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Author for correspondence:

George J. Weng

e-mail: weng@jove.rutgers.edu

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Theory of electric creep and electromechanical coupling with domain evolution for non-poled and fully poled ferroelectric ceramics

Xiaodong Xia^{1,2}, Yang Wang², Zheng Zhong¹ and George J. Weng²

¹School of Aerospace Engineering and Applied Mechanics, Tongji University, Shanghai 200092, People's Republic of China

²Department of Mechanical and Aerospace Engineering, Rutgers University, New Brunswick, NJ 08903, USA

GJW, 0000-0003-1543-1535

Unlike mechanical creep with inelastic deformation, electric creep with domain evolution is a rarely studied subject. In this paper, we present a theory of electric creep and related electromechanical coupling for both non-poled and fully poled ferroelectric ceramics. We consider electric creep to be a time-dependent process, with an initial condition lying on the D (electric displacement) versus E (electric field) hysteresis loop. Both processes are shown to share the same Gibbs free energy and thermodynamic driving force, but relative to creep, the hysteresis loop is just a field-dependent process. With this view, we develop a theory with a single thermodynamic driving force but with two separate kinetic equations, one for the field-dependent loops in terms of a Lorentzian-like function and the other for the time-dependent D in terms of a dissipation potential. We use the 0° – 90° and then 90° – 180° switches to attain these goals. It is demonstrated that the calculated results are in broad agreement with two sets of experiments, one for a non-poled PIC-151 and the other for a fully poled PZT-5A. The theory also shows that creep polarization tends to reach a saturation state with time and that the saturated polarization has its maximum at the coercive field.

1. Introduction

Smart materials especially piezoelectric–ferroelectric materials have attracted increasing interest due to their intrinsic multi-field coupling effects [1] and their wide range of potential applications such as in micro-electromechanical devices, piezoelectric nanotubes for micro-fluidic systems, and ultrafast-switching room-temperature detectors, actuators and sensors [2–4]. In ferroelectrics, the low-field electromechanical responses are linear, but as the loading reaches certain state, nonlinearity will set in. Nonlinearity is caused by domain switch. Domain switch in ferroelectric single crystals, polycrystals and thin films has been a subject of considerable fascination over the past two decades [5–16]. Comprehensive reviews can be found in Kamlah [17], Landis [18] and Potnis *et al.* [19]. Phase-field simulations along this line have further revealed more in-depth microstructural evolution during domain switch in ferroelectric polycrystals [20–26]. In certain applications such as sensors and actuators, ferroelectric materials are subjected to a constant electric load. In this case, the response can be time-dependent, known as electric creep [27–29]. Unlike the rate-independent domain switch, or mechanical creep with inelastic deformation, electric creep is not a commonly studied subject. At present, only a limited number of experimental investigations [30,31] or phenomenological modelling [32–34] is known to exist. The relationship between the time-dependent evolution of electric polarization and the underlying domain growth process remains to be understood.

In this study, we will develop a micro-continuum theory that could account for the domain evolution to predict the creep polarization of ferroelectric ceramics under an external electric field. Along the way, we will also study the related issue of electromechanical coupling. Creep at a given applied electric field evolves with time, but it starts from a nonlinear state on the curve of electric displacement (D) versus electric field (E), or the hysteresis loop. Such a relation is shown in figure 1*a*, where segment $O \rightarrow A$ represents the nonlinear relation of D versus E for the non-poled ceramic, whereas curve $B' \rightarrow A$ represents the fully poled one. The vertical lines from each curve, such as $B_1 \rightarrow B_2$ or $G_1 \rightarrow G_2$, depict the time-dependent evolution of electric displacement at a constant field. These are followed by the nearly horizontal lines characterized by the dielectric permittivity during instantaneous unloading. Similarly, creep strain is obtained due to electromechanical coupling, which starts from a nonlinear state on the curve of mechanical strain (ε) versus electric field (E), or the butterfly loop in figure 1*b*. In this figure, segment $O \rightarrow Q$ represents the nonlinear relation of ε versus E for the non-poled ceramic, whereas curve $P' \rightarrow J \rightarrow Q$ represents the fully poled one. The vertical lines from each curve, such as $Q_1 \rightarrow Q_2$ or $J_1 \rightarrow J_2$, depict the time-dependent evolution of mechanical strain at a constant field. The objective of the creep theory is to predict $O \rightarrow B_1 \rightarrow B_2$ and $B' \rightarrow G_1 \rightarrow G_2$ for creep polarization, as well as $O \rightarrow Q_1 \rightarrow Q_2$ and $P' \rightarrow J_1 \rightarrow J_2$ for creep strain.

Several issues are involved in this development. The first one is to find the Gibbs free energy and thermodynamic driving force for domain switch. A related issue has been addressed by Li & Weng [7] for a dual-phase system and by Weng & Wong [35] for a rank-2 laminated domain pattern. This approach requires Eshelby's [36] S-tensor in the electromechanical context, but explicit results with arbitrarily oriented lamellar domain in a transversely isotropic medium have been derived by Li [37].

The second issue is the kinetic equations. First we note that, relative to the time-evolving creep process that takes place from state B_1 or G_1 , the time spent at a single point of the D versus E curve from $O \rightarrow B_1$ or from $B \rightarrow G_1$ is simply too short to induce any comparable time-dependent evolution of polarization, for the reason that it takes too many discrete points to constitute a continuous curve. As a result, even though the kinetic equation for the creep portion has to be time-dependent, the kinetic equation from $O \rightarrow B_1$ or from $B \rightarrow G_1$, can be treated as instantaneous and thus simply field-dependent. We shall apply a Lorentzian-like function to describe this field-dependent kinetic process. Subsequently, we will adopt the concept of dissipation potential that has proved to be versatile in the study of material degradation and moisture absorption [38–41] to develop a time-dependent kinetic equation for electric creep.

