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Numerical model for the characterization of Maxwell-Wagner relaxation in piezoelectric and flexoelectric composite material

B.H. Nguyen [c], X. Zhuang [c,d], T. Rabczuk [a,b,*]

[a] Division of Computational Mechanics, Ton Duc Thang University, Ho Chi Minh City, Viet Nam
[b] Faculty of Civil Engineering, Ton Duc Thang University, Ho Chi Minh City, Viet Nam
[c] Institute for Continuum Mechanics, Leibniz Universität Hannover, Appelstraße 11, 30167 Hannover, Germany
[d] State Key Laboratory of Structural Analysis for Industrial Equipment, Dalian University of Technology, Dalian 116024, PR China

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ABSTRACT

Bi-layer structures can be engineered to investigate the interfacial polarization (Maxwell-Wagner polarization) of heterogeneous dielectric material, which shows the frequency-dependent property of the effective dielectric permittivity. However, in piezoelectric or flexoelectric heterostructures, behaviors of the effective piezoelectric or flexoelectric coefficients are remained unclear. Therefore, in this work, we present a numerical model of the Maxwell-Wagner polarization effect in a bi-layer structure made of piezoelectric or flexoelectric material. In this model, the conductivity, which qualitatively represents the free charge in a real dielectric material, is introduced to the complex dielectric permittivity. Several numerical examples are performed to validate the model and investigate the frequency dependence of the effective dielectric permittivity, piezoelectric and flexoelectric coefficients as well as the giant enhancement of dielectric constants. It is found that the static (at low frequency) and the instantaneous (at high frequency) effective coefficients are governed by those of the thin and thick layer, respectively. Moreover, both conductivity and volume ratio play essential roles in the enhancement of the dielectric constant that is underpinned by the Maxwell-Wagner effect.

1. Introduction

The Maxwell-Wagner (MW) effect characterizes the interfacial polarization taken place at the interface of two dissimilar materials due to the difference in their dielectric properties, i.e. permittivity and conductivity. The MW mechanism does not only interpret the physics of the dielectric mixture but also is responsible for the colossal dielectric constant ($> 10^3$) in non-ferroelectric materials [1,2], such as $CaCu_3Ti_4O_{12}$ [3,4] and hexagonal $BaTiO_3$ [5,6], in which the interfacial polarization occurs when the space charge accumulates at the internal boundary layers due to the inhomogeneous microstructure or at the external depletion layer of the electrode/sample interface due to the Schottky barrier effect. This mechanism is analogous to a very thin parallel-plate capacitor, thus very high capacitance can be obtained [1,2]. Nowadays, the development of portable electronic devices entails higher demand on energy storage, life cycle and charge time, which heavily depend on the material properties. Therefore, predicting the electric response of high dielectric constant materials is of great importance. Numerical analysis is often a time and cost efficient approach. Modern numerical method, such as finite element method (FEM) or boundary element method (BEM) had been used to predict the effective dielectric constant in periodic, lossy and random dielectric composite material in the work of Sareni et al. [7–9]. Tuncer et al. [10] study the dielectric relaxation in a binary dielectric mixture by using FEM and present comparisons with different dielectric mixture formulas. Similar work can be found in [11]. A review on numerical study based on FEM of dielectric properties of binary dielectric composite structure is reported in [12]. On the other hand, permittivity dispersion of dielectric solid was also studied by the network of resistors and capacitors in the work of Almond and Vainas [13]. In these aforementioned references, the MW effect was considered to describe the frequency dispersion of the effective dielectric permittivity of composite structures, but the resulting large dielectric constant was not discussed. Furthermore, the MW effect is also studied analytically in a serial bi-layer ferroelectric material, showing that due to the MW relaxation, the longitudinal direct piezoelectric coefficient exhibits either relaxation or retardation [14]. As an extension, Turik and Radchenko [15] analytically study a more comprehensive model of

* Corresponding author at: Ton Duc Thang University, Ho Chi Minh City, Viet Nam.

E-mail address: timon.rabczuk@tdt.edu.vn (T. Rabczuk).

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the MW relaxation in piezoactive media, accounting for boundary conditions and transversal piezoelectric response. Within their work, both piezoelectric relaxation and giant enhancement of dielectric permittivity were analytically investigated. However, the flexoelectric effect, as we will briefly introduce in the following, was not considered in these works.

Flexoelectricity, which was theoretically proposed more than half a century ago [16–18], describes the spontaneous induced polarization from strain gradients in all dielectric material regardless of the symmetry of the structure. The effect is appreciable at the submicron or nano length scale as the strain gradient increases. The significance of flexoelectricity can be found not only in microelectromechanical or nanoelectromechanical systems [19–21] but also in different physical phenomena such as strain gradient-driven polarity control [22]. Excellent review articles about flexoelectricity can be found for instance in [23–26]. Nonetheless, the understanding of flexoelectricity needs to be further explored as the theoretical predictions and experimental measurements are comparatively discrepant. While theory conjectures the flexoelectric coefficients have the magnitude of 10^{-9} (C/m) [18,25], measured data in high dielectric constant materials (e.g. ferroelectric materials) can be as high as 10^{-6} (C/m) [27–31] and more recently, the largest effective flexoelectric coefficient with the magnitude of 10^{-3} (C/m) has been reported in oxide semiconductor [32].

