Journal of Mechanics of Materials and Structures

THERMODYNAMICS OF INHOMOGENEOUS FERROELECTRICS Gerard A. Maugin and Liliana Restuccia

Volume 3, Nº 6

June 2008



mathematical sciences publishers

THERMODYNAMICS OF INHOMOGENEOUS FERROELECTRICS

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In a previous paper within the framework of the theory of inhomogeneities, the balance law of the socalled *pseudomomentum* for ferroelectrics was worked out exploiting the presence of *material forces*. Electric polarization density per unit mass and its gradient were introduced as state variables in the state vector. In this paper, starting from the pseudomomentum balance equation, we construct, in a systematic way, the material energy balance law for ferroelectrics which plays a crucial role in applications related to the study of fracture.

1. Introduction

Ferroelectrics are dielectric materials which possess the essential property of exhibiting a local spontaneous electrical polarization. Ferroelectricity generally disappears above a certain temperature, called the transition temperature or Curie point θ_c , at which a ferroelectric crystal passes from a polarized state of low temperature to a nonpolarized state of high temperature. Thermic agitation tends to destroy ferroelectric order. Ferroelectric crystals which don't have a Curie point exist, because they melt before reaching a ferroelectric phase. Rochelle salt has two Curie points, one higher and one lower, between which this crystal is ferroelectric. Ferroelectrics have applications in computer science, in the technology of integrated circuits, and in the fields of electronic microscopy, electronic sensors, optoelectronics, and other technological sectors. Ferroelectric media are characterized by the fact that two ordered structures coexist in them: a crystalline structure which has as order parameter the deformation of the elementary cell (tensorial parameter), and the ferroelectric order parameter consisting of the specific polarization vector. We use in our description a phenomenological approach to deformable ferroelectric crystals, derived in [Maugin 1977a; 1977b; Maugin and Pouget 1980]. Dissipative processes in ferroelectrics were investigated in [Francaviglia et al. 2004]. Electric polarization density per unit mass, possessing its own dynamics and inertia, and its gradient, responsible for nonlocal interactions and the typical ferroeletric ordering, are introduced as state variables in the state vector. In this paper, within the framework of the theory of inhomogeneities [Maugin 1993], from the pseudomomentum balance equation, worked out in [Restuccia and Maugin 2004], the material energy balance law is constructed for inhomogeneous ferroelectrics in the presence of configurational forces. This law plays a crucial role in applications related to the fracture study and the computation of the so-called energy-release rate (energy dissipated at the phase-transition fronts). Inhomogeneities can be caused by abrupt changes of material properties such as density, module of elasticity, and existence of different elements and parts, and by the presence of transition fronts, dislocations, and defects such as cavities, cracks, and inclusions, which can selfpropagate during the processes of fabrication because of changed conditions or surrounding conditions

Keywords: Eshelbian mechanics, material inhomogeneities, fracture mechanics.

that are favorable [Cherepanov 1979]. Such defect propagation can provoke a premature fracture [Maugin 1992]. A crack is one of the most common defects, and it can self-propagate when a critical threshold of a certain strength is reached. To prevent this fracture criteria for propagation of a crack can be introduced in the study of the mechanics of solids [Maugin 1992]. The critical threshold of propagation of a crack can be evaluated by introducing, for instance, the rate of energy restitution and the contour integral (more precisely, Rice's integral). This critical threshold is a precise breaking condition because of the fracture instability of a fractured medium. Fracture criteria were introduced long ago for elastic materials (Lhemon, 1888, on the mesomorphic phase of the matter; Volterra, 1907, on distortions in matter). The technological evolution of the science of materials has introduced new materials into industry that exhibit an interaction between mechanical stress elastic fields and polarization field. One of the first works on inhomogeneities is by Eshelby [1951] (see also [Eshelby 1969; Maugin 1995]), who studied a particular case of inhomogeneity: the presence of a defect in an elastic material. He introduced a fictitious force (the material force) in order to give a more detailed description of energy variation related to a position of imperfection. This force is not to be confused with surface and bulk forces. Eshelby showed that this force can be obtained starting with a contour integral on any surface surrounding the defect. In the absence of a defect, this integral becomes zero and reduces itself to a strict conservation law. Following Maugin, the material force of inhomogeneity is put into evidence by projecting the balance equations of a continuum body onto a material frame [Maugin 1992]. Also Kalpakides and Agiasofitou [2002], Vukobrat [1994] and Huang and Batra [1996] investigated ferroelectrics where the gradient of electric polarization or electric fields is considered.

2. Governing equations for ferroelectrics

We use the standard Cartesian tensor notation in rectangular coordinate systems. The general nonlinear deformation of a body, between a configurational reference \mathcal{K}_R and a current configuration \mathcal{K}_t at the time *t*, is represented by the diffeomorphism

$$\mathbf{x} = \boldsymbol{\chi}(\mathbf{X}, t), \quad \mathbf{X} = \boldsymbol{\chi}^{-1}(\mathbf{x}, t),$$

where \mathbf{x} represents Eulerian coordinates and \mathbf{X} the material coordinates of the same material particle P.

We have the following relations: $F_K^i = \partial x^i / \partial X^K = x_{,K}^i$ (denoting the components of the deformation gradient **F**), $(F^{-1})_j^K = \partial X^K / \partial x_j = X_{,j}^K$, $J_F = \det(F_K^i) > 0$ (the Jacobian of **F**), $x_{,K}^i X_{,j}^K = \delta_j^i$, $F_K^i (F^{-1})_j^K = \delta_j^i$, $X_{,i}^K x_{,L}^i = \delta_L^K$, $(F^{-1})_i^K F_L^i = \delta_L^K$. From the kinematic description one defines the physical and material velocities by $v^i = (\partial x^i / \partial t)|_X$, $V^K = (\partial X^K / \partial t)|_X$, where we have explicitly indicated the time derivatives at fixed **X** (the so-called "material derivative") and at fixed **x**. Now consider in a current configuration \mathcal{K}_t the general equations that govern the quasielectrostatics of thermoelastic ferroelectric insulators [Maugin and Pouget 1980; Restuccia and Maugin 2004]. Suppose that *the material may present continuously distributed material inhomogeneities*, that the range of the considered temperatures is much below the Curie ferroelectric phase-transition temperature θ_c , and that the body occupies the simply connected material volume V_t with regular boundary ∂V_t having unit outward normal **N** in \mathcal{K}_t .

