weibull probability function in terms of the hydrostatic stress in the inclusions. In [4,5], the macroscopic constitutive relationship of particulate-reinforced viscoelastic composite materials was investigated by employing Eshelby's equivalent inclusion method combined with the Mori–Tanaka averaging scheme. It was ascertained that the macroscopic strain rate, the particle-size dispersity, the relaxation time of the matrix, and the interface adhesive strength are key mechanical factors for this kind of particulate-reinforced composites where the microvoids nucleation and growth are the driving damage process.

Interfacial debonding in polymer/nanoparticle composites was studied in [6] by means of Eshelby's equivalent inclusion method combined with an energy-derived debonding criterion stating a relationship between the interfacial adhesion strength and the work of adhesion of the components.

One of the first approaches to the study of the overall elastoplastic response of a two-phase, ductile composite containing aligned oblate inclusions is described in [7]. The same kind of ductile matrix composites was also investigated by means of micromechanical damage models to predict the overall elastoplastic behavior and damage evolution. For example, three-phase ductile matrix composites were studied in [8] by introducing an effective yield criterion based on the ensemble-volume averaging process together with an associative plastic flow rule and a hardening

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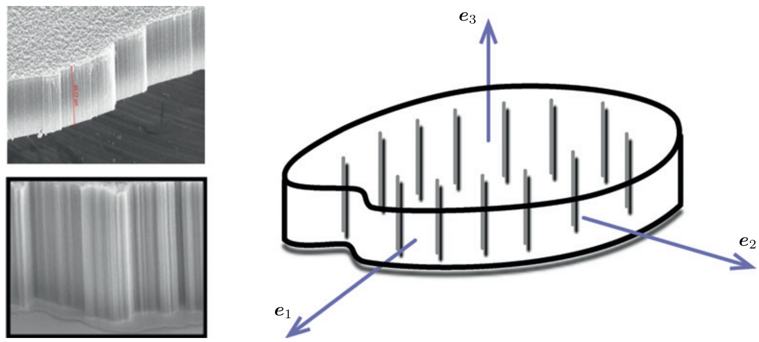

Fig. 1. (left) Forests of carbon nanotubes embedded in a supercapacitor [19], (right) the reference configuration $\mathcal{B}^o$ of the nanocomposite material with the hosting matrix and the CNTs.

law. A key aspect of this work is the evolutionary interfacial particle debonding model in accordance with the Weibull statistical function to describe the varying probability of complete debonding. The theory was applied in the context of composites with different types of inclusions and considering uniaxial and plane loading conditions [9,10].

A similar micromechanical framework employing Eshelby's tensor was proposed in [11,12] to predict the effective response of particulate composites with spherical inclusions suffering interface weakening. The damage model was based on Weibull's probabilistic function to characterize the probability of evolution of weakened interface between the inclusions and the matrix under uni-, bi- and tri-axial tensile loading conditions. Some comparisons with experimental data were reported in [13]. Particulate composites were also studied in [14] by a thermodynamical formulation combining Anderson's and Yilmizer's models accounting for the filler-matrix debonding process in a variety of concentration scenarios. A simple analytical model accounting for the CNTs waviness was combined in [15] with a micro-mechanical approach to deal with interfacial debonding and degradation of the CNTs reinforcing efficiency in nanocomposites.

In this work, we propose a new dynamical formulation for the macroscopic response of carbon nanotube composites accounting for the cumulative debonding of CNTs due to progressive weakening of the interface. The novelty of the present contribution lies in the proposition of a new thermodynamically consistent phase flow law for the evolution of the volume fraction of debonded cylindrical inclusions (here CNTs) in a rate form amenable to a full dynamical formulation which enables parametric studies and fatigue life assessment. This flow law is the combination of the Weibull statistics and a law giving the rate of the effective stress measure which drives the debonding progression. A number of key features of the debonding phenomenology and its consequences on the dynamic response are discussed both in the context of a two- and three-dimensional setting. The achieved dynamical formulation allows general investigations to be carried out into the effects of interfacial damage on various aspects of the mechanical performance of multi-phase composites, including the dynamic response features (i.e., frequencies and mode shapes, frequency–response functions, etc.) and fatigue life assessments. Some of the initial steps and preliminary results of this work were anticipated in [16–18].

