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## Poling of Ferroelectric Ceramics

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**Abstract:** In this study, we use the model Basic Equations to deduce a one-dimensional model for the description of the poling of ferroelectrics ceramics. This is built within the scheme of the thermo dynamical theory of internal variables. The model produces both plastic and electric hysteresis effects in the form of “plasticity”, i.e., rate-independent evolution equations for the plastic strain and the residual electric polarization and both mechanical and electric hardenings. The influence of stresses on ferroelectrics hysteresis loops through piezoelectricity and electrostriction is a natural outcome of this model. Some simple experimental methods for the determination of the material coefficients of the considered ceramics are suggested.

**Key words:** Ferroelectrics, phenomenological model, poling of ceramics, constitutive equations, internal variables

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### INTRODUCTION

It is only recently that general approaches have been presented for the description of coupled electromechanical behaviors of electrically polarized (as also magnetized) deformable solids and, more particularly, crystals. While accounting for some microscopic features, these works offer a fully deductive, phenomenological scheme. Among these, the approach by Collet and Maugin (1974), Maugin and Eringen (1977), Maugin (1980), Maugin and Pouget (1980) and Maugin (1988), following the pioneering works of Toupin, Eringen, Tiersten and others, is based on the fruitful notion of principle of virtual power (for finite velocities and without thermodynamical restrictions). Basing on previous model Bassiouny *et al.* (1988a, b) and Bassiouny (2005) were a framework for phenomenological constitutive laws capable of handling complex multi-axial electromechanical loading and making use of concepts of phenomenological plasticity theory including yield surfaces and isotropic and kinematic hardening was developed and the polarization response of the material during initial poling is characterized and hysteresis loops during cyclic electrical loading were given. This model presents a study to deduce a one-dimensional thermodynamical model for the description of the poling of ferroelectric ceramics.

A rather complete phenomenological constitutive model for ferroelectric materials subject to switching has been developed by Kamlah and Tsakmakis (1999) and Kamlah and Böhle (2001). This constitutive model is based on a framework akin to that of Bassiouny *et al.* (1988a, b) and uses a number of non-linear functions to represent behavior during switching.

The essential ferroelectric property is the possibility of reversal, or change in orientation, of the polarization direction by an electric field. The hysteresis loop is a manifestation of this key property. Additionally, ferroelectricity is characterized by a specific property: mechanical and electric loads of sufficient magnitude may change the dipole orientation of unit cells, leading to domain switching and domain wall motion (Kamlah and Liskowsky, 2005).

The key physical phenomena governing the non-linear response of ferroelectrics below the Curie temperature is switching of the remnant electrical polarization of crystalline material between distinct polarization states. When the remnant polarization switches between distinct states, the switch is in some cases accompanied by a change of shape of the unit cell, giving rise to remnant strain. Consequently, switching may be driven by stress (termed ferroelastic switching) or by electric field (Huber and Fleck, 2001).

The poling process may differ from one substance to another. Mechanical stresses may play a role in the poling. For instance, some Perovskite ferroelectrics such as  $\text{BaTiO}_3$ , it is often straightforward to obtain a single domain plate by application of electric field alone, while for other, such as  $\text{PbTiO}_3$  it is easier to remove a-axis type domain with an applied stress in the plane of the plate while orienting c-axis or 180 domains with a field (Dungan and Storz, 1985).

An alternative procedure for poling ferroelectrics is to switch reverse domains below the Curie temperature with electric fields higher than the coercive field. For several materials high coercivity allows poling in this way only near to  $T_c$  but in most materials electrical poling may be

achieved by cooling a crystal from the paraelectric phase into the ferroelectric phase in an applied electric field parallel to the polar crystallographic axis under constant condition of the electric field (Lines and Glass, 1977).

This model is motivated by the fact that the effective number of dipoles aligned in the direction of the applied field is altered as domains switch and this number obeys a rate law which depends nonlinearly on the electric field.

