



# Dissipative ferroelectricity at finite strains. Variational principles, constitutive assumptions and algorithms



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## ABSTRACT

In recent years increasing interest in functional materials such as ferroelectric polymers and ceramics has been shown. For those materials, electric polarizations and viscous effects cause dissipative phenomena, such as the characteristic *dielectric and butterfly hystereses*. The deformation of polymers is characterized by *large strains and rotations*. This work develops a general framework for the formulation and numerical implementation of ferroelectric materials at finite deformations. In particular, continuous and discrete *variational principles* for the dissipative response of quasi-static finite electro-mechanics are developed, which fully determine the continuous evolution and incremental update problems. These principles recast the framework into a canonically compact structure, that makes the model-inherent *symmetries* of the coupled problem transparent. In the algorithmic setting, two-step update schemes are developed, first for the local internal variables and next for the global primary fields, both fully determined by the exploitation of *incremental potentials*. Specific constitutive assumptions are proposed for finite ferroelectricity. A critical point is the definition of kinematic assumptions in the large-strain context, such as the *multiplicative decomposition* of the deformation gradient into reversible and remanent parts. The proposed formulation allows to reproduce dielectric and butterfly hystereses together with their rate-dependency and accounts for macroscopically non-uniform distributions of the polarization at finite deformations. The performance of the proposed methods is demonstrated by means of benchmark problems undergoing large deformations.

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## 1. Introduction

Ferroelectric polymers show complex hysteresis phenomena due to electric polarizations and viscous effects accompanied by large deformations. These effects are caused by their specific microstructures with fundamental ingredients: First, a non-centro-symmetric molecular constitution which induces the appearance of elementary *micro-dipoles* with certain spontaneous polarization. The ability to orient or align the molecular dipoles yields a *piezoelectric* behavior of the unit cells. Secondly, the capability of *alignment* of unit cells under a certain electro-mechanical loading and the ability of retaining this alignment after unloading, inducing the *dissipative* behavior of those materials. A ferroelectric polymer appears in a *semi-crystalline form* in which the ferroelectric crystallites are embedded in an amorphous matrix. Examples include polyvinylfluoride PVF (Fukada & Nishiyama, 1972; Phelan, Mahler, & Cook, 1971), polyvinylidene Fluoride PVDF (Kawai, 1969; Fukada & Sakurai, 1971; Murayama, Nakamura, Obara, & Segawa, 1976), copolymers of PVDF with trifluorethylene

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TrFE (Higashihata, Sako, & Yagi, 1981) or tetrafluoroethylene TFE (Broadhurst, Davis, DeReggi, Roth, & Collins, 1982), and odd-numbered nylons (Kawai, 1969).

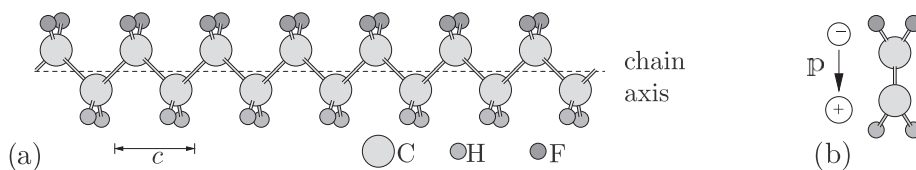
### 1.1. Microstructure of ferroelectric polymers

The characteristic properties of semicrystalline polymers are caused by their microstructural morphology. PVDF ( $\text{CH}_2\text{F}_2$ ) and its copolymers exhibit the largest piezoelectric activity. It is polymorphic and has at least four major crystalline phases, Lovinger (1982). Two of them, the  $\beta$ -phase and the  $\alpha$ -phase are most relevant for practical applications. In the  $\beta$ -phase, the dipoles are aligned in one direction as the fluorine atoms (F) are all positioned on one side of the polymer chain. Fig. 1 shows the polymer chain in a planar zigzag conformation. When crystallizing, two chains are packed into individual orthorhombic unit cells. It is noted in Fig. 2(a) that the fluorine atoms are positioned on one side of the unit cell, resulting in a *net dipole moment*. Because the unit cell is non-centrosymmetric this phase of PVDF causes the *piezoelectric properties*. Contrary, the primary thermodynamically stable  $\alpha$ -phase is composed such that dipole moments cancel out.