We now discuss the governing equations.

Maxwell equations in the quasielectrostatic approximation. Let E, B, D, H, P and M denote the electric field, the magnetic induction, the electric displacement, the magnetic field, the electric polarization and the magnetization per unit volume, all evaluated in a fixed Galilean frame at time t. In the quasielectrostatic case Maxwell's equations read [Maugin 1988]

$$\nabla \times \mathbf{E} = \mathbf{0}, \qquad \nabla \times \mathbf{H} = \mathbf{0}, \qquad \nabla \cdot \mathbf{D} = 0, \qquad \nabla \cdot \mathbf{B} = 0,$$

where Lorentz-Heaviside units are used and neither currents nor electric charge are present. Further

$$\mathbf{D} = \mathbf{E} + \mathbf{P}, \qquad \mathbf{H} = \mathbf{B}, \qquad \mathbf{M} = \mathbf{0},$$

and the associated jump conditions on ∂V_t are:

$$\mathbf{n} \times \llbracket \mathbf{E} \rrbracket = \mathbf{0}, \qquad \mathbf{n} \cdot \llbracket \mathbf{B} \rrbracket = 0, \qquad \mathbf{n} \times \llbracket \mathbf{H} \rrbracket = \mathbf{0}, \qquad \mathbf{n} \cdot \llbracket \mathbf{D} \rrbracket = 0,$$

where $[\![A]\!] = A^+ - A^-$, A^+ and A^- being the field limits as the boundary is approached from outside and from inside. In the Galilean approximation, calling \mathcal{E} , \mathcal{B} , \mathcal{H} , \mathcal{P} and \mathcal{M} the same fields as **E**, **B**, **H**, **P** and **M**, but referring to an element of matter at time t in a frame $\mathcal{K}_c(\mathbf{x}, t)$, we have

$$\mathcal{E} = \mathbf{E} + c^{-1}\mathbf{u} \times \mathbf{B}, \quad \mathcal{B} = \mathbf{B} - c^{-1}\mathbf{u} \times \mathbf{E}, \quad \mathcal{H} = \mathcal{B} - \mathcal{M} = \mathcal{B}, \quad \mathcal{D} = \mathbf{D}, \quad \mathcal{M} = \mathbf{0}, \quad \mathcal{P} = \mathbf{P},$$
(1)

where **u** is the velocity of the reference \mathcal{K}_c with respect to the current reference \mathcal{K}_t . In the quasielectrostatic approximation, terms in **u** are irrelevant. Further, let π denote the *polarization vector* per unit mass in \mathcal{K}_t :

$$\boldsymbol{\pi} = \boldsymbol{\pi}(\mathbf{X}, t) = \mathbf{P}/\rho$$

where $\rho(\mathbf{x}, t)$ is the mass density.

Conservation of mass. This equation reads

$$\dot{\rho} + \rho \nabla \cdot \mathbf{v} = 0 \quad \text{in } V_t, \tag{2}$$
where $\dot{\rho}(\mathbf{X}, t) = \frac{\partial \rho(\mathbf{X}, t)}{\partial t} \Big|_{\mathbf{X}}$ is the material time derivative of ρ and $\mathbf{v} = \frac{\partial \boldsymbol{\chi}(\mathbf{X}, t)}{\partial t} \Big|_{\mathbf{X}}$. Moreover,
$$\frac{\partial \rho_0}{\partial t} \Big|_{\mathbf{X}} = 0, \quad \text{i.e.,} \quad \rho_0(\mathbf{X}) = J_F \rho, \quad \text{in } V_R. \tag{3}$$

Relation (3)₂ indicates that ρ_0 depends at most on **X**. It depends on **X** when the considered body presents *inertial material inhomogeneities*.

Motion equation. In the absence of body force (of purely mechanical origin) this equation reads

$$\operatorname{div} \mathbf{t} + \mathbf{f}^{\operatorname{em}} = \rho \, \dot{\boldsymbol{v}} \quad \text{in } V_t, \tag{4}$$

with the boundary condition

$$t_{ij}n_j = T_i^{\text{em}} \text{ on } \partial V_t,$$

where \mathbf{f}^{em} and \mathbf{T}^{em} are, in the quasielectrostatic approximation, the volume ponderomotive force in a nonrelativistically moving nonmagnetizable dielectric medium and the corresponding surface traction of purely mechanical origin, given by

$$f_i^{\text{em}} = P_j \mathcal{E}_{i,j}, \quad \mathbf{f}^{\text{em}} = (\mathbf{P} \cdot \nabla) \mathcal{E} = -(\nabla \cdot \mathbf{P}) \mathcal{E} + \nabla \cdot (\mathcal{E} \otimes \mathbf{P}), \quad \mathbf{T}^{\text{em}} = \left[\!\!\left[\mathcal{E} \otimes \mathbf{P} + \mathbf{E} \otimes \mathbf{E} - \frac{1}{2} (E^2) \mathbf{1} \right]\!\!\right] \cdot \mathbf{n},$$

and t^{ij} is the nonsymmetric Cauchy stress tensor defined by

$$t^{ij} = \sigma^{ij} + (t^{\text{int}})^{[ij]}.$$
(5)

In (5) σ^{ij} is the *intrinsic stress tensor* (the symmetric Cauchy tensor)

$$\sigma^{ij} = \sigma^{ji}$$

and $(t^{int})^{ij}$ is the interaction stress tensor defined by

$$(t^{\text{int}})^{ij} = \rho^{L} E^{i} \pi^{j} - {}^{L} \mathbb{E}^{ip} \pi^{j}_{,p}, \quad \text{with } t^{[ij]} = (t^{\text{int}})^{[ij]}.$$
(6)

This equation is the local statement of the balance of moment of momentum.