Hysteresis effects typical of poling may be due to rate-independent effects (like in the mechanical behavior known as “plasticity”). This is explored here for the electric behavior (which coupled either through piezoelectricity or electrostriction to stresses) thanks to the introduction of electric internal variables (Maugin, 1981 and Collet, 1985). Plastic behavior will be involved simultaneously with the electric properties. The Model produces both plastic and electric-hysteresis effects in the form of “plasticity”, i.e., rate-independent, evolution equations for the plastic strain, the residual electric polarization and both mechanical and electric hardenings.

**Three-dimensional constitutive equations:** A phenomenological model capable of displaying electromechanical hysteresis effects and domain switch can be constructed by assuming first a specific dependence of the free energy  $\Psi$  per unit volume on a set of state variables, including an internal variable  $\underline{\Pi}^{int}$  which describes an internal electric polarization.

To simplify, assume that there are no variables, which are related to microscopic effects of plasticity and irreversible polarization such as densities of dislocation and domain wall motion.

Assume that the electric polarization  $\underline{\Pi}$  is the sum of a reversible part  $\underline{\Pi}^r$  and a residual contribution  $\underline{\Pi}^R$  that is, (compare with Bassiouny, 2005):

$$\underline{\Pi}^r = \underline{\Pi} - \underline{\Pi}^R \tag{1}$$

Besides the assumption 1 of decomposition of the total polarization, we assume that the total strain tensor  $\underline{\epsilon}$  is also the sum of the reversible part  $\underline{\epsilon}^e$  and a plastic deformation  $\underline{\epsilon}^p$ , (we assume that the deformation occurs in the frame of the theory of small perturbations), thus

$$\underline{\epsilon}^e = \underline{\epsilon} - \underline{\epsilon}^p \tag{2}$$

Consider the following dependence of the free energy  $\Psi$  per unit volume on a set of (observable and internal) state variables

$$\Psi = (\underline{\epsilon}^e, \underline{\Pi}^r, \underline{\Pi}^{int}, \theta) \tag{3}$$

Where,  $\theta$  is the absolute temperature measured from a reference temperature. We should note that  $\Psi$  is concave in  $\theta$  and convex in all other arguments.

Use the standard Cartesian tensor notation in the rectangular coordinates system. All considered fields are material fields written in the  $X_K$ -system and depending on the space-time variables  $(X_K, t)$ . The superimposed dot denotes the material time derivative (Maugin, 1992).

Accounting for the special form of the free energy 3 and the hypothesis 1 and 2, we can write the Clausius-Duhem inequality (compare with Bassiouny, 2005) as:

$$\begin{aligned} (\bar{S} - S)\dot{\theta} - \sigma_{KL}^{visc} \dot{\epsilon}_{KL}^e + \sigma_{KL} \dot{\epsilon}_{KL}^p - E_K^{relax} \dot{\Pi}_K^r \\ - E_K^{int} \dot{\Pi}_K^{int} - E_K^L \dot{\Pi}_K^R - \theta^{-1} Q_K \theta_{,K} \geq 0 \end{aligned} \tag{4}$$

where, in the following definitions have used

$$\begin{aligned} \bar{S} = -\frac{\partial \Psi}{\partial \theta}, \quad \sigma_{KL}^{visc} = \sigma_{KL} - \sigma_{KL}^e, \quad \sigma_{KL}^e = \frac{\partial \Psi}{\partial \epsilon_{KL}^e}, \\ E_K^{Lr} = -\frac{\partial \Psi}{\partial \Pi_K^L}, \quad E_K^{int} = -\frac{\partial \Psi}{\partial \Pi_K^{int}}, \quad E_K^{relax} = E_K^L - E_K^{Lr} \end{aligned} \tag{5}$$

Where,  $S$  is the entropy per unit volume,  $\sigma_{KL}$  is the component of Cauchy stress tensor,  $E^L$  the local electric field represents the interaction between the electric dipoles and the mechanical structure of the solid.  $Q$  is the heat flux vector.

