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Effects of electric fatigue on the butterfly curves of ferroelectric ceramics

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Abstract

The major obstacle for further expansion of the applications of ferroelectric materials is the limited durability and reliability. Recent experiments demonstrated that after a certain number of electric loading cycles, a ferroelectric ceramic may exhibit different strain hysteresis loops on its different positions. Based on a gradual domain switching model, the present paper formulates the strain loops of ferroelectric ceramics induced by electric fatigue. The mechanisms of point defects and domain pinning by space charges are considered in the fatigue model. Our simulations reveal that the spatial distribution and electric property of space charges play a dominant role in the asymmetry of strain loops, while the evolution law of domain switching influences the smoothness of curves. It is found that with the increase in electric loading cycles, both the longitudinal and the transverse strain loops become asymmetric.
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Keywords: Ferroelectric ceramics; Constitutive relation; Electric fatigue; Gradual domain switching; Hysteresis loops

1. Introduction

Ferroelectric materials are of interest for use in smart sensors, actuators and non-volatile memory devices due to their excellent properties, e.g., piezoelectricity, high non-linearity, polarization reversal and hysteresis response [1]. Domain switching and its delayed reverse process under an externally applied electrical or mechanical field have been demonstrated by numerous experimental and theoretical researches to be the source of the hysteretic response of ferroelectrics [2]. The electric field may cause 180° or 90° domain switching and the stress field may cause 90° domain switching. The switching of 180° can induce the direction or sign change in both the polarization vector and the piezoelectric tensor, which result in a hysteresis loop between the electric displacement and the electric field, and a butterfly shaped loop between the strain and the electric field. The butterfly hysteresis loop is symmetric with respect to the positive and negative electric field. However, ferroelectric devices will lose partially its ability of switchable (remanent) polarization under repetitive cyclic electric loading. This phenomenon is referred to as electric fatigue [3], which is a major barrier to the full commercialization of ferroelectric devices, especially in memory applications [4].

Besides the degradation of remanent polarization, the coercive field and the shape of the hysteresis loop may also be affected by electric fatigue. A fatigued ferroelectric sample exhibits an increase in its coercive field, a shrinkage in its polarization hysteresis and an asymmetry in its strain loops [5,6]. Over the last decades, considerable experimental studies have been reported on the asymmetric butterfly curves induced by electric fatigue [6–10]. Weitzing et al. [7], Furuta and Uchino [8] and Lupascu and Verdier [9] found that the right wing of the butterfly curves degraded more rapidly than the left one with the evolution of electric fatigue. However, Nuffer et al. [6] reported a contrary result that the left wing of the butterfly curves degenerated more rapidly than the right. Recently, more systematic experimental studies [10,11] revealed that the asymmetry is location-dependent and that there exist three possible butterfly curves within a same sample. In addition to the butterfly curves (longitudinal strain versus electric field hysteresis), the corresponding reversed butterfly curves (transverse strain versus electric filed hysteresis) also become asymmetric after a large number of electric loading cycles [9].

The underlying physical and mechanical mechanisms of electric fatigue behaviors are extremely complex, and no satisfactory model can be found in the literature to elucidate the observed change in the butterfly curves associated with electric fatigue. Among others, two important types of fatigue mechanisms have been evidenced experimentally. One is associated with point defects [1,12] or microcracking [13], while the other

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is attributed to domain-wall pinning (domain freezing or domain clamping) by space charges through charge injection from electrode [6,14,15]. Accounting for the above mechanisms, some fatigue models [12,16–19] have been developed to describe the deterioration dependence of the remanent polarization on the number of electrical cycles. However, few studies have been made to describe the constitutive curves of ferroelectric ceramics induced by electric fatigue. Recently, Yu et al. [20] proposed a theoretical formulation of the constitutive behaviors of ferroelectric ceramics with electric fatigue effect based on an instantaneous complete domain switching model. The instantaneous complete domain switching assumption may induce several inflexions in the hysteresis loops, which may cause some complexities and difficulties in analytical or numerical analysis of smart structures. As a matter of fact, however, many experimental observations have demonstrated that domain switching is a progressive process accompanied by domain nucleation and domain-wall movement [21]. Considering such a gradual process of domain switching, Yu et al. [22] established a phenomenological constitutive model for ferroelectric ceramics. In this paper, the influence of electric fatigue on the constitutive relations of ferroelectric ceramics is studied by employing the gradual domain switching model and incorporating two typical physical mechanisms (point defects and domain pinning by space charges) of electric fatigue.

2. Fatigue model

2.1. Constitutive equations based on a gradual domain switching model

The constitutive relations of a linear piezoelectric material are given by [1]:

$$D_i = d_{ikl}\sigma_{kl} + k_{ik}E_k, \tag{1}$$

$$\varepsilon_{ij} = s_{ijkl}\sigma_{kl} + d_{kij}E_k, \tag{2}$$

where σ_{kl}, ε_{ij}, D_i, E_k are the stress tensor, the strain tensor, the electric displacement vector and the electric field vector, respectively, s_{ijkl} the elastic compliance tensor, d_{kij} the piezoelectric compliance tensor, and k_{ik} is the dielectric permittivity tensor.