In Equation (6)₁ ${}^{L}\mathbf{E} \equiv ({}^{L}E^{i})$ is called the *local electric field vector* and represents the electric anisotropy field, accounting for the interaction between the polarization of different molecular species with the crystal lattice, while ${}^{L}\mathbf{E} \equiv ({}^{L}\mathbf{E}^{ip})$ accounts for polarization gradients and has the name of *shell-shell interaction tensor*, by identification or analogy with results from the lattice theory of alkali halides. ${}^{L}\mathbf{E}$ is responsible for the typical ferroelectric ordering. In fact, in this phenomenological model, derived in [Maugin 1977a; 1977b; Maugin and Pouget 1980], it is assumed that the medium is formed by *n* coexisting molecular species $\alpha = 1, 2, ..., n$, each one of them giving rise to a field of electric dipoles, which when suitably averaged is represented by a volume density \mathbf{P}_{α} of electrical polarization. Then, the polarization vector per unit volume is the sum of the polarization vectors per unit volume of each molecular species: $\mathbf{P} = \Sigma_{\alpha} \mathbf{P}_{\alpha}$. Letting ρ_{α} be the density of α molecules, $c_{\alpha} \equiv \rho_{\alpha}/\rho$ being the corresponding concentration, we define $\pi_{\alpha} \equiv \mathbf{P}_{\alpha}/\rho_{\alpha}$, where \mathbf{P}_{α} and π_{α} are the polarization vectors per unit volume and mass in \mathcal{K}_{t} for the molecular species α .

Balance equation for the polarization vector. A theorem in [Maugin and Pouget 1980] states that the balance equation for the polarization vector in a deformable nonmagnetizable ferroelectric medium reads (see also [Maugin 1977a; 1977b; Maugin and Pouget 1980])

$$\mathcal{E}^{i} + {}^{L}\!E^{i} + \rho^{-1L} \mathbb{E}^{ij}_{,j} = I \ddot{\pi}^{i} \qquad \text{in } \mathbf{V}_{t}, \tag{7}$$

where *I* is the so-called polarization inertia and \mathcal{E} is the *electromotive intensity* due to external sources; see (1)₁. This equation resembles Newton's law of motion.

After the introduction of the symmetric stress tensor E_t^{ij} (elastic stress tensor) defined by

$${}^{E}t^{ij} = \sigma^{ij} - \rho^{L}E^{(i}\pi^{j)} + {}^{L}\mathbb{E}^{(ik}\pi^{j)}_{k} = {}^{E}t^{ji}$$

Equation (5) reads

$$t^{ij} = {}^{E}t^{ij} + \rho^{L}E^{i}\pi^{j} - {}^{L}\mathbb{E}^{ik}\pi^{j}_{,k} = {}^{E}t^{ij} + (t^{\text{int}})^{ij}.$$
(8)

Conservation of energy. The first law of thermodynamics, in the absence of a heat source by radiation, reads

$$\rho \dot{e} = t^{ji} v_{i,j} - \rho^L E^i \dot{\pi}_i + {}^L \mathbb{E}^{ij} (\dot{\pi}_i)_{,j} - q^k_{,k}.$$
⁽⁹⁾

Entropy inequality and Clausius–Duhem inequality. In this paper we use the following form of the entropy inequality:

$$\rho \dot{\eta} + \boldsymbol{\nabla} \cdot \mathbf{j}_s \ge 0$$

where η is the entropy per unit mass and \mathbf{j}_s is the entropy flux, defined by $\mathbf{j}_s = \mathbf{q}/\theta$.

Introducing Helmholtz's free energy per unit mass $\psi = e - \eta \theta$ by a *Legendre transformation* and using the energy balance equation, the following Clausius–Duhem inequality is obtained:

$$-\rho(\dot{\psi}+\eta\dot{\theta})+t^{ji}v_{i,j}-\rho^{L}E^{i}\dot{\pi}_{i}+{}^{L}\mathbb{E}^{ij}(\dot{\pi}_{i})_{,j}-\theta^{-1}q^{k}\theta_{,k}\geq0,$$

where $0 < \theta \ll \theta_c$.

3. A thermodynamical model for ferroelectrics

In [Restuccia and Maugin 2004], following the general philosophy exposed in the theory of the inhomogeneities [Maugin 1993], in order to put in evidence the material force of inhomogeneity, balance equations of continuum were projected onto \mathcal{K}_R material frame, effecting the following *Piola transformations* (see also [Maugin and Pouget 1980; Lax and Nelson 1976]):

$$\mathbf{T} = J_F \mathbf{F}^{-1} \cdot \mathbf{t}, \quad T^{Ki} = J_F (F^{-1})_j^K t^{ji},$$

$$^E \mathbf{T} = J_F \mathbf{F}^{-1} \cdot ^E \mathbf{t}, \quad ^E T^{Ki} = J_F (F^{-1})_j^K ^E t^{ji},$$

$$^L \mathbf{E} = J_F \mathbf{F}^{-1} \cdot ^L \mathbf{E}, \quad ^L \mathbf{E} \mathbf{E}^{Ki} = J_F (F^{-1})_j^K ^L \mathbf{E}^{ji},$$

$$\mathbf{Q} = J_F \mathbf{F}^{-1} \cdot \mathbf{q}, \quad Q^K = J_F (F^{-1})_j^K q^j,$$

$$\mathbf{J}_{\mathbf{s}} = J_F \mathbf{F}^{-1} \cdot \mathbf{j}_{\mathbf{s}}, \quad J_s^K = J_F (F^{-1})_j^K j_s^j,$$

$$^L \mathbf{E} = \mathbf{F}^T \cdot ^L \mathbf{E}, \quad ^L \mathbf{E}_K = ^L E_i F_K^i,$$

$$^L E^s = \delta^{si} {}^L E_i = \delta^{si} (F^{-1})_i^K {}^L \mathbf{E}_K,$$

$$\mathbf{E} = \mathbf{F}^T \mathbf{\mathcal{E}}, \quad \mathbf{E}_K = \mathcal{E}_i F_K^i,$$

$$\mathbf{\Pi} = J_F \mathbf{F}^{-1} \cdot \mathbf{P}, \quad \Pi^K = J_F (F^{-1})_j^K P^j.$$

Multiplying (8) by $J_F(F^{-1})_I^K$, the following Piola transformation was derived:

$$T^{Ki} = {}^{E}T^{Ki} + \rho_0(F^{-1})_l^{K} {}^{L}E^l \pi^i - {}^{L}\mathbf{E}^{Kl}\pi^i_{,l},$$
(10)

where **T** is the first *Piola–Kirchhoff* stress.