The Clausius-Duhem inequality 4 can be considered as a thermodynamical constraint imposed on the constitutive equation. In the absence of electric ordering effects and polarization inertia,  $E^L$  and the electromotive intensity  $\underline{E}$  are simply related by the intermolecular balance equation written here as (Toupin, 1963; Collet and Maugin, 1974):

$$\underline{E} + \underline{E}^L = 0 \tag{6}$$

This equation suggests a balance of forces acting on the charged particles forming the polarized material point of the medium.

Dissipative processes are distinguish to that which may depend on rates and fluxes, such as viscosity, electric relaxation and heat conduction and those which may be referred to as instantaneous dissipative processes, in the sense that they do not depend on the associated rates (rate-independent plasticity is such a case, but here this notion is applied to electric processes). Accounting to the thermodynamical local-state axiom the entropy  $S$  takes its usual thermostatic definition  $S = \bar{S}$  and from 5 there remains the following dissipative inequality

$$\varphi = \varphi_1 + \varphi_2 > 0 \quad (7)$$

$$\varphi_1 = \sigma_{KL} \dot{\varepsilon}_{KL}^p + E_K^{int} \dot{\Pi}_K^{int} - E_K^L \dot{\Pi}_K^R \quad (8)$$

$$\varphi_2 = \sigma_{KL}^{visc} \dot{\varepsilon}_{KL}^e + \sigma_{KL} \dot{\varepsilon}_{KL}^p - E_K^{relax} \dot{\Pi}_K^f - \theta^{-1} Q_K \dot{\theta}_{,K} \quad (9)$$

Where,  $\varphi_1$  and  $\varphi_2$  refer to the two special classes of dissipative processes referred to above. The state laws defined in 5 must be supplemented by complementary laws for the fields that produce entropy in Eq. 7. For these, we may postulate the existence of dissipation potentials  $D_1$  and  $D_2$ , respectively, homogeneous of degree one and two in the rates so that the complementary laws read

$$\sigma_{KL} = \partial D_1 / \partial \dot{\varepsilon}_{KL}^p, E_K^{int} = -\partial D_1 / \partial \dot{\Pi}_K^{int}, E_K^L = -\partial D_1 / \partial \dot{\Pi}_K^R \quad (10)$$

on one hand and

$$\sigma_{KL}^{visc} = \partial D_2 / \partial \dot{\varepsilon}_{KL}^e, E_K^{relax} = -\partial D_2 / \partial \dot{\Pi}_K^f, Q_K = \partial D_2 / \partial A_K \quad (11)$$

on the other.

For the first set 10 one prefers to introduce a dual dissipation potential  $D_1^*$  via a Legendre-Fenchel transformation, so that evolution equations will replace 10. Indeed, in analogy with rate-independent elastoplasticity with hardening and magnetic hysteresis effects (Kachanov, 1974), instead of  $D_1^*$  one can directly introduce an electric loading function  $f(F_1, \cdot)$ ,  $F_1 = -\{\underline{\sigma}, \underline{E}^{int}, \underline{E}^{int}\}$  such that  $D_1^*$  becomes the indicator of a convex set  $C = \{F_1; f(F_1, \cdot) \leq 0\}$  which defines the domain of reversible electric processes spanned by  $F_1$ . With this, Eq. 10 is replaced by the following evolution equations:

$$\dot{\varepsilon}_{KL}^p = \dot{\lambda} \partial f_1 / \partial \sigma_{KL}, \dot{\Pi}_K^{int} = -\dot{\lambda} \partial f_1 / \partial E_K^{int}, \dot{\Pi}_K^R = -\dot{\lambda} \partial f_1 / \partial E_K^L \quad (12)$$