Figure 1. Schematic of (a) hysteresis loop and (b) butterfly loop with electric creep for the non-poled and fully poled ferroelectric ceramics. (Online version in colour.)

The last one is the homogenization issue. Ferroelectric ceramics are polycrystals, so the best approach to address this issue is to fully consider the orientations of all constituent grains to evaluate their individual domain evolution and then take the orientational average to obtain the overall behaviour of the polycrystalline aggregate. But as there are numerous grains and grain–grain interactions, and the overall properties of the aggregate are not known *a priori*, direct application of Hill’s [42] self-consistent approach for the coupled electromechanical behaviour turns out to be quite a formidable computational task. In a series of works on ferroelectric polycrystals, Su & Weng [43–46] have carried out such polycrystal computations. Many simultaneous, implicit and nonlinear grain interactions had to be synthesized out and the task was indeed formidable. Because our objective here is to develop a simple micro-continuum model to determine the global behaviour of the polycrystals, we will not dwell upon such complex multi-grain calculations but, instead, devise a dual-phase composite model that could properly represent the overall behaviour of polycrystal ceramics. In the dual-phase system, the parent phase will represent the gross behaviour of all unswitched constituent grains, and the product phase will represent the gross behaviour of all grains that have undergone domain switch. We will prove that this dual-phase equivalent system can satisfactorily represent the overall behaviour of multi-grain ceramics by comparing the calculated results with the self-consistent calculations of polycrystals by Su & Weng [43,46] before we proceed to apply it to study the problem of electric creep and electromechanical coupling.

To construct such a dual-phase model, it is almost immediate that the simplest way is to treat the parent phase as a 0° domain and the product phase as the 180° domain. While such a model could capture the evolution of electric displacement under a pure electric field, it could not simultaneously deliver the development of mechanical strain or the butterfly loop. In addition, various phase-field simulations [20–26] have indicated that the 90° domain switching is more often than the 180° switching in both single crystals and polycrystalline ferroelectric ceramics even under a pure electric field. Several polycrystal calculations [43–46] with rhombohedral grains have also revealed the 71° , 110° and 180° switches inside grains. So a better model is to include an intermediate 90° switch, making it a 0° – 90° and then a 90° – 180° switch. This idea was also introduced in [11]. In this way, both electric creep and the broader issue of electromechanical coupling can all be accounted for.

The theory will be so developed. It will then be applied to examine two sets of experiments, one for a non-poled ferroelectric ceramic and the other for a fully poled one. Among several interesting features of the theory, we will demonstrate that the evolution of creep polarization with time tends to reach a saturation state, and that the maximum saturated polarization occurs at the coercive field.

2. Preliminaries of domain switch in the dual-phase equivalent system

To pave the way for the analysis, we first look at the polarization state of non-poled and fully poled ceramics, as depicted in figure 2*a*(i) and *b*(i), respectively. The directions of polarization in the constituent grains are random in (*a*), whereas those in (*b*) are preferably oriented downward following a downward polarization. The overall behaviour of the ceramic in (*a*) is isotropic, whereas in (*b*) it is transversely isotropic. These could be represented by the equivalent isotropic and transversely isotropic phases as shown in figure 2(ii), which correspond to the original state *O* and remnant state *B* (strictly speaking should be *B'*) in figure 1*a*. Then following an upward electric loading, some part of the ceramic will experience a domain switch to lead to *B*₁ and *G*₁, respectively. At this generic state, the ceramic consists of the parent and product phases in the dual-phase system. This representation was originally suggested in Li & Weng [7] and Li [47]. In this dual-phase representation, the parent phase will be taken as phase 0 and the product phase as phase 1, with the volume concentrations *c_r* for the *r*th phase.

For ease of exposition, we shall use a crystal with cubic symmetry to represent the isotropic phase as such a crystal possesses no polarization and its lattice and elastic constants can be easily tailored to reflect those of an isotropic phase. Likewise, we shall use a tetragonal crystal to represent the transversely isotropic phase. These representations are illustrated in figure 2*a,b*. The effective spontaneous strain and polarization of the non-poled state are both zero, and, by taking this state as reference, the effective spontaneous strain and polarization of the poled ceramic can be written as

$$\varepsilon_0^{\text{sp}} = \left\{ \begin{array}{ccc} (a - a_0)/a_0 & 0 & 0 \\ 0 & (a - a_0)/a_0 & 0 \\ 0 & 0 & (c - a_0)/a_0 \end{array} \right\} \quad (2.1)$$

and

$$P_0^{\text{sp}} = [0, 0, -P_s]^T,$$

where a downward polarization is assumed. We take this downward orientation as the 0° domain, for which a subscript 0 is attached to both quantities. In this equation, *a*₀ represents the effective lattice constant of the isotropic phase, and *a* and *c* are those of the transversely isotropic one. *P_s* is its spontaneous polarization, and 'T' stands for the transpose. This transversely isotropic state is depicted by figure 2*b*(ii), while figure 2*a*(ii) represents that of the non-poled state.