Further, multiplying the balance of energy by J_F , we obtain

$$\dot{E} = (T^{Ki}\delta_{ij})\dot{F}^{j}_{k} - \rho_{0}\delta^{is}(F^{-1})^{K}_{s}{}^{L}\mathsf{E}_{K}\dot{\pi}_{i} + {}^{L}\mathbb{E}^{iK}(\dot{\pi}_{i})_{,K} - Q^{K}_{,K},$$
(11)

where $E = \rho_0 e$; doing the same to the entropy inequality and the Clausius–Duhem inequality we get

$$\theta \dot{S} \ge -Q_{,K}^{K} + \theta^{-1} Q^{K} \theta_{,K},$$

$$-(\dot{W} + S\dot{\theta}) + (T^{Ki} \delta_{ij}) \dot{F}_{K}^{j} - \rho_{0} \delta^{is} (F^{-1})_{s}^{K} {}^{L} \mathsf{E}_{K} \dot{\pi}_{i} + {}^{L} \mathbf{E}^{iK} (\dot{\pi}_{i})_{,K} - \theta^{-1} Q^{K} \theta_{,K} \ge 0,$$

$$= \rho_{0} \eta_{K} - F_{K} - S\theta_{K}$$

where $S = \rho_0 \eta$ and $W = \rho_0 \psi = E - S\theta$.

In [Restuccia and Maugin 2004] a thermodynamical model for materially inhomogeneous thermoelastic ferroelectric insulators was proposed, choosing the following state vector

$$C = C(\mathbf{F}, \boldsymbol{\pi}, \nabla_{R}\boldsymbol{\pi}, \boldsymbol{\theta}, \nabla_{R}\boldsymbol{\theta}; \mathbf{X}),$$
(12)

where the physical fields π and $\nabla_R \pi$ are responsible for the internal structure of the medium, the relaxation properties of the thermal field are taken into account, and the explicit dependence on **X** reflects the material inhomogeneity. In (12) the symbol ∇_R denotes the gradient operator in material space. The constitutive dependent variables of the set

$$\mathbf{Z} = \mathbf{Z}(W, S, \mathbf{T}, {}^{L}\mathbf{E}, {}^{L}\mathbf{E}, \mathbf{Q}),$$

were determined as functions of the set C, that is, $\mathbf{Z} = \mathbf{Z}(C)$, and using the expression

 $W = W(\mathbf{F}, \boldsymbol{\pi}, \nabla_R \boldsymbol{\pi}, \boldsymbol{\theta}, \nabla_R \boldsymbol{\theta}; \mathbf{X})$

and the Clausius–Duhem inequality, the *nonlinear constitutive equations*, the *dissipation inequality*, and other results were worked out:

$$T^{Ki} = \frac{\partial W}{\partial F_K^j} \delta^{ji}, \qquad {}^{L}\mathsf{E}_K = -\rho_0^{-1} \frac{\partial W}{\partial \pi_i} \left(\mathbf{F}\right)_K^i, \qquad {}^{L}\mathbf{E}^{iK} = \frac{\partial W}{\partial \pi_{i,K}}, \tag{13}$$

$$S = -\frac{\partial W}{\partial \theta}, \qquad \frac{\partial W}{\partial \theta_{,K}} = 0, \qquad -\theta^{-1} Q^{K} \theta_{,K} \ge 0.$$
(14)

Then

$$W = W(\mathbf{F}, \boldsymbol{\pi}, \nabla_{R}\boldsymbol{\pi}, \boldsymbol{\theta}; \mathbf{X}),$$

but

$$Q^{K} = Q^{K}(\mathbf{F}, \boldsymbol{\pi}, \nabla_{R}\boldsymbol{\pi}, \theta, \nabla_{R}\boldsymbol{\theta}; \mathbf{X}),$$

with $\lim_{\nabla_R \theta \to 0} Q^K(\mathbf{F}, \boldsymbol{\pi}, \nabla_R \boldsymbol{\pi}, \theta, \nabla_R \boldsymbol{\theta}; \mathbf{X}) = 0$ (continuity condition).

From (11), using the constitutive relations and the Legendre transformation $W = E - S\theta$, the energy equation can be rewritten as

$$\theta \frac{\partial S}{\partial t}\Big|_{\mathbf{X}} + \nabla_{R} \cdot \mathbf{Q} = 0, \quad \text{or} \quad \frac{\partial S}{\partial t}\Big|_{\mathbf{X}} + \nabla_{R} \cdot \mathbf{J}_{\mathbf{s}} = -\theta^{-2} \mathcal{Q}^{K} \theta_{,K}.$$
 (15)

4. Material energy balance

In [Restuccia and Maugin 2004], following the philosophy of the theory of the inhomogeneities exposed in [Maugin 1993], in order to place the presence of *material forces* and to obtain the balance of material momentum (called balance of pseudomomentum), the motion equation (4) was projected onto the material manifold \mathcal{M}^3 by applying the operator $J_F \mathbf{F}^T$ at the left of equation (4). This operation is called *convection* or *pull-back*. First, by multiplying Equation (4) by J_F , the following Piola–Kirchhoff form was obtained:

$$T_{i,K}^{K} + J_F f_i^{\text{em}} = J_F \rho \dot{v}_i = \rho_0 \dot{v}_i, \qquad \text{div}_R \mathbf{T} + J_F \mathbf{f}^{\text{em}} = \frac{\partial \mathbf{p}_R}{\partial t} \Big|_{\mathbf{X}}, \tag{16}$$

where $\mathbf{p}_R = \rho_0 \mathbf{v}$ is defined as the *physical linear momentum per unit volume* in \mathcal{K}_R .