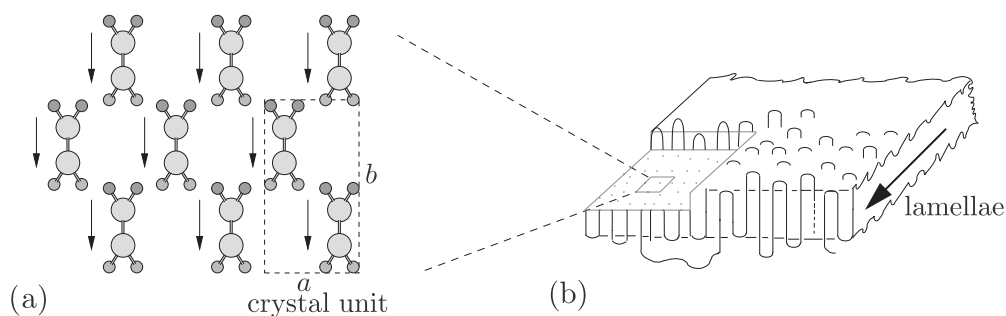
When polymers crystallize, the molecules typically form thin *lamellae*. The polymer molecules are oriented perpendicular to the surface of the lamellae and are folded in a way that they penetrate the lamellae many times, Kepler and Anderson (1992), see Fig. 2(b) for a schematic representation. A morphology of ferroelectric polymers consisting of crystallites dispersed in amorphous regions is shown in Fig. 3(a). Most polymers are *oriented* in order to improve physical properties such modulus, stiffness and recovery. This is achieved by *stretching or rolling* at temperatures below the crystallization temperature as indicated in Fig. 3(b). In the  $\beta$ -phase crystallites form *ferroelectric domains* which are polar, but initially *randomly* oriented within the sample. This accounts for the absence of an overall piezoelectric activity unless the sample is poled. Poling is accomplished by *electroding the polymer surfaces* with metal films, followed by application of a strong electric field to orient the crystallites as sketched in Fig. 3(c), inducing a crystalline *reorientation*. The amorphous phase of semicrystalline polymers supports the crystal orientation, and polarization is stable up to the Curie temperature.

### 1.2. Hysteresis phenomena on the macro-level

Ferroelectric materials like PVDF exhibit characteristic hysteresis phenomena on the macro-level due to above mentioned electric polarization mechanisms on the micro-level. For applied cyclic electric fields high enough to induce ferroelectric switching, strains in the thickness direction follow a *butterfly hysteresis* while the electric displacement shows a *square hysteresis* loop as shown in Fig. 4. Depending on different material compositions and pre-treatments, the shape of the hysteresis loops differs substantially. Hystereses measured on a uniaxially drawn PVDF polymer are depicted in Fig. 4(a). The polymer performs a conventional but rather rounded dielectric hysteresis loop when compared to the hysteresis of undrawn VDF-TrFE copolymers in Fig. 4(b). The *coercive field*  $E_c$  marks the point where the hysteresis intersects the horizontal axis. The *remanent polarization*  $p^r$  corresponds to the point where the loop intersects the vertical axis. Caused by the structure of the crystal unit cell, polymers and copolymers of VDF become thickest when their polarization reverses and thinnest



**Fig. 1.** A schematic view of  $\beta$ -phase: (a) Polymer chain of carbon (C), hydrogen (H) and fluorine (F) atoms in the planar zigzag conformation with repeated crystal unit having lattice dimension  $c = 2.56 \text{ \AA}$ . (b) View parallel to the chain axis with spatial distribution of the charges inducing polarization.



**Fig. 2.** (a) Crystal structure of  $\beta$ -phase containing two chains forming a crystal unit and having lattice dimensions  $a = 8.58 \text{ \AA}$  and  $b = 4.90 \text{ \AA}$ . (b) Several chains aligned and folded yield a polymer crystal (Figure redrawn from Wang et al. (1988)).

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