In studying the *D* versus *E* relation from *O* to *A* in figure 1 for the non-poled ceramic, the domain switch strain and polarization such as the one depicted in figure 3*a*(ii) are

$$\varepsilon_{c \rightarrow T}^{\text{ds}} = \left\{ \begin{array}{ccc} (a - a_0)/a_0 & 0 & 0 \\ 0 & (a - a_0)/a_0 & 0 \\ 0 & 0 & (c - a_0)/a_0 \end{array} \right\} \quad \text{and} \quad P_{c \rightarrow T}^{\text{ds}} = [0, 0, P_s]^T, \quad (2.2)$$

where the superscript 'ds' denotes the 'domain switch', and the subscript '*c* → *T*' signifies the cubic-to-tetragonal (or isotropic-to-transversely isotropic) transition.

On the other hand for the study of *B'* to *A* for the poled ceramic subjected to an upward electric field, the 0°–90° switch occurs before the coercive field with the 45° domain walls, as the one depicted in figure 3*b*(ii). It is accompanied by the domain-switch strain and polarization

$$\varepsilon_{0^\circ \rightarrow 90^\circ}^{\text{ds}} = \left\{ \begin{array}{ccc} (c - a)/a & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & -(c - a)/a \end{array} \right\} \quad \text{and} \quad P_{0^\circ \rightarrow 90^\circ}^{\text{ds}} = [P_s, 0, P_s]^T. \quad (2.3)$$

To complete the cycle, the additional 90°–180° switch with the –45° domain walls occurs after the coercive field, as the one depicted in figure 3*c*(ii). In figure 3, panels *a*(i), *b*(i) and *c*(i) represents the matrix phase. The corresponding domain-switch strain and polarization are

$$\varepsilon_{90^\circ \rightarrow 180^\circ}^{\text{ds}} = \left\{ \begin{array}{ccc} -(c - a)/a & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & (c - a)/a \end{array} \right\} \quad \text{and} \quad P_{90^\circ \rightarrow 180^\circ}^{\text{ds}} = [-P_s, 0, P_s]^T. \quad (2.4)$$

Figure 2. Schematic of the equivalence procedure: (a) non-poled ceramic to an isotropic crystal without polarization; (b) fully poled ceramic to a transversely isotropic one with a polarization. (Online version in colour.)

3. Constitutive equations of the parent and product phases

There are several ways to write the linear constitutive relations of a ferroelectric material, but to study domain switch under an external electric field and mechanical stress, the best one is

$$\left. \begin{aligned} \varepsilon_{ij} &= s_{ijkl}^{(E)} \sigma_{kl} + d_{nij} E_n + \varepsilon_{ij}^{\text{sp}} \\ D_m &= d_{mkl} \sigma_{kl} + \kappa_{mn}^{(\sigma)} E_n + P_m^{\text{sp}} \end{aligned} \right\} \quad (3.1)$$

and

where σ_{ij} and ε_{ij} are stress and strain, E_m and D_m are electric field and electric displacement, $s_{ijkl}^{(E)}$ is the elastic compliance tensor at a constant electric field, $\kappa_{mn}^{(\sigma)}$ is the dielectric permittivity tensor at a constant stress, and d_{mkl} is the piezoelectric compliance tensor. The spontaneous strain $\varepsilon_{ij}^{\text{sp}}$ and polarization P_m^{sp} must be added when the ceramic is in a preferred polarized state. For computational convenience, this set of coupled electromechanical constitutive equations can be written in a unified form as

$$Y_i = M_{ij} X_j + Y_i^{\text{sp}}, \quad i, j = 1 \sim 9, \quad (3.2)$$

where X_i represents the electromechanical load, Y_i the response, M_{ij} the electromechanical compliance matrix and Y_i^{sp} the spontaneous strain and polarization. In terms of Voigt's and Nye's

Figure 3. Schematic of domain switch in the equivalent system subjected to electric loading: (a) isotropic-to-transversely isotropic domain switch in the non-poled ferroelectric ceramic, (b) 0°–90° and (c) 90°–180° transversely isotropic-to-transversely isotropic domain switch in the fully poled ferroelectric ceramic. Panels *a(i)*, *b(i)* and *c(i)* represent the matrix phase. (Online version in colour.)

contracted notations, they are

$$\left. \begin{aligned} X &= [\sigma_1, \sigma_2, \sigma_3, \sigma_4, \sigma_5, \sigma_6, E_1, E_2, E_3]^T, \\ Y &= [\varepsilon_1, \varepsilon_2, \varepsilon_3, \varepsilon_4, \varepsilon_5, \varepsilon_6, D_1, D_2, D_3]^T \\ \text{and} \quad Y^{\text{sp}} &= [\varepsilon_1^{\text{sp}}, \varepsilon_2^{\text{sp}}, \varepsilon_3^{\text{sp}}, \varepsilon_4^{\text{sp}}, \varepsilon_5^{\text{sp}}, \varepsilon_6^{\text{sp}}, D_1^{\text{sp}}, D_2^{\text{sp}}, D_3^{\text{sp}}]^T. \end{aligned} \right\} \quad (3.3)$$

Conversely, equation (3.2) can be written as

$$X_i = L_{ij}(Y_j - Y_j^{\text{sp}}), \quad i, j = 1 \sim 9, \quad (3.4)$$

where $L_{ij} = [M_{ij}]^{-1}$ is the unified electromechanical moduli matrix.