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Next, applying the pull-back operator (\mathbf{F}^T) on the left-hand side of $(16)_1$,

$$F_L^i T_{i,K}^K + J_F F_L^i f_i^{\text{em}} = F_L^i \frac{\partial}{\partial t} \left(\rho_o v_i \right) \Big|_X,$$

the following balance of pseudomomentum projected on \mathcal{M}^3 was obtained:

$$\frac{\partial \boldsymbol{\mathcal{P}}}{\partial t}\Big|_{\mathbf{X}} - \operatorname{div}_{R} \,\hat{\boldsymbol{b}} = \mathbf{f}^{\operatorname{inh}} + \mathbf{f}^{\operatorname{fer}},\tag{17}$$

where

$$\mathcal{P} = -\rho_{0}\mathbf{F}^{T}\cdot\mathbf{v} - (\nabla_{R}\boldsymbol{\pi})\cdot\left(\rho_{0}\boldsymbol{I}\,\dot{\boldsymbol{\pi}}\right), \qquad (18)$$

$$\hat{\boldsymbol{b}} = -\left(\hat{\mathcal{L}}\mathbf{1}_{R} + \mathbf{T}\cdot\mathbf{F} + (\nabla_{R}\boldsymbol{\pi})\cdot{}^{L}\mathbf{H}\mathbf{E}\right), \qquad (18)$$

$$\hat{\mathcal{L}} = \rho_{0}\left(\mathbf{X}\right)\left(\frac{1}{2}\mathbf{v}^{2} + \frac{1}{2}\boldsymbol{I}\,\dot{\boldsymbol{\pi}}^{2} + \boldsymbol{\mathcal{E}}\cdot\boldsymbol{\pi}\right) - W(\mathbf{F},\boldsymbol{\pi},\nabla_{R}\boldsymbol{\pi},\boldsymbol{\theta};\mathbf{X}), \qquad \mathbf{f}^{\text{inh}} = \nabla_{R}\rho_{0}\left(\mathbf{X}\right)\left(\frac{1}{2}\mathbf{v}^{2} + \frac{1}{2}\boldsymbol{I}\,\dot{\boldsymbol{\pi}}^{2} + \boldsymbol{\mathcal{E}}\cdot\boldsymbol{\pi}\right) - \nabla_{R}\mathbf{W}|_{\text{expl}}, \qquad \mathbf{f}^{\text{th}} = S\nabla_{R}\boldsymbol{\theta}, \qquad \mathbf{f}^{\text{fer}} = \rho_{0}\boldsymbol{\pi}\cdot\nabla_{R}\boldsymbol{\mathcal{E}} - \mathbf{F}^{T}\cdot(\boldsymbol{\Pi}\cdot\nabla_{R})\boldsymbol{\mathcal{E}}.$$

In these expressions \mathcal{P} is the *pseudomomentum*, a material *covector* on \mathcal{M}^3 , $\hat{\boldsymbol{b}}$ is referred to as the *Eshelby* (material) stress tensor accounting for ferroelectric exchange effects, \mathbf{f}^{inh} is the *material inhomogeneity* force, \mathbf{f}^{th} is called the *thermal material force*, and \mathbf{f}^{fer} is a new material force which reflects the presence of ferroelectric effects (see also [Maugin and Pouget 1980]). The inhomogeneity force \mathbf{f}^{inh} here has its canonical definition

$$\mathbf{f}^{\text{inh}} = \frac{\partial \hat{\mathcal{L}}}{\partial \boldsymbol{X}}\Big|_{\text{expl}},$$

where the potential $\hat{\mathcal{L}}$ would be the Lagrangian density if irreversible processes were not present. Now, although (11) already provides an expression of the local energy equation, we construct the expression of the material energy balance that plays a crucial role in applications related to the study of fracture in a medium. Using the already obtained results, upon scalar multiplication of (17) by the material velocity, we obtain

$$\frac{\partial \boldsymbol{\mathcal{P}}}{\partial t}\Big|_{\mathbf{X}} \cdot \mathbf{V} - \left(\operatorname{div}_{R} \hat{\boldsymbol{b}}\right) \cdot \mathbf{V} = \mathbf{f}^{\operatorname{inh}} \cdot \mathbf{V} + \mathbf{f}^{\operatorname{th}} \cdot \mathbf{V} + \mathbf{f}^{\operatorname{fer}} \cdot \mathbf{V}.$$
(19)

We evaluate each contribution separately, systematically using the following relations (see [Fomethe and Maugin 1996]):

$$\mathbf{V} = \frac{\partial \mathbf{\chi}^{-1}(\mathbf{x}, t)}{\partial t}\Big|_{\mathbf{x}}, \quad v^{i} = -F_{K}^{i}V^{K}, \quad V^{K} = -(F^{-1})_{i}^{K}v^{i},$$
$$\dot{\mathbf{A}}(\mathbf{X}, t) = \frac{\partial \mathbf{A}(\mathbf{X}, t)}{\partial t}\Big|_{\mathbf{X}}, \quad \dot{\mathbf{A}} = \frac{\partial \mathbf{A}}{\partial t}\Big|_{\mathbf{x}} + \mathbf{v} \cdot \nabla \mathbf{A}, \quad \frac{\partial \mathbf{A}}{\partial t}\Big|_{\mathbf{x}} = \dot{\mathbf{A}} + \mathbf{V} \cdot \nabla_{R}\mathbf{A},$$
$$A_{,K}^{i} = A_{,j}^{i}F_{K}^{j}, \quad A_{,j}^{i} = A_{,K}^{i}(F^{-1})_{j}^{K}, \quad \mathbf{v} \cdot \nabla \mathbf{A} = \frac{\partial \mathbf{A}}{\partial t}\Big|_{\mathbf{X}}, \quad \mathbf{V} \cdot \nabla_{R}\mathbf{A} = \frac{\partial \mathbf{A}}{\partial t}\Big|_{\mathbf{x}}, \quad \mathbf{V} \cdot \nabla_{R}\mathbf{A} = \frac{\partial \mathbf{A}}{\partial t}\Big|_{\mathbf{x}}, \quad \mathbf{V} \cdot \nabla_{R}\mathbf{A} = \frac{\partial \mathbf{A}}{\partial t}\Big|_{\mathbf{x}}, \quad \dot{F}_{K}^{i} = v_{,K}^{i} = v_{,j}^{i}F_{K}^{j}, \quad v_{,j}^{i}v^{j} = \frac{\partial v^{i}}{\partial t}\Big|_{\mathbf{X}} - \frac{\partial v^{i}}{\partial t}\Big|_{\mathbf{x}}, \quad \frac{\partial \rho_{0}}{\partial t}\Big|_{\mathbf{X}} = 0,$$