It had been both theoretically and experimentally confirmed that the large flexoelectric response can be found in high dielectric constant materials, in which the MW effect plays an essential role. Therefore, the aim of this work is to propose a numerical study of the MW effect in a serial bi-layer piezoelectric or flexoelectric structure. It should be noted that, although more complex composite could be investigated, a simple serial bi-layer structure is chosen because: (i) the numerical model can be validated with available theoretical model (e.g. [15], only for piezoelectricity); (ii) resemble the experimental setup, in which one thin surface layer stays on top of a bulk layer [1,5,32]. In our numerical model, the free charge, which accounts for the interfacial polarization under certain range of frequency, is characterized by the conductivity of the material and is introduced to the complex dielectric permittivity. With appropriate boundary conditions, frequency dependence of the effective material parameters can be obtained as a result of a boundary value problem in piezoelectric or flexoelectric domain. To the contrary of the well developed continuum model of piezoelectricity (e.g. [33]), continuum modeling of flexoelectricity has been recently developed [34–38]. Owing the size effect from strain gradient, the computational model of flexoelectricity necessitates C^1 continuity of the basis functions in a Galerkin framework. This difficulty can be overcome by local maximum-entropy meshfree method (LME) [39–42], mixed FEM [43–45], triangular Argyris elements [46] and isogeometric analysis (IGA) [47]. While the LME and IGA methods enjoy to satisfy the C^1 continuity condition conveniently, mixed formulation FEM requires additional degrees of freedom as well as the fulfillment of the inf-sup condition [48]. In this paper, we employed IGA [49,50] since we believe it is computationally less costly than LME approximations. Additionally, by employing the advantages of handling complex geometry of IGA, our numerical framework could be extended to study the interfacial polarization in more complicated piezoelectric or flexoelectric composite structures.

The remainder of the paper is organized as follows: In Section 2, we present the constitutive relations derived from the electric Gibbs free energy density and the Maxwell-Wagner interfacial polarization model in the piezoelectric and flexoelectric domain based on the dielectric relaxation. Consequently, the frequency dependence of the effective material parameters can be obtained by solving the boundary value problem numerically based on the Hamilton's variational principle and isogeometric analysis which are presented in Section 3. We then report our numerical results of the MW effect in piezoelectric and flexoelectric bi-layer structures in Section 4 and discuss our observations in Section 5.

2. Theory background

2.1. Constitutive equations

In this section we summarize the linear theory of dielectric solids possessing piezoelectricity and flexoelectricity proposed by [34,35]. The electric Gibbs energy density is expressed in terms of the linear strain tensor ϵ_{ij}, strain gradient $\epsilon_{jk,l}$ and electric field E_i [36]:

$$\mathcal{G}(\epsilon_{ij}, E_i, \epsilon_{jk,l}) = \frac{1}{2} c_{ijkl}\epsilon_{ij}\epsilon_{kl} - e_{ikl}E_i\epsilon_{kl} - \mu_{ijkl}E_i\epsilon_{jk,l} - \frac{1}{2}\kappa_{ij}E_iE_j, \tag{1}$$

in which c_{ijkl} is the fourth-order elastic tensor, e_{ikl} is the third-order piezoelectric tensor, κ_{ij} is the second-order dielectric tensor and μ_{ijkl} is the fourth-order flexoelectric tensor. Note that tensor μ_{ijkl} is combined from the direct and converse flexoelectric tensors through integration by parts, details has been shown in [36]. We also remark that higher order tensors (i.e. fifth-order and sixth-order tensor) are neglected for the sake of simplicity. Within the linear regime, the strain ϵ_{ij}, the strain gradient $\epsilon_{jk,l}$ and the electric field E_i are defined from the displacement filed u_i and electric potential ϕ as

$$\epsilon_{ij} = \frac{1}{2}(u_{i,j} + u_{j,i}), \tag{2a}$$

$$\epsilon_{jk,l} = \frac{1}{2}(u_{j,kl} + u_{k,jl}), \tag{2b}$$

$$E_i = -\phi_{,i}. \tag{2c}$$

Consequently, the constitutive relations can be derived from the electric Gibbs energy density as follows

$$\sigma_{ij} = \hat{\sigma}_{ij} - \tilde{\sigma}_{ijk,k} = c_{ijkl}\epsilon_{kl} - e_{kij}E_k + \mu_{lijk}E_{l,k}, \tag{3a}$$

$$D_i = e_{ikl}\epsilon_{kl} + \mu_{ijkl}\epsilon_{jk,l} + \kappa_{ij}E_j, \tag{3b}$$

in which the classical stress $\hat{\sigma}_{ij}$ and electric displacement D_i as well as the higher order stress $\tilde{\sigma}_{ijk}$ are defined as

$$\hat{\sigma}_{ij} = \frac{\partial \mathcal{G}}{\partial \epsilon_{ij}}, \quad D_i = -\frac{\partial \mathcal{G}}{\partial E_i}, \tag{4a}$$

$$\tilde{\sigma}_{ijk} = \frac{\partial \mathcal{G}}{\partial \epsilon_{ij,k}}. \tag{4b}$$

By means of Eq. (3), the electric Gibbs energy density can be rewritten as

$$\mathcal{G}(\epsilon_{ij}, E_i, \epsilon_{jk,l}) = \frac{1}{2}\hat{\sigma}_{ij}\epsilon_{ij} + \frac{1}{2}\tilde{\sigma}_{ijk}\epsilon_{ij,k} - \frac{1}{2}D_iE_i, \tag{5}$$

which will be used to defined the boundary value problem in the following section.

2.2. Formulation

Let us consider an elastic dielectric solid occupying a volume Ω bounded by the boundary $\partial\Omega$. Under the assumption of free body force, the Hamilton's variational principle can be employed to obtain equilibrium equations and boundary conditions such that

$$\delta \int_{t_1}^{t_2} (K - G + W)\mathrm{d}t = 0, \tag{6}$$

in which the kinetic energy K, the internal energy G and the external work W are defined as

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