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Effect of the electric conductivity on the modeling of the poling process of ferroelectric components

Holger Schwaab [a,c], Marco Deluca [b,c], Peter Supancic [b,c], Marc Kamlah [a,*]

[a] Karlsruhe Institute of Technology (KIT), Institute for Applied Materials, Hermann-von-Helmholtz-Platz 1, 76344 Eggenstein-Leopoldshafen, Germany
[b] Institut für Struktur- und Funktionskeramik, Montanuniversität Leoben, Franz-Josef- Straße 18, 8700 Leoben, Austria
[c] Materials Center Leoben Forschung GmbH, Roseggerstraße 12, 8700 Leoben, Austria

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ABSTRACT

Piezoceramic materials, despite being semiconductors with a high but finite ohmic resistance, are generally modeled as perfect insulators. In this work the influence of the electric conductivity on the poling process of piezoceramic materials is investigated. To solve the electromechanically coupled boundary value problem, a reduced form of the Maxwell equations is implemented inside a hybrid finite element formulation. In this formulation the electric displacement is available as nodal quantity (i.e. degree of freedom) which is used instead of the electric field to determine the evolution of the remanent polarization. The material model is fully ferroelectric/ferroelastic coupled, whereas the material behavior is described by a set of yield functions and the history dependence is stored in internal state variables representing the remanent polarization and the remanent strain. The simulation of poling processes for three different components are presented. First and second, the poling of a radially poled hollow cylinder and of a stack actuator is investigated. Here, a residual electric field appears after poling, leading to significant time-dependent changes of stresses and deformation due to subsequent charge transportation processes. The third example is a bending actuator composed of two layers of hard-PZT and soft-PZT material. It is shown that considering the electric conductivity new strategies for the poling process can be developed in order to improve the bending actuation. In this way, we show the importance of considering charge transportation processes in simulations of the poling of ferroelectrics, which seems not to have been recognized so far.

1. Introduction

Piezoelectric ceramics are due to their electromechanical coupling widely used in transducers, actuators and sensors. Materials such as lead zirconate titanate (PZT) and barium titanate ($BaTiO_3$), always piezoelectric on the micro-scale, in their as-sintered polycrystalline ceramic form they show no trace of piezoelectricity due to the random orientation of crystal orientation and the ferroelectric domains therein. Because of this, the so-called poling process is necessary in order to activate the macroscopic coupling. This is performed by applying electric fields high enough to force a permanent alignment of the microdipoles (the domains) (Jaffe et al., 1971). During this process large strains are introduced, which can lead to the initiation of cracks. Residual stresses often remain after poling in the component, thus influencing its

* Corresponding author. Tel.: +49 721 60 82 5860.
E-mail address: marc.kamlah@kit.edu (M. Kamlah).

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electromechanical properties and leading to a subsequent fatigue if high cycle loadings are applied (Lupascu, 2004). Nonlinear simulations are the only way to determine the poling state, the mechanical stresses and the strains as well as the remaining electric field within the component during and after the poling process. Knowledge of these quantities is essential for the development and the optimization of piezoelectric devices. Usually piezoceramic materials are modeled as perfect insulators, although they are in reality semiconductors with a high but finite ohmic resistance (Jaffe et al., 1971). The charge transfer influences the residual electric field after poling, whereas the already present electromechanical coupling may in turn affect mechanical properties.

