



Influence of second-phase inclusions on the electro-deformation of ferroelectric ceramics



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ABSTRACT

Theoretical estimates are given for the overall dissipative response of ferroelectric ceramics with second-phase inclusions, under arbitrary electromechanical loading histories. The ferroelectric behavior of the constituent phases is described via a stored energy density and a dissipation potential in accordance with the theory of generalized standard materials. An implicit time-discretization scheme is used to generate a variational representation of the overall response in terms of a single incremental potential. Estimates are then generated by constructing sequentially laminated microgeometries of particulate type whose overall incremental potential can be computed exactly. Because they are realizable, by construction, these estimates are guaranteed to conform with any material constraints, to satisfy all pertinent bounds, and to exhibit the required convexity properties with no duality gap. By way of example, the theory is used to study the influence of metallic particles and of microcavities on the electro-deformability of a lead zirconate titanate. In particular, the role of remanent polarization fluctuations on the piezoelectric properties is assessed.

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1. Introduction

The search for electro-deformable materials with specific combinations of properties not found in monolithic ferroceramics has motivated the development of an increasing variety of two-phase ferroelectric composites. A natural option consists in dispersing in a polycrystalline ferroelectric ceramic second-phase inclusions such as metallic particles (e.g., Duan et al., 2000; Ning et al., 2012; Zhang et al., 2010) or microcavities (e.g., Piazza et al., 2010). In practice, these composite materials are first synthesized by a suitable fabrication process, then permanently polarized by application of a strong electric field, and finally employed within their piezoelectric regime. Now, the influence of a second phase on the final piezoelectric properties of the composite system is not evident a priori in view of the intricate role of material heterogeneity in the poling process. The purpose of this work is to estimate theoretically such influence in terms of the constitutive properties of the phases and the microstructural characteristics of the composite. A wide range of micromechanical models have already been proposed for that purpose—see, for instance, the monograph of Topolov and Bowen (2009)—, but all

proposals invariably treat the ferroceramic matrix of a poled composite as a stress-free phase with uniform piezoelectric properties. Due to material heterogeneity, however, strong spatial variations of the electric field can arise during the poling process which, in turn, can result in residual stresses and non-uniform piezoelectric coefficients within the permanently poled specimen (e.g., Idiart, 2014). To account for these features, the entire poling process must be simulated.

Given that ferroelectricity is largely hysteretic, the problem calls for a methodology to estimate the overall response of two-phase deformable dielectrics with complex particulate microstructures and with constituent phases that can simultaneously store and dissipate electro-mechanical energy. Estimates of this sort for rigid dielectrics have been recently derived by Idiart (2014). These estimates rely on the generalized-standard material model for ferroelectricity proposed by Bassiouny et al. (1988)—which identifies the irreversible electric polarization as an internal variable—and on the variational representation of Miehe and Rosato (2011) for the macroscopic response of heterogeneous ferroelectric solids in terms of an effective incremental potential. A special class of microgeometries is then identified such that it reproduces the essential geometrical features of the actual composite microstructure while at the same time allow the exact computation of this effective potential. The class consists of certain sequentially laminated microgeometries which have been successfully used already to model other types of particulate systems with nonlinear behavior such as viscoplastic composites and porous

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media (e.g., [Danas et al., 2008](#); [deBotton and Hariton, 2002](#); [Idiart, 2008](#)), and non-ohmic composite conductors ([Hariton and deBotton, 2003](#); [Idiart and Ponte Castañeda, 2013](#)). The predictions always conform with material constraints, satisfy all pertinent bounds, and exhibit the required convexity when applicable. This consistency is guaranteed by the fact that the estimates are realizable –i.e., exact for a given class of material systems– by construction. The present work provides a generalization of this approach to *deformable* dielectrics.

We begin in [Section 2](#) by formulating the problem of a heterogeneous dielectric body undergoing small deformations. The overall response is defined for a general composite system in [Section 3](#) and then given for sequentially laminated systems in [Section 4](#). By way of example, specific results are reported in [Section 5](#) for a lead zirconate titanate with either metallic particles or microcavities. We conclude the presentation by identifying a potential issue with a class of constitutive models commonly used for monolithic polycrystalline ferroceramics.

2. The composite material model

2.1. The material system and field equations

The material system under study is idealized here as a heterogeneous body occupying a domain Ω and made up of a continuous matrix containing a uniform dispersion of second-phase inclusions. The matrix phase will be identified with the index $r = 1$ while the inclusions will be collectively identified with the index $r = 2$. Each phase occupies a domain $\Omega^{(r)} \subset \Omega$ ($r = 1, 2$) such that $\Omega = \Omega^{(1)} \cup \Omega^{(2)}$. The domains $\Omega^{(r)}$ are described by a set of characteristic functions $\chi^{(r)}(\mathbf{x})$, which take the value 1 if the position vector \mathbf{x} is in $\Omega^{(r)}$ and 0 otherwise.