Where,  $\dot{\lambda}$  is called the "plastic multiplier" and  $\dot{\lambda} \geq 0$

if  $f_1 = 0$  and  $\dot{f}_1 = 0$  and  $\dot{\lambda} = 0$  if  $f_1 < 0$  or  $\dot{f}_1 < 0$ . Viscosity, electric relaxation and thermal conduction remain described by Eq. 11 with a dissipation function

$$D_2 = \left\{ \dot{\varepsilon}_{KL}^e, \dot{\Pi}_K^f, \dot{A}_K \right\}$$

The material coefficients are introduced by performing an expansion of the free energy. In doing so, a split into thermo-electro-elastic contribution  $\Psi_{pe}$  and the energy stocked by the internal variables,  $\Psi_{int}$  is achieved:

$$\Psi = \Psi_{pe}(\underline{\varepsilon}^e, \underline{\Pi}^f, \theta) + \Psi_{int}(\underline{\Pi}^{int}, \theta) \quad (13)$$

With, for instance,

$$\Psi_{int} = (1/2) Q_{KL}(\theta) \Pi_K^{int} \Pi_L^{int} \quad (14)$$

$$Q_{KL} = \partial^2 \Psi_{int} / \partial \Pi_K^{int} \partial \Pi_L^{int} \quad (15)$$

In fact,  $Q_{KL}$  will depend on  $\underline{E}^L$ . With the above and in the absence of any plastic deformation, an electric version of the celebrated Drucker inequality can be obtained in the form

$$\dot{\underline{E}} \cdot \dot{\underline{\Pi}}^R \geq 0 \quad (16)$$

on account of Eq. 6 and the convexity of  $\Psi$ .

Proceeding as in Bassiouny *et al.* (1988a), the plastic multiplier  $\dot{\lambda}$  can be determined from the consistency condition (for  $\dot{\lambda} \neq 0$ ). One gets:

$$\dot{\lambda} = h^{-1} H(f_1) \left\langle \frac{\partial f_1}{\partial \underline{E}} \cdot \dot{\underline{E}}^L \cdot \frac{\partial f_1}{\partial \theta} \dot{\theta} + \frac{\partial f_1}{\partial \underline{\sigma}} \cdot \dot{\underline{\sigma}} \right\rangle \quad (17)$$

$$h = \left( \frac{\partial f_1}{\partial E_K^{int}} \right) \cdot Q \cdot \left( \frac{\partial f_1}{\partial E_K^{int}} \right) \quad (18)$$

where,  $h$  is called the electric hardening modulus,  $H(f_1)$  is the Heaviside function and  $\langle \cdot \rangle$  indicates the positive part.

Several examples of the loading functions, the influence of stresses on these functions via electromechanical couplings and both isotropic and kinematical hardening have been studied by Bassiouny *et al.* (1988a).

**One dimensional model for poling of ceramics:** As an application of the model presented in the previous sections we consider the case where the vectorial internal variable  $\underline{\Pi}^{int}$  has the physical interpretation of a

polarization density defined by  $\underline{N} = \sum_i (p_i \cdot \underline{e}_\alpha) \underline{e}_\alpha$  where,  $\underline{e}_\alpha$

is a unit vector in the direction of the poling field,  $p_i$  is an elementary electric dipole and the summation is carried out over each subpart of the sample (in comparison with Chen, 1980, Chen and Montgomery, 1980; Bassiouny *et al.* 1988b). The norm of the vector  $\underline{N}$  gives us an indication of the number of the aligned dipoles. Accounting for Eq. 5 and expanding the material coefficients of the studied ceramic and retaining terms at most linear in  $\underline{N}$ , we have the state laws (notation of Voigt for stress and strain,  $\alpha, \beta = 1, 2, \dots, 6$ )

$$E_K^{lr} = (\chi_{KL}^{-1} + \chi_{KLQ} N_Q) \Pi_K^f + (P_{K\alpha} + P_{K\alpha Q} N_Q) E_\alpha + (\Theta_K + \Theta_{KQ} N_Q) \bar{\theta} + B_{KL} N_L \quad (19)$$