The influence of the electric conductivity on acoustic waves in piezoelectric semiconductor materials is well known (Auld, 1969; Wauer and Suherman, 1997; Yang and Zhou, 2005). The electric field produces currents and space charges resulting in dispersion and acoustic loss. The interaction between a traveling acoustic wave and the mobile charges is called the acoustoelectric effect. In general a damping of the acoustic wave is obtained, although by an initial or biasing dc electric field the acoustic wave can be also amplified (White, 1962). In the above-mentioned papers the system is described by a set of ordinary differential equations (one point models), which do not allow considering the discretization of the local field and the piezoelectric behavior is described by linear electromechanically coupled functions. It is also well known that the electric conductivity has a significant impact on the poling process of composite structures. The piezoelectric properties of PZT/PU (polyurethane) are improved by adding a small amount (1.0 vol%) of graphite (Sakamoto et al., 2002; Sa-Gong et al., 1986). By adding graphite, the electric conductivity of PU increases, causing space charges to drift and accumulate at the particle-matrix interface in the polymer phase during the poling process (up to 1 h) (Kwok et al., 2007). This phenomenon stabilizes the poling orientation in the PZT and hence the electric field for poling can be reduced. A similar enhancement of the piezoelectric constant is observed in Chen et al. (1998) by soaking the PZT/PVDF composite in an *N*-dimethylacetamide solvent. Wong and Shin (2005) believe that this is related to an increase of the electric conductivity in the matrix material. In Or et al. (2003b) the poling behavior of ferroelectric multilayered composite structures is modeled and a comparison with the experimental results by Furukawa et al. (1986) is shown. For the composite system of a PZT and with a polymer, the electric field acting on the ceramic phase increases over time, while the external field remains constant. Therefore, a higher conductivity in the polymer phase speeds up the poling process.

The significant influence of the electric conductivity on the poling process of composite structures is demonstrated in the above-mentioned publications. The models explaining this behavior (Wong and Shin, 2005; Wei et al., 2007; Or et al., 2003a) only represent the ferroelectric behavior of piezoceramic materials in one dimension, and are restricted to a specific geometry under additional assumptions. The mechanical stresses applied by the matrix can lead to a depolarization of the PZT that cannot be predicted by these material models.

In this work a general framework to model the weak electric conductivity of piezoceramic materials inside a finite-element is presented. Therefore a hybrid form of the electromechanical weak formulation (Ghandi and Hagood, 1997) is used, taking the electric displacement $\dot{\vec{D}}$ and the strain $\boldsymbol{S}$ as independent variables, similar to the vector potential formulation proposed by Landis (2002). However, to overcome the problem of defining correct boundary conditions in the 3D vector potential formulation, the scalar potential φ is retained as a primary variable in the hybrid formulation. A macroscopic phenomenological material model is used to represent the hysteretic behavior of ferroelectric ceramics. The model is fully ferroelectric/ferroelastic coupled, and thus allows considering major nonlinear effects like ferroelectric, butterfly and ferroelastic hystereses, as well as coupling phenomena such as the depolarization driven by mechanical stresses.

This paper is organized as follows: in the first part, the hybrid finite element formulation including the electric conductivity is shown together with the constitutive material model. In the second part three examples are presented. The first one reports the simulation results of poling a hollow cylinder in radial direction. For this geometry an electric field appears directly after poling and the electric conductivity leads to a time-dependent change of stresses, strains and deformation. Similar effects appear in the second example where the poling process of a rounded electrode tip in a stack actuator is simulated. The results for these two systems represent a class of geometries where inhomogeneous polarization after poling appears. The non-divergence-free polarization field causes a residual electric depolarization field and the electric conductivity leads to the time-dependent effect shown in this paper. In the third example a simulation of poling a bi-layer composite bending actuator is presented. A similar system is investigated in Steinhausen et al. (2008) with a point model not considering the ferroelastic behavior. The bending actuator is composed of a soft- and a hard-PZT ceramic and with the presented poling process it is possible to obtain a polarization in each layer with opposite directions, which leads to improve bending actuation. This poling texture is only possible due to the electric conductivity of piezoceramic materials.

2. Weak formulation of the electromechanical problem including the electric conductivity

On the basis of the governing equations, the electromechanical boundary value problem is presented here under the assumptions of small deformations, isothermal and electrical quasi-static conditions, but including the electric conductivity. Static mechanical equilibrium requires

$$\text{div }\boldsymbol{\sigma}+\vec{f}^{B}=0, \tag{1}$$