It follows that, for the equivalent cubic-to-tetragonal domain switch in the non-poled ceramic, the electromechanical compliance tensor of the parent phase is given as

$$M_0 = [s_{11}, s_{12}, s_{44}, \kappa_{11}], \quad \text{with } s_{44} = 2(s_{11} - s_{12}), \quad (3.5)$$

and for the product phase

$$M_1 = [s_{11}, s_{12}, s_{13}, s_{33}, s_{44}, s_{66}, d_{31}, d_{33}, d_{15}, \kappa_{11}, \kappa_{33}], \quad \text{with } s_{66} = 2(s_{11} - s_{12}), \quad (3.6)$$

in Voigt–Nye notations, where the superscripts ‘ E ’ and ‘ σ ’ in s_{ijkl} and κ_{ij} in equation (3.1) have been dropped for brevity. In addition, we can write $\Delta M = M_1 - M_0$ to represent the difference of the electromechanical compliance between the product and the parent phase, and $L_0 = M_0^{-1}$, to represent the electromechanical moduli matrix of the parent phase.

4. Gibbs free energy and thermodynamic driving force

To derive the kinetic equations, Gibbs free energy and thermodynamic driving force of the dual-phase system are needed. The former can be evaluated from [35,36,48,49]

$$\begin{aligned} G(\bar{X}, c_1) = & \frac{1}{2} \int_V \sigma_{ij}(\varepsilon_{ij} - \varepsilon_{ij}^{\text{ds}}) dV - \int_S \bar{F}_i(u_i - u_{i(0)}^{\text{sp}}) dS \\ & + \frac{1}{2} \int_V E_i(D_i - P_i^{\text{ds}}) dV - \int_S \bar{\phi}(D_i - P_{i(0)}^{\text{sp}}) n_i dS, \end{aligned} \quad (4.1)$$

where c_1 is the volume concentration of the product phase; $u_{i(0)}^{\text{sp}}$ and $P_{i(0)}^{\text{sp}}$ are the spontaneous displacement and polarization of the parent phase, respectively, and $\bar{F}_i$ and $\bar{\phi}$ are the mechanical traction and electric potential on the boundary of the system, respectively. On the other hand, the free energy of the reference state (i.e. $c_1 = 0$ and no domain switch) is given by

$$\begin{aligned} G_0(\bar{X}) = & \frac{1}{2} \int_V \sigma_{ij} \varepsilon_{ij} dV - \int_S \bar{F}_i(u_i - u_{i(0)}^{\text{sp}}) dS + \frac{1}{2} \int_V E_i D_i dV - \int_S \bar{\phi}(D_i - P_{i(0)}^{\text{sp}}) n_i dS \\ = & -\frac{1}{2} \bar{X}^T Y^0 - \bar{X}^T Y_{(0)}^{\text{sp}}, \end{aligned} \quad (4.2)$$

where $\bar{X} = (\bar{\sigma}, \bar{E})^T$ is the unified external loading given by $\bar{F}_i$ and $\bar{\phi}$, and $Y^0 = M_0 \bar{X}$ is the electromechanical response of the parent phase under the same external $\bar{X}$.

Then the difference of the free energies is given by

$$\Delta G(\bar{X}, c_1) = G(\bar{X}, c_1) - G_0(\bar{X}). \quad (4.3)$$

It is this difference that ultimately gives rise to the thermodynamic driving force. To obtain an explicit expression for ΔG , the domain shape needs to be specified. With a general ellipsoidal shape, it can be shown that [7,35,49,50]

$$\Delta G(\bar{X}, c_1) = -\frac{1}{2} c_1 [\bar{X}_1^T Y^{\text{ds}} + \bar{X}^T (Y^{\text{ds}} + Y^*)], \quad (4.4)$$

where $\bar{X}_1$ is the average electromechanical field in the product phase, given by

$$\bar{X}_1 = B_1 Q [\bar{X} - c_0 L_0 (I - S) Y^{\text{ds}}], \quad (4.5)$$

and S is Eshelby’s S -tensor, and Y^{ds} and Y^* are given by

$$\left. \begin{aligned} Y^{\text{ds}} &= Y_{(1)}^{\text{sp}} - Y_{(0)}^{\text{sp}} \\ Y^* &= \Delta M \bar{X}_1. \end{aligned} \right\} \quad (4.6)$$

The quantity Y^* is the Eshelby-type equivalent electromechanical eigenfield introduced into the inclusion regions so that L_1 can be replaced by L_0 to yield the same $\bar{X}_1$. Furthermore,

$$\left. \begin{aligned} B_1 &= [I + L_0 (I - S) \Delta M]^{-1} \\ Q &= (c_1 B_1 + c_0 I)^{-1}, \end{aligned} \right\} \quad (4.7)$$

and

and I is the symmetric identity matrix. A commonly observed domain morphology in BaTiO_3 crystals is a lamellar structure [51]. Its explicit expressions can be found in [37] and in the appendix of the electronic supplementary material.

It follows that ΔG is given by

$$\Delta G(\bar{X}, c_1) = -\frac{1}{2}(\bar{X}^T A \bar{X} + \bar{X}^T B Y^{\text{ds}} + Y^{\text{ds}T} C Y^{\text{ds}}), \quad (4.8)$$

where

$$A = \Delta M B_1 Q, \quad B = I - c_0 \Delta M B_1 Q L_0 (I - S) + (B_1 Q)^T \quad \text{and} \quad C = -c_0 [B_1 Q L_0 (I - S)]^T. \quad (4.9)$$