$$T_\alpha^e = (C_{\alpha\beta} + C_{\alpha\beta Q} N_Q) E_\beta - (P_{K\alpha} + P_{K\alpha Q} N_Q) \Pi_K^f + (\tau_\alpha + \tau_{\alpha Q} N_Q) \bar{\theta} + S_{\alpha Q} N_Q \quad (20)$$

$$\eta = \eta_0 + (\phi + \phi_Q N_Q) \bar{\theta} - (\tau_\alpha + \tau_{\alpha Q} N_Q) E_\alpha + (\Theta_K + \Theta_{KQ} N_Q) \Pi_K^r + \rho_K N_K \quad (21)$$

and

$$E_K^{int} = X_{KL}^{-1} N_L - S_{\alpha K} E_\alpha + B_{KL} \Pi_K^r + \rho_K \bar{\theta} \quad (22)$$

where,  $|\bar{\theta}| \leq \theta_0 < \theta_c$  with  $\theta_c$  is the Curie temperature,  $B_{KL}$  may be called the orientation polarization.  $S_{\alpha Q}$  and  $\rho_K N_K$  express the stress and the entropy during domain switching,  $\chi_{KlQ} N_Q$ ,  $P_{K\alpha Q} N_Q$ ,  $\tau_{\alpha Q} N_Q$ ,  $\Theta_{KQ} N_Q$  and  $\phi_Q N_Q$  are the changes in the inverse electric susceptibility, piezoelectricity, thermoelastic coefficients and the change in the pyroelectric vector and in the specific heat due to domain switching and  $\eta$  is the entropy per unit mass.

As the constitutive Eq. 19-22 include tensors of odd rank; they are valid for materials with no center of symmetry. These equations constitute a fully coupled, three-dimensional thermodynamical set of equations describing the poling of ferroelectric ceramics.

As in Eq. 19-22 the domain switching will affect most of the macroscopic properties of ferroelectrics. Consequently, the starting point for the experimental study of ferroelectric is the preparation of single crystals which are both single domains electrically and untwined crystallographically. Thus it seems natural that we seek certain simplifications when need arises and at the same time not destroy the essence of our present approach.

The purpose of this study is to consider the simplest non trivial constitutive relations and rate laws which are sufficient to yield results comparable to experimental observations. To shed some light and be more specific, let us consider the following one-dimensional form of the free energy

$$\Psi = \Psi (S, \Pi^r, N, \theta) \quad (23)$$

Where,  $S$  is the strain  $\theta$  is the absolute temperature and  $N$ , in fact, gives the effective number of the aligned dipoles.

Considerations shall restrict to a fairly thin specimen which is large in lateral extent and whose surfaces are stress free. Expanding Eq. 23 and using the definitions 5 in one-dimensional form and then developing the material coefficients of the considered specimen and retaining only the linear terms in the independent variable  $N$  we get the following one-dimensional constitutive equations

$$E^{Lr} = (\chi + \chi' N) \Pi^r + (e + e' N) S + (\Theta + \Theta' N) \bar{\theta} + bN \quad (24)$$

$$T^e = (C + C' N) S - (e + e' N) \Pi^r + (\tau + \tau' N) \bar{\theta} + aN \quad (25)$$

$$\eta = \eta_0 + (\phi + \phi' N) \bar{\theta} + (\Theta + \Theta' N) \Pi^r - (\tau + \tau' N) S + dN \quad (26)$$

and the following linear one-dimensional form to the internal electric field

$$E^{int} = XN - aS + b\Pi^r + d\bar{\theta} \quad (27)$$

where,  $\chi' N$ ,  $e' N$ ,  $\Theta' N$ ,  $C' N$ ,  $\tau' N$  and  $\phi'$  are the changes in the dielectric, piezoelectric, pyroelectric, elastic and thermoelastic coefficients and the specific heat constant due to domain switching,  $bN$ ,  $aN$  and  $dN$  are the additional polarization, stress and entropy occur during domain switching.