For a ferroelectric material, domain switching leads to a change in the macroscopic remanent strain tensor $\varepsilon^{\mathrm{r}}_{ij}$ and the remanent polarization vector P^{r}_i. Accounting for the non-linear effect of domain switching, the constitutive relations in Eqs. (1) and (2) become [23]:

$$D_i = d_{ikl}\sigma_{kl} + k_{ik}E_k + P^{\mathrm{r}}_i, \tag{3}$$

$$\varepsilon_{ij} = s_{ijkl}\sigma_{kl} + d_{kij}E_k + \varepsilon^{\mathrm{r}}_{ij}. \tag{4}$$

The remanent strain $\varepsilon^{\mathrm{r}}_{ij}$ and the remanent polarization P^{r}_i related to domain switching can be derived by different approaches. Yu et al. [20] developed a constitutive model with effects of electric fatigue by using the assumption of instantaneous complete domain switching. The disadvantage of this assumption is the lack of smoothness in the constitutive curves. In fact, however, domain switching is an evolutionary process with electric loading. Therefore, we will use in the present paper a more accurate yet still simple phenomenological constitutive model in the spirit of gradual domain switching [22], which will be reviewed very briefly in this section.

To study the electric fatigue effect, we consider the case of cyclic electric loading. Ferroelectric ceramics are usually poled by a high electric field prior to their application. It might be assumed, without loss of generality, that the poling is along the x_3-direction. For a fully poled ferroelectric ceramics, the corresponding remanent polarization and remanent strain in this direction are denoted as P^{r}_3 and $\varepsilon^{\mathrm{r}}_{33}$, respectively. The constitutive relations in Eqs. (3) and (4) reduce to

$$D_3 = k_{33}E_3 + \bar{P}^{\mathrm{r}}_3, \tag{5}$$

$$\varepsilon_{33} = \bar{d}_{333}E_3 + \bar{\varepsilon}^{\mathrm{r}}_{33}, \tag{6}$$

where the overbars stand for the current values of the corresponding parameters. The piezoelectric tensor is assumed to be proportional to the remanent polarization [23], i.e.:

$$\bar{d}_{333} = d_{333}\frac{\bar{P}^{\mathrm{r}}}{|\bar{P}^{\mathrm{r}}_{\mathrm{s}}|}, \tag{7}$$

where $\bar{P}^{\mathrm{r}}$ is the current value of the remanent polarization, and $\bar{P}^{\mathrm{r}}_{\mathrm{s}}$ is the fully poled saturation magnitude of the remanent polarization of the ceramic.

According to Landau–Devonshire theory [1], the remanent strain of a ferroelectric ceramic is approximately proportional to the square of the remanent polarization, that is

$$\bar{\varepsilon}^{\mathrm{r}}_{33} = Q_{\mathrm{eff}}(\bar{P}^{\mathrm{r}}_3)^2, \tag{8}$$

where Q_{eff} is the effective electrostriction coefficient. The evolution of remanent polarization $\bar{P}^{\mathrm{r}}_3$ can be derived from the volume fraction of domain switching $f^{\mathrm{E}}_{\mathrm{d}}$ according to the domain nucleation theory by

$$\bar{P}^{\mathrm{r}}_3 = f^{\mathrm{E}}_{\mathrm{d}}P^{\mathrm{r}}_3. \tag{9}$$

The volume fraction $f^{\mathrm{E}}_{\mathrm{d}}$ of domain switching under cyclic electric loading is given approximately by

$$f^{\mathrm{E}}_{\mathrm{d}} = \exp\left(-\frac{b^{\mathrm{E}}}{|E_3 \pm E_{\mathrm{c}}|}\right), \tag{10}$$

where the material constant b^{E} is the activation field under an electric field. The evolution of the volume fraction of domain switching in Eq. (10) in a complete electrical cycle has the following explicit expressions:

$$f^{\mathrm{E}}_{\mathrm{d}}(E_3) = \begin{cases} \exp\left[-\dfrac{b^{\mathrm{E}}}{E_3 + E_{\mathrm{c}}}\right] & -E_{\mathrm{c}} \le E_3 \le E_{\max} \\ \exp\left[\dfrac{b^{\mathrm{E}}}{E_3 + E_{\mathrm{c}}}\right] & -E_{\max} \le E_3 < -E_{\mathrm{c}} \\ \exp\left[\dfrac{b^{\mathrm{E}}}{E_3 - E_{\mathrm{c}}}\right] & -E_{\max} \le E_3 \le E_{\mathrm{c}} \\ \exp\left[-\dfrac{b^{\mathrm{E}}}{E_3 - E_{\mathrm{c}}}\right] & E_{\mathrm{c}} < E_3 \le E_{\max} \end{cases}, \tag{11}$$

where $E_{\max}$ is the initial saturation polarization field.

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