



# Piezoresistive fiber-reinforced composites: A coupled nonlinear micromechanical–microelectrical modeling approach



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## ABSTRACT

Piezoresistive composites are materials that exhibit spatial and effective electrical resistivity changes as a result of local mechanical deformations in their constituents. These materials have a wide array of applications from non-destructive evaluation to sensor technology. We propose a new coupled nonlinear micromechanical–microelectrical modeling framework for periodic heterogeneous media. These proposed micro-models enable the prediction of the effective piezoresistive properties along with the corresponding spatial distributions of local mechanical–electrical fields, such as stress, strain, current densities, and electrical potentials. To this end, the high fidelity generalized method of cells (HFGMC), originally developed for micromechanical analysis of composites, is extended for the micro–electrical modeling in order to predict their spatial field distributions and effective electrical properties. In both cases, the local displacement vector and electrical potential are expanded using quadratic polynomials in each subvolume (subcell). The equilibrium and charge conservations are satisfied in an average volumetric fashion. In addition, the continuity and periodicity of the displacements, tractions, electrical potential, and current are satisfied at the subcell interfaces on an average basis. Next, a one way coupling is established between the nonlinear mechanical and electrical effects, whereby the mechanical deformations affect the electrical conductivity in the fiber and/or matrix constituents. Incremental and total formulations are used to arrive at the proper nonlinear solution of the governing equations. The micro–electrical HFGMC is first verified by comparing the stand-alone electrical solution predictions with the finite element method for different doubly periodic composites. Next, the coupled HFGMC is calibrated and experimentally verified in order to examine the effective piezoresistivity of different composites. These include conductive polymeric matrices doped with carbon nano-tubes or particles. One advantage of the proposed nonlinear coupled micro-models is its ability to predict the local and effective electro-mechanical behaviors of multi-phase periodic composites with different conductive phases.

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## 1. Introduction

Piezoresistance is a one-way coupled electrical–mechanical property that measures the change in electrical resistance of the material due to applied deformation (stress and strain). Unlike piezoelectric materials, piezoresistive materials (PMs) change their electrical resistance or conductivity without producing any electric potential. Different classes of PM can be manufactured and designed depending on their in situ composition of conductive constituents that form an internal network needed for crossing the percolation-threshold of conductivity. The gage factor (GF) is a PM property that characterizes the relative change in resistance divided by the applied strain. Polymeric, hydrogel, silicon and other non-conductive materials can be chemically altered or doped with

conductive fibers and particles to embed new piezoresistive behavior. There is a great potential of this class of engineered PMs for sensors and MEMS. It is not surprising to have the Noble Prize in Chemistry for the year 2000 to be on conductive polymers, awarded to Heeger, MacDiarmid and Shirakawa (Shirakawa et al., 1977).

Doping polymeric materials with multi-wall carbon nanotubes (MWCNTs) with a relatively small volume fraction (less than 2%) is a technique to engineer new PMs. The properties of this polymeric piezoresistive composite matrix (PPCM) have been extensively studied in the last decade (Loh et al., 2008; Fernberg et al., 2009; Wichmann et al., 2009; Bautista-Quijano et al., 2010; Ciselli et al., 2010; Loyola et al., 2010). These studies indicate that a relatively high GF can be achieved using MWCNT as a simple nanoparticle candidate without the need to chemically alter the polymer chains as used in conjugate conductive polymers. The experimental treatments presented in these early PPCM studies are usually

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limited to uniaxial loading with little attention to complex stress states and multiaxial loading conditions. In addition, these studies do not fully address the two sources of nonlinear mechanical behaviors, namely the nonlinear elastic deformation of the polymeric matrix followed by damage due to high volume of coalescing voids and microcracks. The nonlinear mechanical responses lead to an increase in order of magnitudes in the piezoresistive behavior as a function of the applied deformations. Doped cement-based carbon nanotubes were made to explore this PM electrical resistance with good results even at lower added relative weight of 0.05% (Han et al., 2010). Sensor type devices made from PPCM have been recently proposed due to their ease of manufacturing and the effective GF property. Doped polymeric films with MWCNT (up to 1%) have been proposed for strain measurements on the surface of a structural component (Bautista-Quijano et al., 2010). A similar approach was used to study a multi-layered and tailored carbon nanotube composite strain sensor (Loh et al., 2008).

Despite the recent intensive research in PPCMs, their piezoresistance behavior is not well understood and predictive theories remain empirical, one-dimensional and phenomenological. The relation between morphology of the constituents as a function of local spatial deformations is poorly understood. The piezoresistant effect in conducting fiber-filled composites has been studied by a continuum percolation model (Lin et al., 2010) and was proposed to study the piezoresistant behavior of conducting fiber reinforced composites. The deformation of the fiber in the form of bending and rotation were taken into account in the simulation model. Compression loading increased the conductivity and percolation threshold that yields a positive piezoresistive material coefficient. Their simulations highlighted the strong relation with microstructural deformation-based changes.

