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Rate dependent non-linear magneto-electro-mechanical response of layered magneto electric composites: Theoretical and experimental approach

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a r t i c l e i n f o

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A B S T R A C T

Magnetoelectric (ME) composite structures have a wide range of applications in various industries due to the presence of ME coupling behavior. In the present work, ME coupling behavior is accounted for by considering mechanically bonded ferroelectric and magnetostrictive phases. Firstly, the rate dependent behaviors offerroic phases are observed by performing experimental investigations at room temperature. To depict the response of ferroic materials, thermodynamically consistent model is proposed. In order to account for the rate dependent behavior, a penalty function is introduced. Simulation studies have been performed by obtaining model parameters from the experimental studies and results are presented. Simulated results are in good agreement with the experimental data. This work is further extended to capture the response of ME composites by employing a simple homogenization technique. Mechanical, electrical and ME coupling behavior of ME composites are characterized in terms of effect of the volume fraction of the ferroic phases.

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1. Introduction

Multiferroic material with the presence of ferroic ordering such as ferroelectricity and ferromagnetism displays ME coupling behavior. This coupling phenomenon which tunes electrical polarization for an applied magnetic field or magnetization for an applied electric field is termed as ME effect [\[1\].](#page--1-0) In recent decades, multiferroic magneto electric (ME) materials have gained an importance due to their applications in various fields such as energy harvesting, magnetic field sensors and information storage devices [\[2–5\].](#page--1-0)

Presence of ME effect was experimentally observed in a single phase material like Cr_2O_3 [\[6\].](#page--1-0) However, single phase materials exhibit low ME effect (20 mV/cmOe) at room temperature which limits their applications in the practice [\[7\].](#page--1-0) In order to enhance ME effect at room temperature, several composite configurations were prepared using ferroelectric and ferromagnetic material phases. Depending upon the connectivity between the constituent phases, composites can be classified as particulate, fibrous and laminated composites [\[8\].](#page--1-0) Composites prepared in laminated configuration exhibit highest ME effect without altering physical properties of constituent phases [\[9\].](#page--1-0) However, under external stimuli, con-

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[http://dx.doi.org/10.1016/j.sna.2017.09.018](dx.doi.org/10.1016/j.sna.2017.09.018) 0924-4247/© 2017 Elsevier B.V. All rights reserved. stituent phases of ME composites exhibit nonlinear constitutive behavior. Hence, a mathematical model is necessary to predict the nonlinear constitutive behavior of ferroelectric and magnetostrictive phases.

In literature, several constitutive models are developed to depict the response of ferroelectric ceramics. In a broader perspective, constitutive models can be categorized as micromechanical and phenomenological models. In micromechanical models, the response is captured by considering the material at domain level. Effective response of the polycrystalline ferroelectric ceramics is obtained by averaging the response of all grains oriented in different crystallographic directions. To capture nonlinear response of a single grain, volume fraction of domain variants were considered as internal variables. For an incremental loading, internal variables could be updated by satisfying the switching criteria $[10-14]$. Application of micromechanical models are not efficient for large scale structures owing to their less computational efficiency due to the presence of large number of variables. Similarly, in magnetostrictive materials, micromechanical models were able to capture the macroscopic response by analyzing the three dimensional domain structures. To simulate devices, microscopic framework is computationally inefficient due to the presence of large number of domains [\[15–19\].](#page--1-0) Hence, phenomenological modeling with lesser number of variables provide a computationally efficient method to analyze large scale devices.

In phenomenological modeling, constitutive equations could be developed by employing a thermodynamic free energy potential. To capture nonlinear and hysteretic behavior, internal variables would be introduced along with a yield condition. By using evolution equations, internal variables can be updated for incremental loading. Various phenomenological models were reported to capture rate independent behavior of ferroelectric ceramics [\[20–24\].](#page--1-0) A three dimensional model was built based on a symmetric tangent approach by considering the remnant parts of strain and polarization as internal variables [\[21\].](#page--1-0) The rate independent response of the ferroelectric ceramics were captured by examining the movement of switching surface and irreversible part of electric field was considered as internal variable [\[22\].](#page--1-0) In another investigation, a theoretical model was built based on the framework of phenomenological model and internal variables were expressed in terms of volume fraction of the domain variants [\[24\].](#page--1-0) Meanwhile, the nonlinear constitutive models were also developed to capture the rate independent behavior of magnetostrictive materials [\[25–29\].](#page--1-0) A thermodynamically consistent macroscopic model was built by considering Gibbs energy as a free energy function and by expressing the strain as a quadratic function of magnetization [\[26\].](#page--1-0) In another work, a macroscopic model was built by considering the irreversible parts of magnetization and strain as the internal variables. For an incremental loading, internal variables were updated by employing a quadratic yield function with kinematic hardening [\[25\].](#page--1-0) A macroscopic model was built based on a thermodynamic framework and irreversible parts of magnetic field and strain were considered as internal variables [\[27\].](#page--1-0)