2. Problem formulation

The composite material here investigated is a multi-phase Cauchy continuum whereby the phases evolve according to a set of criteria. The material occupies a region of $\mathbb{E}^3$ denoted by $\mathcal{B}^o$ in its reference state. The material points of $\mathcal{B}^o$ are described by the position vector $\boldsymbol{x}$. Here boldface italic small caps letters indicate vectors of $\mathbb{E}^3$, boldface italic capital letters indicate second-order tensors while blackboard bold capital letters denote fourth-order tensors. Gibbs' notation is adopted according to which the application of tensor $\boldsymbol{A}$ on vector $\boldsymbol{u}$ is denoted by $\boldsymbol{A} \cdot \boldsymbol{u}$ and the tensor product of vectors $\boldsymbol{u}$ and $\boldsymbol{v}$ is denoted by $\boldsymbol{uv}$ instead of the more conventional $\boldsymbol{u} \otimes \boldsymbol{v}$. The inner product of fourth-order tensor $\mathbb{A}$ with second-order tensor $\boldsymbol{B}$ is denoted by $\mathbb{A} : \boldsymbol{B}$ while the product between fourth-order tensors $\mathbb{A}$ and $\mathbb{B}$ is indicated by $\mathbb{A} \bullet \mathbb{B}$. The inner product between second-order tensors $\boldsymbol{A}$ and $\boldsymbol{B}$ is thus denoted by $\boldsymbol{A} : \boldsymbol{B}$.

The material in its pristine state is made of two phases, namely, the hosting matrix treated as an isotropic medium and the small volume-fraction phase of carbon nanotubes treated as transversely isotropic elastic inclusions [17]. In most of the applications, the CNTs are fabricated in the form of aligned forests immersed in a hosting material (see Fig. 1 left), situation that allows to model the inclusions in a way fairly simpler than the case of randomly oriented CNTs.

In this state, the CNTs are perfectly bonded to the hosting matrix and their volume fraction is denoted by $\phi_1(\boldsymbol{x}, t)$. During a loading history, debonding phenomena occur for various reasons such as (i) onset of the ultimate interface strength, (ii) progressive weakening of the interface due to fatigue over several stress cycles, (iii) electrochemical degradation of the interface. The volume fraction of debonded CNTs described by $\phi_2(\boldsymbol{x}, t)$ progressively increases from zero, with the consequence that the problem during the debonding phase becomes that of a three-phase material, in which the hosting matrix exhibits a volume fraction equal to $1 - \phi_1 - \phi_2$ (see Fig. 2). Of course, the volume fraction of perfectly bonded CNTs at the initial time t^o is prescribed as $\phi_1(t^o) = \phi_1^o$ and the upper bound of the fully debonded CNTs is $\phi_2 = \phi_1^o$.

The main objective of the present formulation is to describe the evolving damage-type behavior of the composite material due to debonding between CNTs and hosting matrix. While it is true that localized large strains may occur at the interfaces, the description

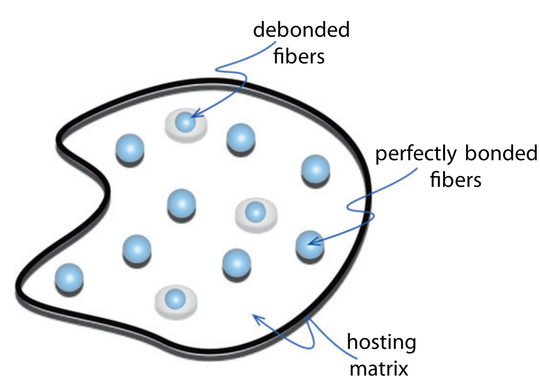

Fig. 2. The current configuration $\mathcal{B}$ of the material with the hosting matrix incorporating the phase of perfectly bonded CNTs and the phase of debonded CNTs.

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