Piezoresistive behavior of carbon fiber reinforced polymeric (CFRP) composites has been previously investigated because of their widespread engineering applications and the inherent conductivity of the fiber. Longitudinal tension tests have shown increased resistivity (Todoroki and Yoshida, 2004) while others pointed out to the nonlinear resistivity due to the rapid increase in fiber breakage (Prabhakaran, 1990; Park et al., 2002). Tests for uniaxial layer of CFRP with different volumes of carbon fiber reinforcement under both monotonic and cyclic loading were conducted (e.g., Abry et al., 1999). An unsymmetrical piezoresistive matrix was found from multi-axial tests of a longitudinal CFRP layer (Todoroki et al., 2009). Nonlinear piezoresistive behavior was also found by testing bi-axial laminate and plain woven samples (Boschetti-de-Fierro et al., 2009). Electrical impedance tomography (EIT) was proposed to characterize the location and magnitude of damage in piezoresistive composites (Schueler et al., 2001). Testing biaxial CFRP composites using DC and AC currents (Abry et al., 2001) showed they are suitable for fiber breakage and matrix cracking modes, respectively.

Several models for piezoresistive composite laminates have been proposed (Prabhakaran, 1990; Schueler et al., 2001; Park et al., 2002; Xia et al., 2003). They used spring type analogy (resistances) to describe the collective electrical response. These discrete models are one-dimensional and were only successful in describing the axial change of resistance. They are also limited to a composite medium with a non-conductive matrix. Classical linear lamination theory has been coupled with electrostatic theory for the piezoresistivity analysis of laminated composites (Xiao et al., 1999). To our best knowledge, there is no prior work on nonlinear micro-mechanical-electrostatic models for piezoresistive composites.

In addition, the polymeric matrix can be doped with conductive particles, such as micro-spheres and MWCNTs. The latter have anisotropic piezoresistive behaviors both due to the polymeric matrix's inherent nonlinear mechanical response and due to damage

evolution in the matrix and fiber constituents. Piezoresistive fiber reinforced polymeric composites (PFRP) have also been examined for self health structural monitoring. The piezoresistive response of composites with zinc conductive fillers have been proposed as a uniaxial stress self-material measurement (Abyaneh and Kulkarni, 2008) with several order of magnitudes changes in resistivity as pressure reaches a critical value. The effects of different carbon fibers and reinforcement arrangements on the overall piezoresistive behavior have been studied (Boschetti-de-Fierro et al., 2009). Linear and nonlinear range was found for the GF as a function of the applied mechanical loading. The piezoresistive behavior was examined and compared for PFRP composites and concrete materials with short fibers (Chung, 1998). It was found that these PMs can be used for self-health-monitoring where the concrete composite gives excellent strain sensitivity (GF = 700) while the composite gives good matrix cracking damage detection ability. Damage mode detection in PFRP composites was also considered (Chung and Wang, 2003; Chung, 2007), such as in flexure, tension, fatigue, and impact loadings. The anisotropic distribution of the electrical current in these heterogeneous anisotropic composites was identified as a challenging issue to deal with in order to better diagnose the damage mode and magnitude in these materials and structures. It was suggested that rivets or fasteners in aerospace PFRP composite structures can be used as electrodes for measurements of piezoresistance and electrical changes in strength and fatigue (De Baere et al., 2007, 2010).

The electromechanical piezoresistive behavior was also explored (Fernberg et al., 2009) for laminated PFRP composites with longitudinal long fibers and doped epoxy matrix with MWCNT. The tests showed that CNT-doped epoxy under tension is highly piezoresistive. However, for the PFRP layer, different factors can contribute to the electrical resistivity measurements. These were attributed to geometrical changes (deformation) in the specimen. Transverse cracking in glass fiber cross-ply laminates with a doped matrix showed an increase in the measured electrical resistance. The authors identified three factors contributing to the overall piezoresistive behavior. These are geometrical changes of the structure (deformations), inherent piezoresistive fiber and matrix response, and accumulation of micro-damage.

The effective conductivity (thermal and electrical) in heterogeneous materials has been the subject of a large volume of publications and research over more than four decades. Many micromechanical theories have been proposed and studied with different interaction conditions imposed at the interface-interphase between the fiber and matrix constituents. In addition, the geometry of the fiber and volume fractions has also been considered. The elastic and electric fields in a piezoelectric inhomogeneity was considered (Benveniste, 1992) and an analytical solution was obtained for an ellipsoidal type inclusion. A unified formulation (Benveniste and Milton, 2003) for a composite with ellipsoidal inclusions was considered with uncoupled conductivity and elasticity and coupled thermoelectricity and piezomagnetolectricity. An assemblage of uniform aspect-ratio was considered of ellipsoidal inclusions that could include a void and a coating. Thus the two-phase composite media was solved for the above uncoupled and coupled conduction problems on the basis of potential. Different and similar recent advanced approaches for conductivity type linear effective micro-electromechanical models and studies have also been published (Tane et al., 2005; Duan et al., 2006; Park et al., 2008; Sevostianov and Kachanov, 2008; Lee and Kim, 2010). Having said that, these outlined micro-models are primarily concerned with generating the effective behavior of the heterogeneous material, and typically consider the periodic medium with deformation-independent electromechanical constituents and without evolving damage. Thus, these may not help obtaining a solution for the piezoresistive properties if the objective is to

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