This in turn provides the thermodynamic driving force of domain switch at a given external load $\bar{X}$ by taking its negative partial derivative with respect to c_1 [7,35],

$$f_{\text{driv}} = - \left. \frac{\partial \Delta G(\bar{X}, c_1)}{\partial c_1} \right|_{\bar{X}} - f_s = \frac{1}{2}(\bar{X}^T A_f \bar{X} + \bar{X}^T B_f Y^{\text{ds}} + Y^{\text{ds}T} (C_f - C_s) Y^{\text{ds}}), \quad (4.10)$$

where

$$\left. \begin{aligned} A_f &= A - c_1 \Delta M B_1 Q (B_1 - I) Q, \\ B_f &= B + c_1 \{ -[B_1 Q L_0 (I - S)]^T + \Delta M B_1 Q [I + c_0 (B_1 - I) Q] L_0 (I - S) \} \\ C_f &= C + c_1 \{ [B_1 Q (I + c_0 (B_1 - I) Q)] L_0 (I - S) \}^T, \end{aligned} \right\} \quad (4.11)$$

and

f_s is the resistance force due to the depolarization field, which must be subtracted to provide a positive driving force. It is defined as

$$f_s = \frac{1}{2} Y^{\text{ds}T} C_s Y^{\text{ds}} = \frac{1}{2} Y^{\text{ds}T} \{ B_1 Q L_0 (I - S) \}^T Y^{\text{ds}}. \quad (4.12)$$

From equation (4.11), it can be seen that the first term of the driving force with A_f comes from the electromechanical heterogeneity, ΔM , the second term with B_f comes from the energy consumption during domain switch and the third one with C_f is attributed to the self-energy due to the existence of electromechanical eigenfield, Y^{ds} , in the product phase.

5. Overall response and kinetic equations

With this thermodynamic driving force, we can derive the kinetic equation for the evolution of product volume concentration, c_1 , to calculate the nonlinear D versus E and ε versus E curves and the evolution of D and ε as a function of time under creep condition.

(a) Overall response of the ceramics

From the dual-phase homogenization theory, it is known that the overall electromechanical response of a dual-phase ferroelectric material can be calculated from [35]

$$\bar{Y} = M_0 \bar{X} + c_1 (Y^{\text{ds}} + Y^*) + Y_{(0)}^{\text{sp}}. \quad (5.1)$$

For the problem under an upward electric field, this provides the overall electric displacement and axial strain in the 3-direction for the non-poled ceramic

$$\left. \begin{aligned} \bar{D}_3 &= \kappa_{33}^{(\sigma)} \bar{E}_3 + c_1 (P_3^{\text{ds}} + P_3^*) + P_{(0)3}^{\text{sp}} \\ \bar{\varepsilon}_3 &= c_1 (\varepsilon_3^{\text{ds}} + \varepsilon_3^*) + \varepsilon_{(0)3}^{\text{sp}}, \end{aligned} \right\} \quad (5.2)$$

and

and for the fully poled one

$$\left. \begin{aligned} \bar{D}_3 &= \kappa_{33}^{(\sigma)} \bar{E}_3 + c_1 (P_3^{\text{ds}} + P_3^*) + P_{(0)3}^{\text{sp}} \\ \bar{\varepsilon}_3 &= d_{33} \bar{E}_3 + c_1 (\varepsilon_3^{\text{ds}} + \varepsilon_3^*) + \varepsilon_{(0)3}^{\text{sp}}. \end{aligned} \right\} \quad (5.3)$$

and

In both the cases, it is clear that volume concentration of the product phase, c_1 , is the figure of merit. Its evolution leads to the variation of the overall electric displacement, $\bar{D}_3$ and strain, ε_3

under an applied electric field, $\bar{E}_3$, which will be written henceforth as D , ε and E , respectively, for simplicity.

The need to know c_1 at an applied E and time t calls for the kinetic equations of domain growth.

(b) Kinetic equation for the field-dependent nonlinear D versus E and σ versus E curves

Before we can consider electric creep at B_1 (or Q_1) for the non-poled case and at G_1 (or J_1) for the fully poled one in figure 1a,b, we have to arrive at B_1 (or Q_1) from O or at G_1 (or J_1) from B' or P' first. This nonlinear path, as explained in the Introduction, can be taken to be field-dependent. In this case, the value of c_1 depends only on the load, that is on the thermodynamic driving force. It has been found that the corresponding kinetic equation can be expressed in a Lorentzian-like function [35]

$$\frac{dc_1}{df_{\text{driv}}} = \frac{\eta}{(1 - f_{\text{driv}}/\lambda_1)^2 + (f_{\text{driv}}/\lambda_2)^2}, \quad (5.4)$$

where λ_1 , λ_2 and η are material constants that control the growth of c_1 with respect to the thermodynamic driving force. As the applied field E increases, the driving force f_{driv} also increases in accordance with equation (4.10), and this leads to the increase of c_1 following this kinetic equation. The new c_1 , when substituted into equation (5.2) or (5.3), leads to the increase of the overall electric displacement, D , and strain, ε . This generates the nonlinear paths OB_1 and $B'G_1$ in figure 1a as well as OQ_1 and $P'J_1$ in figure 1b. As this process is rate-independent, this c_1 will be designated as 'static' and denoted as $c_1^{(s)}$.

(c) Kinetic equation for the time-dependent electric creep

Now at B_1 or G_1 , the volume concentration, c_1 , of the product phase will continue to evolve due to electric creep. At a generic state of creep, the total c_1 is then the sum of the initial $c_1^{(s)}$ and this creep-induced part, $c_1^{(c)}$,

$$c_1(t) = c_1^{(s)} + c_1^{(c)}(t). \quad (5.5)$$

The static $c_1^{(s)}$ has already been given by equation (5.4). To derive the creep part, a rate-dependent kinetic equation is needed.