However, under external stimuli domain reorientation inherently depends upon the frequency of loading. Hence, the rate dependent behavior of the ferroelectric ceramics and magnetostrictive materials need to be investigated. Rate dependent behavior of ferroelectric ceramics was experimentally reported and it was observed that increase in the frequency of applied field, results in increase in coercive field [\[30\].](#page--1-0) Based on an incremental variational framework, a theoretical model was built with concept analogous to viscoplasticity and irreversible polarization was considered as internal variable [\[31\].](#page--1-0) A rate dependent nonlinear constitutive model was built based on the single integral form by considering the electric field with higher order terms which accounts for the nonlinear behavior $[32]$. The effect of the frequency and grain size on the response of noncrystalline poly crystal ferroelectrics were examined using a phase field model [\[33\].](#page--1-0) To lessen the sharp profiles of butterfly curves, a theoretical model was established by assuming a quadratic relation between the remnant strain and polarization [\[34\].](#page--1-0)

Various theoretical models were also reported to capture the frequency dependent response of magnetostrictive materials. By incorporating the principles of structural dynamics, a dynamic model was built based on the Jiles–Atherton model and strain response was captured for various operating frequencies [\[35\].](#page--1-0) In another work, a nonlinear constitutive model was reported based on thermodynamic framework which captures the magnetostrictive behavior by considering the eddy current effects [\[36\].](#page--1-0) A nonlinear dynamic model was built by accounting for magnetoelasto-thermal coupling and hysteresis losses. Effect of hysteresis losses on the resonance frequency was examined in terms of amplitude reduction and shift of resonance frequency [\[37\].](#page--1-0) Nonlinear and inelastic behavior of magnetostrictive material was captured using a variational formulation approach in which field variables were obtained by satisfying local and global variational minimization and stationary principles [\[38\].](#page--1-0)

An important aspect of ME composites is their product property which illustrates ME coupling potential of a composite. In literature, various works were emphasized in improving the product property by altering various parameters such as geometry, annealing temperature and connectivity of constituents.[\[39–43\].](#page--1-0) Meanwhile, various works were reported to capture the linear constitutive behavior of composite by obtaining effective properties as a function of volume fraction of constituent phases [\[44–50\].](#page--1-0)

Recently, there has been a growing interest in examining the nonlinear hysteretic behavior of ME composites [\[51,52\].](#page--1-0) Hysteretic behavior of ME composites useful in developing self-biased MEMS scalable energy harvesting devices and self biased magnetic sensors [\[53\].](#page--1-0) In practice, self-biased ME composites can be used without a DC magnetic source. This is due to the presence of ME coefficient at zero DC bias. In literature, few theoretical models were reported to capture the rate independent nonlinear and hysteretic behavior of ME composites [\[54–57\].](#page--1-0) Nonlinear behavior of trilayered ME composite was obtained by employing a nonlinear constitutive relation for the magnetostrictive material and the linear constitutive relation for the ferroelectric material. Further, ME response was captured using a theoretical model which was built based on equivalent circuit by considering the effect of insulating layers and the mechanical losses [\[55\].](#page--1-0) In another work, response of multiferroic composites was captured by employing physically motivated and phenomenological models for ferromagnetic materials [\[56\].](#page--1-0)

In the present work, on the basis of thermodynamic framework, a rate dependent constitutive model is formulated for ferroelectric and magnetostrictive materials. The model is built such that the internal variables can be captured without an iterative procedure which makes it computationally efficient. Subsequently, the rate dependent behavior is accounted for by introducing a penalty type function. Then, a study has been made to capture the frequency dependent nonlinear constitutive behavior of ME composites. To obtain the response of the composite, Voigt homogenization technique is employed along with the proposed constitutive models for the ferroelectric and magnetostrictive phases. Proposed rate dependent constitutive models were employed to capture the hysteretic behavior of ME response of composite. Hence, this formulation can be adopted to capture the hysteretic behavior of ME composites with less computational effort.

2. Experimental description

To analyze the rate dependent behavior of the multiferroic composite, the rate dependent behavior of each constituent in the composite is studied. To perform this study, cyclic loading of external field with various frequencies are applied and the nonlinear hysteretic response of the ferroelectric and the ferromagnetic material are observed. In the following sub-sections, the experimental setups established to perform the present study are described.

2.1. Experimental setup for electrical loading

Ferroelectric material behavior is characterized by obtaining electrical response in terms of dielectric displacement and mechanical response in terms of longitudinal strain. Function generator (Tektronix AFG3022B) is used to input bipolar waveform and then it is amplified using high voltage amplifier (TREK PD05034). An electric field is applied in the thickness direction of piezoelectric ceramic (PZT 5A) sample of size 15 mm \times 15 mm \times 1.5 mm. Laser vibrometer (NLV-2500-5) is employed to measure the longitudinal strain. Data for measuring polarization is obtained by using the Electrometer (Keithley 6517B) and sawyer tower circuit. DAQ card (NI 9215) along with Lab-view application software is used to collect data from all sources. To avoid external vibration during experiment, whole setup is placed on a vibration isolation table.

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