Electromechanical peridynamics modeling of piezoresistive response of carbon nanotube nanocomposites

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A B S T R A C T

In this work, a coupled electromechanical peridynamics formulation is presented which is used to study the electrical and piezoresistive response of a carbon nanotube (CNT) reinforced polymer nanocomposite material. CNT nanocomposites are multiscale materials which have unique piezoresistive properties arising from mechanisms operating from the nanoscale to the macroscale. The origin of piezoresistivity in CNT nanocomposites is a nanoscale phenomenon known as electron hopping or the electrical tunneling effect which allows an electric current to flow between neighboring CNTs even when not in contact, thereby forming a conductive network. A nanoscale representative volume element of a CNT bundle is chosen, i.e. a local region of high CNT volume fraction within the polymer matrix, wherein coupled electromechanical peridynamic equations are solved to evaluate the effective electrical and piezoresistive properties. The peridynamics formulation is used to introduce electron hopping in a unique way, through electron hopping bonds which have a horizon distance and conductivity dictated by the appropriate physics operating at the nanoscale. The effective electromechanical response depends on parameters such as CNT volume fraction, properties of the polymer matrix between CNTs and applied strain which are investigated in detail. Both quasistatic and dynamic loading conditions are considered where the effective electromechanical response is found to depend on variations in the local conductivity of intertube regions.

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1. Background

1.1. Carbon nanotube reinforced polymer nanocomposites

Carbon nanotubes, well known to posses extraordinary mechanical, thermal and electrical properties are being seen as excellent candidates for use as fillers in polymer based composite materials [1]. A number of studies have confirmed the ultra – high stiffness and strength of carbon nanotubes (CNTs), with reported values of Young's moduli ranging from a few hundred GPa to as high as 3.6 TPa and tensile strengths up to 150 GPa [2–6]. CNTs possess unique electrical properties and may act as conductors or semi-conductors depending on their geometric structure, with electrical conductivity of single walled carbon nanotubes measured upwards of 1×10^4 S/m [7–11]. The addition of CNTs to a polymer medium is shown to have a marked effect on mechanical properties [12–16], but more significantly, electrical conductivity is observed to increase by many orders of magnitude with the addition of very low weight percentages of CNTs (i.e. 9–10 orders of magnitude increase with <1 wt% of CNTs) [17–22].

More important to the present work, the use of CNT reinforced polymer nanocomposites for structural health monitoring purposes is being increasingly researched for both strain sensing and damage detection [23–30]. The unique piezoresistive properties of such composites can be associated with three major mechanisms, (1) changing morphology of CNT networks at the nano and microscale due to deformation and damage [31,32], (2) electron hopping by which electrons can hop between CNTs even when not directly in contact [31,33–35] and (3) inherent piezoresistivity of CNTs in response to axial and bending deformations [32,36–38]. The primary contributor to piezoresistivity is believed to be electron hopping (also referred to as quantum tunneling or electrical tunneling) occurring between CNTs at the nanoscale, thereby allowing a high current density to flow through a CNT network in an otherwise non-conducting polymer [33,39–42]. Electron hopping refers to the phenomena when electrons in a CNT have

energies high enough to jump to a neighboring CNT, overcoming the potential barrier of the polymer in between. A key parameter for this effect is electron hopping range which is the maximum CNT separation distance at which electron hopping begins to take place between neighboring CNTs. The maximum hopping distance depends on the potential barrier of the polymer, which is often taken to be in the range of 1 eV to 5 eV [43]. There is no general consensus in literature regarding electron tunneling range and many values have been reported from 1 nm to 30 nm [18,33–35,44].

The mechanism of observable nanocomposite piezoresistivity spans several scales from nano to macro. Individual CNTs at the nanoscale agglomerate together under Van der Waals' forces forming CNT bundles. Under deformation, the changing intertube distances within a CNT bundle continuously make and break electron hopping pathways. CNT bundles are large enough that they start to become discernible at the microscale. In addition, there is often bridging between individual CNT bundles at the microscale, which is a contributing factor to the electrical percolation observed at very low weight percentages of CNTs. Dispersion at the microscale plays an extremely important role in contributing to piezoresistivity observed at the macroscale [45]. To fully understand the effect of these phenomena on effective macroscale properties, computational models need to be constructed at the meaningful scales, taking into account all the relevant physics associated with each scale of interest. In this work, a simple nanoscale representative volume element, reflective of the local arrangement of CNTs within a bundle is modeled with a non-local continuum method, specifically, peridynamics, with the aim of estimating the electromechanical response. A unique advantage of using peridynamics is that unlike in finite element based methods, the highly non-local effect of electron hopping can be taken into account within the peridynamic equations. No ad hoc techniques are required as the changing nanoscale morphology in response to the deformation directly dictates the formation and disruption of electron hopping pathways in a natural fashion. Macroscale observable quantities such as piezoresistive coefficients can be obtained by volume averaging techniques, under quasistatic and dynamic loading conditions.