To this end, we note that $c_1^{(c)}$ not only depends on time but also on the applied electric field E . A plausible way to start is to write it in terms of two state variables

$$c_1^{(c)}(E, t) = \chi(E)\alpha(t), \quad (5.6)$$

where $\alpha(t)$ is a dimensionless parameter with the interval from 0 to 1 that indicates the extent of creep. The limiting conditions are $\alpha = 0$ and $\alpha = 1$, which represent the starting and saturated states, respectively. Variable $\chi(E)$ reflects the saturated magnitude of $c_1^{(c)}(E, t)$ at E as $t \rightarrow \infty$. Since domain evolution is known to be the fastest at the coercive field, E_c , it can be set as

$$\chi(E) = pe^{-(E-E_c)^2/q}, \quad (5.7)$$

which exhibits a maximum at E_c , as shown in the electronic supplementary material figure S1, where p and q are constants governing its magnitude.

It is now necessary to derive the evolution of the internal variable $\alpha(t)$ that is, $\dot{\alpha}(t)$, as a function of f_{driv} . As electric creep is an irreversible dissipation process, the principle of non-equilibrium thermodynamics

$$-\left. \frac{\partial \Delta G(\bar{X}, c_1)}{\partial t} \right|_{\bar{X}} dt \geq 0 \quad (5.8)$$

must be satisfied, where equality holds only for the reversible process. As the thermodynamic system evolves towards equilibrium, the change of ΔG continues to decrease. In the rate-dependent process, a dissipation potential ξ can be defined as [39,52]

$$\xi = - \left. \frac{\partial \Delta G(\bar{X}, c_1)}{\partial t} \right|_{\bar{X}}. \quad (5.9)$$

Several forms of dissipation potential have been proposed in the literature. Here we suggest

$$\xi = \frac{h\chi(E)\dot{\alpha}^2}{(1-\alpha)^n} \quad (5.10)$$

for the electric creep. This choice is motivated by the creep potential for wood cell by Pan & Zhong [38]. It is also a modified version of the dissipation models proposed by Rajagopal *et al.* [52] for the degradation of polymers, and by Pan & Zhong [39] for the degradation of natural fibres. Here h is a constant governing the creep speed, and parameter n is referred as a rate sensitivity index.

Now we note that the rate of Gibbs free energy at a constant E can be rewritten as

$$- \left. \frac{\partial \Delta G(\bar{X}, c_1)}{\partial t} \right|_{\bar{X}} = - \frac{\partial \Delta G(\bar{X}, c_1)}{\partial c_1} \dot{c}_1 = f_{\text{driv}} \dot{c}_1, \quad \text{or } \xi = f_{\text{driv}} \dot{c}_1. \quad (5.11)$$

With $\dot{\alpha} = \dot{c}_1/\chi$ from equation (5.6), and with equations (5.11) and (5.10), we arrive at the rate-dependent kinetic equation for electric creep

$$\dot{c}_1 = Q(E)f_{\text{driv}}, \quad \text{with } Q(E) = \frac{(1-\alpha)^n \chi(E)}{h}, \quad (5.12)$$

where $Q(E)$ is a load-dependent kinetic coefficient.

(d) Influence of an axial compression

As the free energy and thermodynamic driving force both include the mechanical term, the theory can also be applied to study the effect of an applied stress. We shall specifically consider the condition of an axial compression on the poled ceramic, which is of considerable interest and some experimental data are also available. In this case, the static kinetic equation for the 90° switch is also given by the Lorentzian-like function equation (5.4). The kinetic equation for the time-dependent creep can also be expressed by equation (5.12), but the $\chi(E)$ function must be replaced by $\chi(\sigma_3)$, where σ_3 is the magnitude of axial compression which for brevity will be written as σ . This function may be taken as

$$\chi(\sigma) = p e^{-(\sigma - \sigma_c)^2/q}, \quad (5.13)$$

where σ_c is the coercive stress field, and p and q are constants governing its magnitude. Equation (5.12) is now replaced by

$$\dot{c}_1 = Q(\sigma)f_{\text{driv}}, \quad \text{with } Q(\sigma) = \frac{(1-\alpha)^n \chi(\sigma)}{h}, \quad (5.14)$$

where parameters α and h must be re-calibrated for the condition of mechanical loading. Then with this $\dot{c}_1$, its evolution with time can be determined. The overall electromechanical response, that is, $\bar{D}_3$ and $\bar{\epsilon}_3$, can be determined from equation (5.1) again. For the non-poled ceramic, we have

$$\left. \begin{aligned} \bar{D}_3 &= c_1(P_3^{\text{ds}} + P_3^*) + P_{(0)3}^{\text{sp}} \\ \bar{\epsilon}_3 &= s_{33}^{(E)} \bar{\sigma}_{33} + c_1(\epsilon_3^{\text{ds}} + \epsilon_3^*) + \epsilon_{(0)3}^{\text{sp}} \end{aligned} \right\} \quad (5.15)$$

and

and for the fully poled one

$$\left. \begin{aligned} \bar{D}_3 &= d_{33} \bar{\sigma}_3 + c_1(P_3^{\text{ds}} + P_3^*) + P_{(0)3}^{\text{sp}} \\ \bar{\epsilon}_3 &= s_{33} \bar{\sigma}_3 + c_1(\epsilon_3^{\text{ds}} + \epsilon_3^*) + \epsilon_{(0)3}^{\text{sp}} \end{aligned} \right\} \quad (5.16)$$

and

The kinetic equation in equation (5.12) or (5.14), $\dot{c}_1 = Qf_{\text{driv}}$, actually resembles the well-known time-dependent Ginzburg–Landau (TDGL) kinetic equation for the evolution of order parameter τ_i [53,54],

$$\dot{\tau}_i = -L_{ij} \frac{\delta G}{\delta \tau_j}, \quad (5.17)$$

where $-\delta G/\delta \tau_j$ is the thermodynamic driving force that is conjugate to the growth of τ_j in the system, and L_{ij} are the load-independent anisotropic kinetic coefficients. In our creep problem, the order parameter is c_1 , and our kinetic coefficient Q is isotropic and load-dependent. The TDGL theory has been widely used in phase-field simulations of ferroelectric polycrystals [21–26] and ferromagnetic materials [55].