We restrict attention to isothermal processes produced by quasistatic electromechanical interactions. These interactions are exerted by a fixed electrostatic potential $\hat{\phi}$ applied via surface electrodes occupying a portion $\partial\Omega_v$ of the body boundary $\partial\Omega$ and by a surface displacement $\hat{\mathbf{u}}$ applied on a portion $\partial\Omega_u$ of the body boundary. For simplicity, we disregard the possible presence of free charges within the material. The governing field equations are then given by –see, for instance, [Kamlah \(2001\)](#)–

$$\nabla \cdot \mathbf{D} = 0 \quad \text{and} \quad \mathbf{E} = -\nabla\phi \quad \text{in } \mathbb{R}^3, \tag{1}$$

$$\nabla \cdot \boldsymbol{\sigma} = 0 \quad \text{and} \quad \boldsymbol{\varepsilon} = \nabla \otimes_s \mathbf{u} \quad \text{in } \Omega, \tag{2}$$

with

$$\mathbf{D} = \begin{cases} \epsilon_0 \mathbf{E} & \text{in } \mathbb{R}^3 \setminus \Omega \\ \epsilon_0 \mathbf{E} + \mathbf{P} & \text{in } \Omega \end{cases} \tag{3}$$

and boundary conditions

$$\phi = \hat{\phi} \quad \text{on } \partial\Omega_v \quad \text{and} \quad [\mathbf{D} \cdot \mathbf{n}] = 0 \quad \text{on } \partial\Omega \setminus \partial\Omega_v, \tag{4}$$

$$\mathbf{u} = \hat{\mathbf{u}} \quad \text{on } \partial\Omega_u \quad \text{and} \quad [\boldsymbol{\sigma} \mathbf{n}] = \mathbf{0} \quad \text{on } \partial\Omega \setminus \partial\Omega_u. \tag{5}$$

In these expressions, ϕ and \mathbf{u} are continuous fields representing the electrostatic potential and the displacement, \mathbf{D} , \mathbf{E} , \mathbf{P} , $\boldsymbol{\sigma}$ and $\boldsymbol{\varepsilon}$ are, respectively, the electric displacement, the electric field intensity, the material polarization, and the stress and strain tensors, $[\cdot]$ denotes the jump across $\partial\Omega$, \mathbf{n} is the outward normal vector to $\partial\Omega$, and ϵ_0 denotes the electric permittivity of vacuum. In turn, ∇ is the standard nabla operator and the symbol \otimes_s represents the symmetric part of the tensor product. Along internal surfaces of discontinuity, the various fields must satisfy the jump conditions

$$[\phi] = 0, \quad [\mathbf{D} \cdot \mathbf{n}] = 0, \quad [\mathbf{u}] = 0, \quad [\boldsymbol{\sigma} \mathbf{n}] = \mathbf{0}, \tag{6}$$

where \mathbf{n} denotes the normal vector to the discontinuity surface. In addition, the electrostatic potential must vanish at infinity, i.e., $\phi \rightarrow 0$ as $|\mathbf{x}| \rightarrow \infty$.

The above field equations must be supplemented with constitutive relations describing the electromechanical response of each phase. We adopt the thermodynamic approach of [Bassiouny et al. \(1988\)](#) wherein dissipative processes are characterized by an irreversible polarization \mathbf{p} playing the role of an internal variable. This framework is general enough to characterize simple responses such as linear polarizability as well as complex responses such as rate-dependent ferroelectricity –see, for instance, [Kamlah \(2001\)](#), [Miehe and Rosato \(2011\)](#).

The total energy of the material system and its surroundings is thus written as

$$\mathcal{E} = \int_{\Omega} \epsilon(\mathbf{x}, \boldsymbol{\varepsilon}, \mathbf{P}, \mathbf{p}) \, dV + \int_{\mathbb{R}^3} \frac{1}{2} \epsilon_0 \mathbf{E}^2 \, dV \tag{7}$$

where the first term corresponds to the energy stored in the composite material while the second term is the electrostatic energy of the electric field. The energy density ϵ is taken to depend explicitly on position due to the heterogeneity of the body. In turn, the dissipation of the system is assumed to be of the form

$$\mathcal{D} = \int_{\Omega} \frac{\partial \varphi}{\partial \dot{\mathbf{p}}}(\mathbf{x}, \dot{\mathbf{p}}) \cdot \dot{\mathbf{p}} \, dV, \tag{8}$$

where φ is a convex, positive function of the irreversible polarization rate $\dot{\mathbf{p}}$ such that $\varphi(\cdot, \mathbf{0}) = 0$, which is used to characterize the microscopic domain switching in the ferroelectric phase. The form (8) guarantees a positive dissipation.