where the objective vector field \mathbf{A} is a geometrical time-dependent object which is form invariant under rigid-body changes of coordinate frames in K_t and its components transform tensorially. Then, from equations (18) and (19) we have

$$\frac{\partial}{\partial t}(-\rho_0 \mathbf{F}^T \cdot \mathbf{v})\Big|_{\mathbf{X}} \cdot \mathbf{V} = \frac{\partial}{\partial t}(\rho_0 v^2)\Big|_{\mathbf{X}} - \rho_0 \frac{\partial}{\partial t} \left(\frac{1}{2} v^2\right)\Big|_{\mathbf{x}}$$
(20)

and

$$-\frac{\partial}{\partial t} \left[\nabla_{R} \boldsymbol{\pi} \cdot \left(\rho_{0} I \dot{\boldsymbol{\pi}} \right) \right] \Big|_{\mathbf{X}} \cdot \mathbf{V} = -\left(\rho_{0} I \ddot{\pi}^{i} \pi_{i,L} + \rho_{0} I \dot{\pi}^{i} \dot{\pi}_{i,L} \right) V^{L}$$
$$= -\rho_{0} I \ddot{\pi}^{i} \frac{\partial \pi_{i}}{\partial t} \Big|_{\mathbf{X}} + \rho_{0} I \ddot{\pi}^{i} \frac{\partial \pi_{i}}{\partial t} \Big|_{\mathbf{X}} - \rho_{0} I \dot{\pi}^{i} \frac{\partial \dot{\pi}_{i}}{\partial t} \Big|_{\mathbf{X}} + \rho_{0} I \dot{\pi}^{i} \frac{\partial \dot{\pi}_{i}}{\partial t} \Big|_{\mathbf{X}}.$$
(21)

The second term on the left-hand side of (19) gives

$$(\operatorname{div}_{R}\hat{\boldsymbol{b}})\cdot\mathbf{V} = -\hat{\mathcal{L}}_{,L}V^{L} - \left(T_{i}^{K}F_{L}^{i} + {}^{L}\mathbb{E}^{iK}\pi_{i,L}\right)_{,K}V^{L}.$$
(22)

Evaluating the contributions in the right side of (22) we have

$$-\hat{\mathcal{L}}_{,L}V^{L} = -\frac{\partial\hat{\mathcal{L}}}{\partial t}\Big|_{\mathbf{x}} + \frac{\partial\hat{\mathcal{L}}}{\partial t}\Big|_{\mathbf{x}},$$

where

$$-\frac{\partial \hat{\mathcal{L}}}{\partial t}\Big|_{\mathbf{x}} = \frac{\partial W}{\partial F_{K}^{j}} \frac{\partial F_{K}^{j}}{\partial t}\Big|_{\mathbf{x}} + \frac{\partial W}{\partial \pi_{i}} \frac{\partial \pi_{i}}{\partial t}\Big|_{\mathbf{x}} + \frac{\partial W}{\partial \pi_{i,K}} \frac{\partial \pi_{i,K}}{\partial t}\Big|_{\mathbf{x}} + \frac{\partial W}{\partial \theta} \frac{\partial \theta}{\partial t}\Big|_{\mathbf{x}} + \frac{\partial W}{\partial X^{L}}\Big|_{\exp1} \frac{\partial X^{L}}{\partial t}\Big|_{\mathbf{x}} - \left(\frac{1}{2}v^{2} + \frac{1}{2}I\dot{\pi}^{2} + \mathcal{E}\cdot\boldsymbol{\pi}\right)\frac{\partial\rho_{0}}{\partial t}\Big|_{\mathbf{x}} - \rho_{0}\frac{\partial}{\partial t}\left(\frac{1}{2}v^{2} + \frac{1}{2}I\dot{\pi}^{2} + \mathcal{E}\cdot\boldsymbol{\pi}\right)\Big|_{\mathbf{x}}$$
(23)

and

$$\frac{\partial \hat{\mathcal{L}}}{\partial t}\Big|_{\mathbf{X}} = \frac{\partial}{\partial t} \left[\rho_0(\mathbf{X}) \left(\frac{1}{2} v^2 + \frac{1}{2} I \dot{\pi}^2 + \boldsymbol{\mathcal{E}} \cdot \boldsymbol{\pi} \right) \right] \Big|_{\mathbf{X}} - \frac{\partial W}{\partial t} \Big|_{\mathbf{X}}.$$
(24)

In the following, we use the constitutive equations (13) and (14)₁ at fixed **x** in the term $(\partial \hat{\mathcal{L}}/\partial t)|_{\mathbf{x}}$. We further have

$$-\left(T_{i}^{K}F_{L}^{i}\right)_{,K}V^{L} = T_{i,K}^{K}v^{i} - T_{i}^{K}\frac{\partial F_{K}^{i}}{\partial t}\Big|_{\mathbf{x}} + T_{i}^{K}v_{i,K} = \nabla_{R}\cdot(\mathbf{T}\cdot\mathbf{v}) - T_{i}^{K}\frac{\partial F_{K}^{i}}{\partial t}\Big|_{\mathbf{x}},$$
(25)

$$-\left({}^{L}\mathbb{E}^{iK}\pi_{i,L}\right)_{,K}V^{L} = -{}^{L}\mathbb{E}^{iK}_{,K}\frac{\partial\pi_{i}}{\partial t}\Big|_{\mathbf{x}} + {}^{L}\mathbb{E}^{iK}_{,K}\frac{\partial\pi_{i}}{\partial t}\Big|_{\mathbf{X}} - {}^{L}\mathbb{E}^{iK}\frac{\partial\pi_{i,K}}{\partial t}\Big|_{\mathbf{x}} + {}^{L}\mathbb{E}^{iK}\frac{\partial\pi_{i,K}}{\partial t}\Big|_{\mathbf{X}},$$
(26)