1.2. Peridynamics

Peridynamics is a recently developed non-local theory of continuum mechanics [46,47] which recasts the balance laws as integral equations instead of the typical partial differential equations of classical local continuum mechanics. The peridynamics equations are strongly non-local as opposed to a weak non-locality or a strain gradient type non-locality. The strong non-locality stems from the internal force density being represented as an integral of forces between material particles as opposed to an integral of stress in a weak non-local theory [48] or as a divergence of stress in classical continuum theories. In addition, the primary kinematic field variable used in constitutive equations is the displacement as opposed to derivatives (strain or strain gradient) which removes all continuity requirements on the displacement field [49]. As such, peridynamic equations are valid even in the presence of discontinuities like cracks, hence making it straightforward to model crack initiation and growth. Peridynamics assumes that material particles within a finite volume called the horizon region interact with each other, with the interactions being expressed using either a bond based or a state based approach. The bond based theory, by employing a central force model, was developed first [46] to correlate displacements to the forces between material particles. However, the application of the central force model restricts the observed Poisson's ratio of the peridynamic material to 1/3 in 2D and 1/4 in 3D [47]. In order to model a wider range of materials

using peridynamics, the state based theory was developed which takes into account the collective deformation of all bonds within a material particle's horizon and hence improves the constitutive description of the material [47]. Regardless of the choice of peridynamic model, numerical solutions to practical problems can be obtained by the spatial and temporal discretization of the relevant peridynamic equations [50].

Peridynamics has been applied successfully to a wide range of problems such as dynamic fracture [51–53], fracture of fiber reinforced composites [54,55], impact damage of composites [56,57], transient heat conduction [58,59], thermomechanical structural analysis [60,61] and nanostructured materials [62] among others. Adaptive spatial refinement of peridynamic particles and scaling of micromodulus has been discussed in [63,64]. The significance of the horizon has been investigated in [62,65–67] emphasizing the relation between the nonlocal horizon and the length scale of the material. The work done till date in both the bond based and state based theory has established peridynamics as a viable tool for materials modeling especially in dynamic simulation of materials with evolving discontinuities. Work presented here is aimed at exploring application of peridynamics to modeling coupled electromechanical response of a composite material at the nanoscale, incorporating the relevant physics of the scale, in this case, electron hopping.

2. Computational model

2.1. Peridynamics for elasticity

In the non-local theory of peridynamics, every material particle $\boldsymbol{x}$ in the continuum is assumed to interact with every other particle $\boldsymbol{x}'$ within some region of influence called the horizon $\mathcal{H}_{\boldsymbol{x}}$ through a *peridynamic bond*. Consider two particles $\boldsymbol{x}$ and $\boldsymbol{x}'$ in the reference configuration separated by their relative distance $\xi = \boldsymbol{x}' - \boldsymbol{x}$. If $\boldsymbol{x}$ and $\boldsymbol{x}'$ are displaced by $\boldsymbol{u}$ and $\boldsymbol{u}'$ respectively, their relative displacement is given by $\eta = \boldsymbol{u}' - \boldsymbol{u}$ such that their relative position in the deformed configuration is given by $\xi + \eta$ (see Fig. 1).

The peridynamic equation of motion for a particle $\boldsymbol{x}$ at time t is given by

$$\rho \ddot{\boldsymbol{u}}(\boldsymbol{x},t) = \int_{\mathcal{H}_{\boldsymbol{x}}} \boldsymbol{f}(\boldsymbol{u}',\boldsymbol{u},\boldsymbol{x}',\boldsymbol{x},t)dV_{\boldsymbol{x}'} + \boldsymbol{b}(\boldsymbol{x},t) \tag{2.1}$$

where ρ and $\ddot{\boldsymbol{u}}$ denote the density and acceleration of the material particle $\boldsymbol{x}$, $\boldsymbol{f}$ is known as the pairwise force function (units of force per unit volume squared) of the bond between $\boldsymbol{x}$ and $\boldsymbol{x}'$ and $\boldsymbol{b}$ is the body force per unit volume at $\boldsymbol{x}$, all at time t. The net internal force
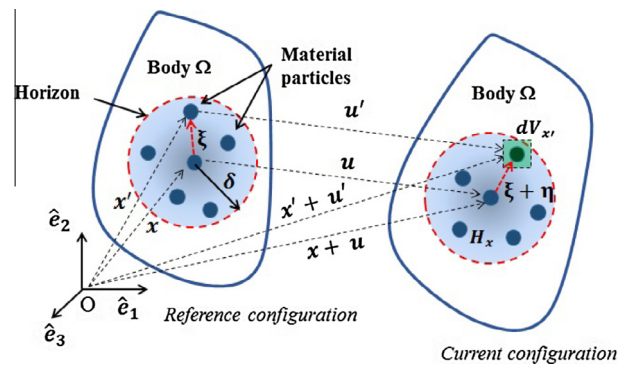
Fig. 1. Kinematics of peridynamic material particles, the blue shaded region corresponds to the horizon region of the material particles at $\boldsymbol{x}$, and $\boldsymbol{x}'$ corresponds to an interacting material particle within the horizon region of $\boldsymbol{x}$, having a volume $dV_{\boldsymbol{x}'}$ shown with the green shaded region. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)