Thermodynamic arguments then imply that the constitutive relations of the material are given by (see [Bassiouny et al., 1988](#))

$$\begin{aligned} \mathbf{E} &= \frac{\partial \epsilon}{\partial \mathbf{P}}(\mathbf{x}, \boldsymbol{\varepsilon}, \mathbf{P}, \mathbf{p}), \quad \boldsymbol{\sigma} = \frac{\partial \epsilon}{\partial \boldsymbol{\varepsilon}}(\mathbf{x}, \boldsymbol{\varepsilon}, \mathbf{P}, \mathbf{p}) \\ \text{and} \quad \frac{\partial \epsilon}{\partial \mathbf{p}}(\mathbf{x}, \boldsymbol{\varepsilon}, \mathbf{P}, \mathbf{p}) + \frac{\partial \varphi}{\partial \dot{\mathbf{p}}}(\mathbf{x}, \dot{\mathbf{p}}) &= \mathbf{0}, \end{aligned} \tag{9}$$

where the first two expressions relate the electric field intensity and stress with the polarization and strain, and the last expression provides the evolution law for the irreversible polarization \mathbf{p} . In the case of nonsmooth potentials, the derivatives in (9) should be understood in the sense of the subdifferential of convex analysis. These constitutive relations conform to the so-called generalized standard material model provided the energy $\epsilon(\mathbf{x}, \cdot, \cdot, \cdot)$ is convex ([Germain et al., 1983](#)). In that case, the polarization can be eliminated from the constitutive description in favor of the electric field intensity by defining the free energy density

$$\psi(\mathbf{x}, \boldsymbol{\sigma}, \mathbf{E}, \mathbf{p}) \doteq \sup_{\mathbf{P}, \boldsymbol{\varepsilon}} [\boldsymbol{\sigma} \cdot \boldsymbol{\varepsilon} + \mathbf{E} \cdot \mathbf{P} - \epsilon(\mathbf{x}, \boldsymbol{\varepsilon}, \mathbf{P}, \mathbf{p})] + \frac{1}{2} \epsilon_0 \mathbf{E}^2, \tag{10}$$

where the first term corresponds to a partial Legendre transformation of ϵ with respect to \mathbf{P} and $\boldsymbol{\varepsilon}$. Note that the function ψ is thus convex in \mathbf{E} and $\boldsymbol{\sigma}$ but concave in \mathbf{p} . The constitutive relations (9) can then be written as

$$\begin{aligned} \mathbf{D} &= \frac{\partial \psi}{\partial \mathbf{E}}(\mathbf{x}, \boldsymbol{\sigma}, \mathbf{E}, \mathbf{p}), \quad \boldsymbol{\varepsilon} = \frac{\partial \psi}{\partial \boldsymbol{\sigma}}(\mathbf{x}, \boldsymbol{\sigma}, \mathbf{E}, \mathbf{p}) \\ \text{and} \quad \frac{\partial \psi}{\partial \mathbf{p}}(\mathbf{x}, \boldsymbol{\sigma}, \mathbf{E}, \mathbf{p}) - \frac{\partial \varphi}{\partial \dot{\mathbf{p}}}(\mathbf{x}, \dot{\mathbf{p}}) &= 0. \end{aligned} \tag{11}$$

Making use of the characteristic functions $\chi^{(r)}$, the potentials ψ and φ are finally expressed as

$$\begin{aligned} \psi(\mathbf{x}, \boldsymbol{\sigma}, \mathbf{E}, \mathbf{p}) &= \sum_{r=1}^2 \chi^{(r)}(\mathbf{x}) \psi^{(r)}(\boldsymbol{\sigma}, \mathbf{E}, \mathbf{p}), \\ \varphi(\mathbf{x}, \dot{\mathbf{p}}) &= \sum_{r=1}^2 \chi^{(r)}(\mathbf{x}) \varphi^{(r)}(\dot{\mathbf{p}}), \end{aligned} \tag{12}$$

where $\psi^{(r)}$ and $\varphi^{(r)}$ denote, respectively, the free energy densities and dissipation potentials of each phase r .

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