$${}^{L}\mathbb{E}_{,K}^{iK}\frac{\partial\pi_{i}}{\partial t}\Big|_{\mathbf{X}} + {}^{L}\mathbb{E}^{iK}\frac{\partial\pi_{i,K}}{\partial t}\Big|_{\mathbf{X}} = \nabla_{R} \cdot ({}^{L}\mathbb{E}\cdot\dot{\boldsymbol{\pi}}),$$
(27)

$$\mathbf{f}^{\text{inh}} \cdot \mathbf{V} = \left(\frac{1}{2}v^2 + \frac{1}{2}I\dot{\pi}^2 + \boldsymbol{\mathcal{E}} \cdot \boldsymbol{\pi}\right) \frac{\partial\rho_0}{\partial t} \Big|_{\mathbf{x}} - \frac{\partial W}{\partial t} \Big|_{\text{expl}},\tag{28}$$

and

$$\mathbf{f}^{\text{th}} \cdot \mathbf{V} = S\theta_{,L} V^{L} = S \frac{\partial \theta}{\partial t} \Big|_{\mathbf{x}} - S\dot{\theta} = S \frac{\partial \theta}{\partial t} \Big|_{\mathbf{x}} + \dot{W} - \dot{E} - \nabla_{R} \cdot \mathbf{Q},$$
(29)

where we have used the relation $-S\dot{\theta} = \dot{W} - \dot{E} - \nabla_R \cdot \mathbf{Q}$, obtained by the Legendre transformation $W = E - S\theta$ and the entropy balance equation (15)₁.

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Finally, using the relations

$$\rho_{0}\pi^{i}\mathcal{E}_{i,L}V^{L} = \rho_{0}\left(\pi^{i}\mathcal{E}_{i}\right)_{,L}V^{L} - \rho_{0}\mathcal{E}_{i}\pi^{i}_{,L}V^{L}, \quad \mathbf{f}^{\text{fer}}\cdot\mathbf{V} = \rho_{0}\pi^{i}\mathcal{E}_{i,L}V^{L} - F_{L}^{i}\Pi^{K}\mathcal{E}_{i,K}V^{L},$$
$$\rho_{0}I\ddot{\pi}^{i}\frac{\partial\pi_{i}}{\partial t}\Big|_{\mathbf{X}} = \rho_{0}I\dot{\pi}^{i}\frac{\partial\dot{\pi}_{i}}{\partial t}\Big|_{\mathbf{X}} = \rho_{0}\frac{\partial}{\partial t}\left(\frac{1}{2}I\dot{\pi}^{2}\right)\Big|_{\mathbf{X}},$$

the balance equation for the polarization vector (7) multiplied by $\rho_0 \frac{\partial \pi_i}{\partial t} \Big|_{r}$,

$$\rho_0 \mathcal{E}^i \frac{\partial \pi_i}{\partial t} \Big|_{\mathbf{x}} + \rho_0 {}^L E^i \frac{\partial \pi_i}{\partial t} \Big|_{\mathbf{x}} + {}^L \mathbb{I}_{,K}^{K} \frac{\partial \pi_i}{\partial t} \Big|_{\mathbf{x}} = \rho_0 I \ddot{\pi}^i \frac{\partial \pi_i}{\partial t} \Big|_{\mathbf{x}},$$

operating some transformations and substituting all the contributions (20)–(29) in (19), we obtain the energy balance equation in the form

$$\frac{\partial}{\partial t} \left[E + \rho_0(\mathbf{X}) \left(\frac{1}{2} v^2 + \frac{1}{2} I \dot{\pi}^2 - \boldsymbol{\mathcal{E}} \cdot \boldsymbol{\pi} \right) \right] \Big|_{\mathbf{X}} - \nabla_R \cdot \left(\mathbf{T} \cdot \mathbf{v} + {}^L \mathbf{I} \mathbf{E} \cdot \dot{\boldsymbol{\pi}} - \mathbf{Q} \right) = H, \tag{30}$$

where

$$H = -\rho_0 \boldsymbol{\pi} \cdot \dot{\boldsymbol{\mathcal{E}}} - \boldsymbol{F}^T \cdot (\boldsymbol{\Pi} \cdot \boldsymbol{\nabla}_R) \, \boldsymbol{\mathcal{E}} \cdot \boldsymbol{V}.$$
(31)

In the left-hand side of (30) there appear the partial time derivatives at fixed **X** of the internal energy E, the kinetic energy of the material lattice, the kinetic energy of the polarization vector that has own inertia, the interaction energy between the electric and the polarization fields and the material energy fluxes related to the Piola–Kirchoff stress **T**, the shell-shell interaction tensor ${}^{L}\mathbf{E}$ and the negative of the heat flux. In the right-hand side there are energy sources due to material forces which reflect the presence of ferroelectric effects.

Next we have

$$\mathbf{f}^{\text{fer}} \cdot \mathbf{V} = \rho_0 \pi^i \mathcal{E}_{i,L} V^L - F_L^i \Pi^K \mathcal{E}_{i,K} V^L = -\rho_0 \pi^i (F^{-1})^L_q v^q \mathcal{E}_{i,L} - \rho_0 F_L^i (F^{-1})^K_j \pi^j \mathcal{E}_{i,K} V^L \\ = -\rho_0 \pi^i \mathcal{E}_{i,q} v^q + \rho_0 \pi^j \mathcal{E}_{i,j} v^i = -\rho_0 \pi^i v^j (\mathcal{E}_{i,j} - \mathcal{E}_{j,i}) = 0,$$

where we have taken into consideration that $\nabla \times \boldsymbol{\mathcal{E}} = 0$ (we are in quasielectrostatic approximation) and

$$\Pi^{K} = J_{F}(F^{-1})_{j}^{K} P^{j} = \rho J_{F}(F^{-1})_{j}^{K} \pi^{j} = \rho_{0}(F^{-1})_{j}^{K} \pi^{j}.$$