With the initial condition, $\alpha = 0$ the kinetic equation (5.12) or (5.14) provides the rate $\dot{c}_1$, which is also $\dot{c}_1^{(c)}$, that can be used to update α from equation (5.6). The evolution of the product domain $c_1(E, t)$ (or $c_1(\sigma, t)$) with respect to time at a given E (or σ) can be obtained with a forward computational scheme.

At this point, the theory of electric creep and electromechanical coupling is completely established. It includes both the field-dependent hysteresis and butterfly loops, and the subsequent time-dependent electric creep and creep strain at an applied E or σ .

6. Results and discussion

To place the developed theory in perspective, we now apply it to calculate the creep polarization and strain of the equivalent cubic-to-tetragonal domain switch in the non-poled ferroelectric ceramic, and those of the equivalent tetragonal-to-tetragonal 90° domain switches in the fully poled ferroelectric ceramic. Theoretical predictions will be compared with the experimental data of Zhou & Kamlah [27] and Viola *et al.* [29], respectively, to validate the present model.

But before we proceed, we will demonstrate, as promised, that the calculations from the dual-phase equivalent systems could well represent the results from the polycrystal calculations. This comparison is shown in figure 4a and b, respectively, for the non-poled and fully poled PLZT 8/65/35 ceramics. The polycrystal results were calculated by Su & Weng [43,46] based on Hill's self-consistent scheme [42]. The material properties used in the validation are listed in the electronic supplementary material, table S1). It is seen that the simple dual-phase model could well capture the calculated results of the far more complex polycrystal model.

Now in accordance with the experimental conditions of Zhou & Kamlah [27] and Viola *et al.* [29], the spontaneous properties of the product phase and coercive field of these two ceramics are given as

- (i) for non-poled ferroelectric ceramic (PIC-151):

$$\varepsilon^{\text{SP}} = [-0.003, -0.003, 0.00385, 0, 0, 0]^T, \quad P^{\text{SP}} = 0.35 \text{ C m}^{-2}, \quad E_c = 1.0 \text{ kV mm}^{-1} \text{ and}$$

- (ii) for fully poled ferroelectric ceramic (PZT-5A):

$$\varepsilon^{\text{SP}} = [-0.00134, -0.00134, 0.002, 0, 0, 0]^T, \quad P^{\text{SP}} = 0.35 \text{ C m}^{-2}, \quad E_c = 1.7 \text{ kV mm}^{-1}.$$

In addition, their elastic, piezoelectric and dielectric constants of the parent and product domains are listed in the electronic supplementary material, table S2, and other material constants for the growth rates of domain walls are also listed in the electronic supplementary material, table S3.

(a) Field- and time-dependent properties of the non-poled ferroelectric ceramic PIC-151

First we compare the calculated results with the experimental data of a non-poled ferroelectric ceramic (PIC-151) conducted by Zhou & Kamlah [27]. In their experiment, the sample PIC-151 was originally set without polarized direction, and subjected to full cycles of electric

Figure 4. The validation for the dual-phase equivalent method for a polycrystal system: (a) non-poled PLZT 8/65/35 and (b) fully poled PLZT 8/65/35 ceramic. (Online version in colour.)

Figure 5. The field-dependent electric displacement versus electric field relation of the non-poled ferroelectric ceramic PIC-151 subjected to the electric loading along the positive 3-direction. (Online version in colour.)

loading. The nonlinear D versus E relations of the ceramic is calculated by equation (5.2), with the kinetic equation (5.4). Figure 5 depicts the calculated nonlinear D versus E curve and the experimental data of this non-poled ceramic. As the electric loading increases from zero, the low-field response is seen to be within the linear regime, but soon nonlinearity sets in. As the loading reaches the coercive field around $1.0 \text{ kV}^{-1} \text{ mm}$, the curve starts to display the sigmoidal shape. Beyond this range, the process of domain switch slows down and the response eventually returns to the linear range. Overall the theoretical curve is entirely consistent with the experimental data.

This curve then serves as the initial condition of the electric and mechanical creep process at three applied electric fields, $E = 0.5, 1.0$ and $2.0 \text{ kV}^{-1} \text{ mm}$; these three holding levels correspond to $E < E_c$, $E = E_c$ and $E > E_c$, respectively. The experimental data and the corresponding calculated, time-dependent, creep polarizations and strains are shown in figure 6a and b, respectively. The experimental data are also seen to be well captured. We also find that both creep polarization and creep strain of the ceramic are the highest at the coercive field, and that a higher field does not necessarily generate a higher electric or mechanical creep. These phenomena indicate that the kinetic process is sensitive to the electric field as reflected in the χ function. All the curves eventually reach their saturation state. The maximum of the saturated creep polarization occurs at the coercive field. This phenomenon can be attributed to the added domain switch near the coercive field. The corresponding evolution of the internal variable α and product concentration c_1 can be seen in the electronic supplementary material, figure S2.

