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A constrained domain-switching model for polycrystalline ferroelectric ceramics. Part I: Model formulation and application to tetragonal materials

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Abstract

A micromechanical model is proposed to study the constrained domain-switching process in polycrystalline ferroelectric ceramics. It is assumed that the depolarization field induced by domain switching is completely compensated by free charges, while the stress caused by non-180° switching is considered in an Eshelby inclusion manner. The model assumes that each grain contains multi-domains and the domain-switching criterion is based on potential energy density. Two switching options, which are based on Hwang et al. [Hwang SC, Lynch CS, McMeeking RM. Acta Metall Mater 1995;43:2073] and Berlincourt and Krueger [Berlincourt D, Krueger HHA. J Appl Phys 1959;30:1804], are used in the model development. Details of the switching process are analyzed for tetragonal ferroelectric/ferroelastic ceramics under electric loading or uniaxial compression (tension) by using an inverse-pole-figure method. Numerical results show that during electric poling, only a few per cent 90° switching can occur in $BaTiO_3$ ceramics, which agrees well with experimental observations.

Keywords: Charge screening; Constitutive modeling; Domain switching; Ferroelectric ceramics; Poling

1. Introduction

Perovskite ferroelectric ceramics such as barium titanate ($BaTiO_3$) and lead titanate zirconate (PZT) are commonly used to make actuators, transducers, etc., due to their electromechanical coupling, ultra-fast response and compact size [1]. This class of materials shows excellent linear response at low electric fields but under a large electric field or high stress significant nonlinearities in response occur due to domain switching [2]. Domain switching in ferroelectric ceramics is crystal symmetry related. Only 180° and 90° domain switching exists in tetragonal ceramics, and 180°, 109° and 71° switching in rhombohedral ceramics. In ferroelectric single crystals, perfect alignment of polarization can be achieved and a single domain state can exist after poling by a strong DC field. However, this is not the case in ferroelectric ceramics, where the crystallite axes arrange in a random way and multiple domain states could exist after poling. The theoretical achievable polarization and strain in perovskite ferroelectric ceramics under electric or mechanical poling have been obtained analytically [3–5] and numerically [6,7]. However, strain measurements and X-ray diffraction studies show that after poling the fraction of completed 90° switching is 10-12% in $BaTiO_3$ ceramics [8,9] and 44–51% in tetragonal PZT and lead magnesium niobate titanate (PMN-PT) ceramics near the morphotropic phase boundary (MPB) [8,10]. This implies that a considerable amount of 90° switching is constrained by neighboring grains.

Modeling of domain switching in ferroelectric materials has received much attention, and both phenomenological models [11,12] and micromechanical models [13–18] exist in the literature. In the latter models, the material usually consists of numerous grains, each of which is a single domain or contains multi-domains [15,18]. Domain

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switching is then determined by an energy-based switching criterion, with intergranular interactions either neglected [13] or taken into account in an Eshelby inclusion manner [14–16] or by finite element methods [17,18]. According to the authors' knowledge, only Wan and Bowman [19] have specifically addressed the fraction of 90° switching (or non-180° switching) in domain switching modeling by assuming that the depolarization field is proportional to the fraction of completed 90° switching. Although the above-mentioned domain-switching models assume that the material is purely tetragonal, most experimental results of PZT ceramics are near the MPB where tetragonal and rhombohedral phases coexist. Recently, Li et al. [20] showed through a rigorous mathematical analysis that unless caused by a very large internal stress, no change of remnant strain is generated in a polycrystalline tetragonal or rhombohedral ferroelectric ceramic via non-180° domain switching. On the other hand, in PZT ceramics near the MPB, non-180° switching can almost be achieved similar to the case of a single crystal. Li et al.'s [20] analysis can accurately interpret most experimental results except the case of rhombohedral ceramics during electric poling where considerable remnant strain by 71° or 109° domain switching has been observed [21,22].

In this paper, a micromechanical model is developed to study the constrained domain-switching phenomenon in ferroelectric/ferroelastic ceramics with grains containing multiple domains by using two previously reported polarization-switching models, i.e. Hwang et al.'s model [13] and Berlincourt and Krueger's definition of complete 180° switching [10]. A description of the proposed constrained domain-switching model is presented in Section 2. In Sections 3 and 4, the domain-switching processes in tetragonal ferroelectric and ferroelastic polycrystalline materials are analyzed in detail under electric loading and uniaxial compression (tension) by using an inverse-pole-figure method. A discussion is presented in Section 5 and the conclusions are given in Section 6. In a subsequent paper [23], a combined switching assumption is presented and used to study constrained domain switching in rhombohedral materials.

2. Constrained domain-switching model

Assume that a polycrystalline ferroelectric ceramic is made up of numerous randomly oriented grains, each of which contains N types of domains, where $N = 6$ for the tetragonal case and $N = 8$ for the rhombohedral case. In an unpoled ceramic, the fraction of each type of domain in a grain is $1/N$, thus both the remnant polarization and strain of each grain is zero. The present model is also based on the assumptions that a polycrystalline ferroelectric ceramic is dielectrically and elastically isotropic and shows linear dielectric and elastic behavior unless domain switching occurs (i.e. all nonlinear polarization and strain are caused by domain switching).

2.1. Charge-screening effect and internal stress by non-180° domain switching

The charge-screening effect in real ceramics [21,22,24] is taken into account in the proposed model. That is, the depolarization field induced by polarization gradient or polarization change during domain switching is completely compensated by free charges. The free charges are trapped by unbalanced polarization and turn in to space charges, which cannot be driven by the applied electric field unless the polarization switches. When domain switching occurs, the space charges are released and move to the surface of a material. In fact, this is the principle that is used to measure the electric hysteresis loops in the Sawyer–Tower circuit and mechanical depolarization in short circuits. The remnant polarization of a ferroelectric ceramic cannot be measured if there is no charge-screening effect. However, the depolarization field may not be completely compensated and it can be very large within a thin layer just below the crystal surface [25]; this is called the Lehovec effect [9,26,27]. This large electric field tends to orient the ferroelectric axis in the thin layer perpendicular to the surface and make domain switching more difficult than that inside a crystal [9,28]. The Lehovec effect is neglected in the present study and the present model is suitable only for bulk materials.

The internal stress induced by spontaneous strain change during non-180° domain switching cannot be compensated in a manner similar to the unbalanced polarization. The spontaneous strain change by non-180° switching is similar to the plastic strain in ductile materials [20]. According to the Taylor rule of plasticity [29], a crystal must have at least five slip systems for a polycrystalline to be ductile. In terms of the deformation modes, a ferroelectric crystal is similar to a ferroelastic crystal or a shape memory alloy [30] which has two independent slip systems for the tetragonal case and three for the rhombohedral case. Thus in tetragonal or rhombohedral polycrystalline ferroelectric ceramics, non-180° domain switching generates large internal stresses because of the spontaneous strain change during switching, i.e. such switching could be constrained by neighboring grains and may not occur. However, in orthorhombic ferroelectric ceramics or PZT ceramics near the MPB, where tetragonal and rhombohedral phases coexist, there are five or more independent deformation modes, which render the material ductile. In this case, non-180° domain switching does not generate very large internal stresses from neighboring grains and can be almost completely accomplished [20,30]. The present model excludes these "ductile" polycrystallines and focuses only on the constrained domain-switching phenomenon in tetragonal and rhombohedral ferroelectric ceramics.

Following Hwang et al. [14] and Huber et al. [15], the internal stress induced by non-180° domain switching in tetragonal and rhombohedral ceramics is considered in an Eshelby inclusion manner [31]. For simplicity, each