Then $\mathbf{f}^{\text{fer}} \cdot \mathbf{V} \equiv 0$. That means \mathbf{f}^{fer} has no dissipative content. Now we transform *H*:

$$H = -\rho_0 \boldsymbol{\pi} \cdot \dot{\boldsymbol{\mathcal{E}}} - \boldsymbol{F}^T \cdot (\boldsymbol{\Pi} \cdot \boldsymbol{\nabla}_R) \, \boldsymbol{\mathcal{E}} \cdot \mathbf{V} = -\rho_0 \pi^i \dot{\mathcal{E}}_i - F_L^j \boldsymbol{\Pi}^K \mathcal{E}_{j,K} V^L$$
$$= -\rho_0 \pi^i \dot{\mathcal{E}}_i - \rho_0 F_L^j \left(F^{-1} \right)_q^K \pi^q \mathcal{E}_{j,K} V^L = -\rho_0 \pi^i \dot{\mathcal{E}}_i + \rho_0 \pi^i \mathcal{E}_{i,j} v^j = -\rho_0 \pi^i \left(\frac{\partial \mathcal{E}_i}{\partial t} \right) \Big|_{\mathbf{x}}.$$

Using the relations $\mathcal{E} = -\nabla \varphi(x, t)$ and $\rho_0 \pi \cdot \nabla \equiv \mathbf{\Pi} \cdot \nabla_R$, we then obtain

$$H = \rho_0 \boldsymbol{\pi} \cdot \boldsymbol{\nabla} \left(\frac{\partial \varphi}{\partial t} \right) \Big|_{\mathbf{x}} = (\boldsymbol{\Pi} \cdot \boldsymbol{\nabla}_R) \left(\frac{\partial \varphi}{\partial t} \right) \Big|_{\mathbf{x}} = \boldsymbol{\nabla}_R \cdot \left(\boldsymbol{\Pi} \frac{\partial \varphi}{\partial t} \Big|_{\mathbf{x}} \right) - (\boldsymbol{\nabla}_R \cdot \boldsymbol{\Pi}) \frac{\partial \varphi}{\partial t} \Big|_{\mathbf{x}}.$$
 (32)

Finally, using (32) the energy balance equation (30) reads

$$\frac{\partial}{\partial t} \left[E + \rho_0(\mathbf{X}) \left(\frac{1}{2} v^2 + \frac{1}{2} I \dot{\pi}^2 - \boldsymbol{\mathcal{E}} \cdot \boldsymbol{\pi} \right) \right] \Big|_{\mathbf{X}} + \nabla_R \cdot \left(\mathbf{T} \cdot \mathbf{v} + \mathbf{\Pi} \frac{\partial \varphi}{\partial t} \Big|_{\mathbf{x}} + {}^L \mathbf{H} \cdot \dot{\boldsymbol{\pi}} - \mathbf{Q} \right) = H, \quad (33)$$

with

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$$H = -\left(\nabla_{R} \cdot \mathbf{\Pi}\right) \frac{\partial \varphi}{\partial t}\Big|_{\mathbf{x}}, \qquad E = \bar{E}(\mathbf{F}, \boldsymbol{\pi}, \nabla_{R} \boldsymbol{\pi}, \boldsymbol{\theta}; \mathbf{X}).$$
(34)

Although it is not given as a strict conservation law, this expression of the energy conservation is of interest because (i) it can be used directly for the evaluation of the *energy-release rate* in the fracture study (compare to the case of classical dielectric-piezoelectrics in [Maugin and Dascalu 1993]), and (ii) it makes the comparison with classical electroelasticity easy in the appropriate reduction.

Indeed, in this simplified case we have

$$I = 0, \quad {}^{L}\mathbf{E} = 0$$

and, for quasielectrostatics, equations (33) and (34) yield

$$\frac{\partial}{\partial t} \left[E - \rho_0(\mathbf{X}) \boldsymbol{\mathcal{E}} \cdot \boldsymbol{\pi} \right] - \boldsymbol{\nabla}_R \cdot \left(\mathbf{T} \cdot \mathbf{v} + \boldsymbol{\Pi} \frac{\partial \varphi}{\partial t} \Big|_{\mathbf{x}} - \mathbf{Q} \right) = -(\boldsymbol{\nabla}_R \cdot \boldsymbol{\Pi}) \frac{\partial \varphi}{\partial t} \Big|_{\mathbf{x}}.$$
(35)

Simultaneously, equations (7) and (13)₂ yield $\mathcal{E}^i + {}^L E^i = 0$,

$$\mathcal{E}_i = -{}^L E_i = -(F^{-1})_i^{K\ L} \mathsf{E}_K = \rho_0^{-1} \frac{\partial W}{\partial \pi_j} (F^{-1})_i^K F_K^j, \quad \mathcal{E}_i = \rho_0^{-1} \frac{\partial W}{\partial \pi_i}$$

Thus, $E - \rho_0 \pi \cdot \mathcal{E} = W + S\theta - \rho_0 \pi \cdot \mathcal{E}$. Setting

$$\bar{W} = W(\mathbf{F}, \boldsymbol{\pi}, \theta) - \rho_0 \boldsymbol{\pi} \cdot \boldsymbol{\mathcal{E}} = \bar{W}(\mathbf{F}, \boldsymbol{\mathcal{E}}, \theta; \mathbf{X}), \quad \pi_j = \rho_0^{-1} \frac{\partial W}{\partial \mathcal{E}_j},$$

we finally obtain from equation (35)

$$\frac{\partial}{\partial t}\bar{W}(\mathbf{F},\boldsymbol{\mathcal{E}},\theta;\mathbf{X})\Big|_{\mathbf{X}}-\nabla_{R}\cdot\left(\mathbf{T}\cdot\mathbf{v}+\mathbf{\Pi}\frac{\partial\varphi}{\partial t}\Big|_{\mathbf{x}}-\mathbf{Q}\right)=-\left(\nabla_{R}\cdot\mathbf{\Pi}\right)\frac{\partial\varphi}{\partial t}\Big|_{\mathbf{x}},$$

which is equation (21) in [Maugin and Dascalu 1993] if we discard the temperature effect.

Acknowledgements

This work has been partially supported by GNFM of INDAM. Restuccia is grateful to W. Muschik for useful remarks and stimulating discussions.

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Received 13 Mar 2008. Accepted 26 Apr 2008.

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