Now turn to the problem of determining a rate law governing the internal variable  $N$ , For this purpose, we turn to Eq. 12<sub>2,3</sub> which assume now the following forms:

$$\dot{N} = -\lambda \frac{\partial f_1}{\partial E^{int}}, \quad \dot{\Pi}^r = -\lambda \frac{\partial f_1}{\partial E^L} \quad (28)$$

Accounting to the fact that the electric internal variable is nothing but the residual polarization up to a constant Eq. 28 yield

$$\dot{N} = \pm Q^{-1} \langle \pm \mathcal{E} \rangle \quad \text{where, } Q = \frac{\partial^2 \Psi_{int}}{\partial N^2} \quad (29)$$

and the sign means in/or opposite the direction of the poling field. Eq. 29<sub>1</sub> is the rate law governing the evolution with time of the density of the aligned dipoles, In view of Eq. 29<sub>2</sub> and 29<sub>1</sub> is a nonlinear evolution equation governing the evolution of the poling process.

Equation 24-27 and Eq. 29 constitute a fully coupled one-dimensional thermodynamical set of equations describing the electromechanical hysteresis effects and the poling of the ferroelectric ceramics.

**Determination of the material coefficients:** All the material coefficients introduced in the modeling are defined at two different states of the sample, the virgin state and the poled state.

It is assumed that the virgin state corresponds to a natural reference state in which the material behaves isotropically. Mathematically, a virgin, or thermally depoled specimen is one for which  $\Pi_K^r = 0$ ,  $N_K = 0$  and  $\bar{\theta} = 0$ . This definition is useful for the determination of the elastic coefficients, the inverse electric susceptibility tensor and thermoelastic coefficients  $\tau_\alpha$  via standard methods.

Next define the poled state as a state for which

$$\mathcal{E}_K = 0, \quad T_\alpha^e = 0, \quad N_K = \hat{N}_K \quad (30)$$

The vector  $\hat{N}$  can be measured experimentally.  $\eta_0$  is determined by assuming that the entropy level at the

poled state is zero. i.e.,  $\eta = 0$ . Thus at this state  $\underline{N}$  is normalized with respect to the maximum value of the density of the aligned dipoles, so that  $\|\underline{N}\| = 1$ .

In fact, we can take  $\underline{N} = (1, 0, 0)$ . In this manner sets of material coefficients obtain in both one-and three-dimensional form for the changes due to the poling process. For instance, for the effect of poling on pyroelectricity we obtain

$$\Theta'_{K1} = -\Theta_K + P_{\beta K} \tau_{\alpha} S_{\alpha\beta} - \left[ (\chi_{KL}^{-1})^{pol} + P_{\beta K} P_{\alpha L} S_{\alpha\beta} \right] \left( \frac{\partial \Pi_L^r}{\partial \theta} \right)^{pol} \quad (31)$$

$$P_{\alpha K} P_{\beta L} = \left[ \left( \frac{\partial E_K^{Lr}}{\partial \Pi_L^r} \right)^{pol} - (\chi_{KL}^{-1} + \chi_{KLI}) \right] \tilde{C}_{\alpha\beta}^{pol} \quad (32)$$

Where,  $S_{\alpha\beta}$  is the elastic compliance and use the following notation

$$\tilde{A}_{..} = A_{..} + A_{..Q} N_Q$$

$$\tilde{A} = \tilde{A} \Big|_{\underline{N}=(1,0,0)}$$

Equation 31 can be useful in the observation of domain switching through the pyroelectric measurement, by determining the rate of variation of the reversible polarization with temperature in the poled state (Lines and Glass, 1977). Eq. 32 provides a means of determining the effect of domain switching on the electromechanical coupling coefficients through the slope of  $(E^{Lr}, \Pi^r)$  curve in the poled state.

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