Figure 6. The evolution of (a) creep polarization and (b) creep strain for the non-poled ferroelectric ceramic PIC-151 [27] with respect to time when subjected to constant electric loading. (Online version in colour.)

Figure 7. The field-dependent (a) electric displacement and (b) strain versus electric field relation of the fully poled ferroelectric ceramic PZT-5A subjected to the electric loading along positive 3-direction. (Online version in colour.)

(b) Field- and time-dependent properties of the fully poled ferroelectric ceramic PZT-5A

Next we compare the predicted results with experiments of a fully poled ferroelectric ceramic (PZT-5A) conducted by Viola *et al.* [29]. In their experiment, the sample PZT-5A is originally set polarized along the negative 3-direction, and subjected to full cycles of electric loading. As with the non-poled case, the time-independent nonlinear D versus E and ε versus E relations of the ceramic are calculated first by equation (5.3) with the kinetic equation (5.4). Figure 7a shows the calculated D versus E results and the experimental data. Figure 7b shows the calculated ε versus E results and the experimental data, which display a butterfly shape. The calculated nonlinear D versus E and ε versus E relations are in agreement with the test data.

Then three holding levels of electric field are selected for the creep tests: $E = 1.2, 1.7$ and 2.5 kV mm^{-1} that also correspond to $E < E_c$, $E = E_c$ and $E > E_c$. Figure 8a,b depicts the corresponding evolution of creep polarization and creep strain for this fully poled ceramic with respect to time. As can be seen, the predictions from the present model are also in close agreement with the experiment. All these curves eventually also reach their saturation state. A saturated electric displacement is known to have significant effect on the initiation and propagation of cracks in piezoelectric ceramics [15]. It is observed that creep strain is negative before the coercive field but positive afterward, and it is also evident that the maximum of saturated creep polarization again occurs at the coercive field. The corresponding evolution of α and c_1 can also be seen in the electronic supplementary material, figure S3.

Figure 8. The evolution of (a) creep polarization and (b) creep strain for the fully poled ferroelectric ceramic PZT-5A [29] with respect to time when subjected to constant electric loading. (Online version in colour.)

Figure 9. (a) The field-dependent strain versus stress field relation of the non-poled ferroelectric ceramic PIC-151 subjected to the compressive stress loading along the 3-direction, (b) the evolution of creep strain for the non-poled ferroelectric ceramic PIC-151 [28] with respect to time when subjected to constant compressive stress. (Online version in colour.)

(c) The effect of stress on the non-poled PIC-151

We have also applied the theory outlined in §5d to calculate the effect of an axial compression on the non-poled PIC-151. Figure 9a depicts the calculated stress–strain relation. The low-field response is seen to be within the linear regime, but soon nonlinearity sets in. As stress reaches the coercive field at 50 MPa, the curve starts to display the sigmoidal shape. Beyond this range the process of domain switch slows down and the response returns to the linear range. The overall stress–strain curve is consistent with the experimental data.

This curve then serves as the initial condition of the creep process at $\sigma = 25$ and 50 MPa. The experimental data and the calculated creep strains are shown in figure 9b. The data are seen to be well captured too. We also find that creep strain of the ceramic is highest at the coercive stress. This phenomenon is consistent with the electric creep behaviour under electric field.

7. Concluding remarks

Electric creep of ferroelectric ceramics is not a commonly studied subject, but its occurrence can have direct impact to the performance of piezoelectric/ferroelectric devices. Similar effects can also be associated with fatigue and ageing properties. Their influence in general is not limited to electrical performance but can extend into mechanical reliability. For instance, fatigue crack growth can compromise the intrinsic electromechanical properties and quick time response of

the devices to external excitations [56,57], and ageing of ferroelectric ceramics can lead to time-dependent instability and degradation of dielectric constant, piezoelectricity and ferroelectricity [58,59]. Much of electric creep, fatigue and ageing of ferroelectric ceramics remain to be explored.

In this study, we have developed a theory of electric creep and related electromechanical coupling for both non-poled and fully poled ceramics. The development has involved three key components. The first one is the Gibbs free energy and thermodynamic driving force of a dual-phase system, the second one is the kinetic equations and the last one is the homogenization scheme that permits the calculation of overall electromechanical response under an applied field.

The theory has been put to test against two separate sets of experimental data, one for a non-poled PIC-151 and the other for a fully poled PZT-5A. It is demonstrated that the theory could capture the test results well. Among other interesting features of the predictions, it shows that the time-dependent evolution of creep polarization is strongly dependent on the applied field, and that it tends to reach a saturation state with time. The maximum saturated electric polarization is found to occur at the coercive field.

We conclude by saying that, despite the simplicity of the dual-phase equivalent system, this theory is based on the principles of micromechanics, irreversible thermodynamics, dissipation potential and domain evolution. Unlike traditional phenomenological models, this micro-continuum model has directly brought the underlying physical processes up to the overall macroscopic level in a quantitative way. The simple structure of the theory could also render it widely useful for the study of both non-poled and fully poled ferroelectric ceramics.

Data accessibility. All the data are displayed in the tables and figures.

Authors' contributions. X.X. carried out the analytic modelling, analysed the data and drafted the manuscript; Y.W. participated in data analysis and discussions, and revised the manuscript; Z.Z. designed the analytic modelling, analysed the data and revised the manuscript; G.J.W. designed the analytic modelling, analysed the data and finalized the manuscript. All authors gave final approval for publication.

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