

A thermodynamically consistent formulation for transversely isotropic nonlinear ferroelectric hysteresis

H. Romanowski*, J. Schröder

Institute of Mechanics, University of Duisburg-Essen, Campus Essen, Department of Civil Engineering, 45117 Essen, Germany

Abstract

In this investigation we discuss a thermodynamic consistent model for an assumed transversely isotropic ferroelectric crystal. Due to the ferroelectric phase transition, that occurs at the so-called electric coercive field, we observe, within a certain range of temperature and under applied oscillating electric fields, two leading hysteresis effects. These are the polarization-electric-field and the strain-electric-field (butterfly) hysteresis loop. The main goal of this paper is to capture these fundamental characteristics of such ceramics.

Keywords: Butterfly-hysteresis; Ferroelectric; Invariant-formulation; Polarization

1. Introduction

In the last years, a variety of microscopically and thermodynamically motivated constitutive models for the description of ferroelectric ceramics has been proposed in the literature. A thermodynamically consistent formulation of the electro-mechanical hysteresis can be found in [1]. In these contributions concepts of plasticity theory including yield surfaces and isotropic and kinematic hardening, based on the Helmholtz free energy, are used. Based on these concepts several approaches for thermodynamically motivated constitutive laws of ferroelectric ceramics have been developed, see e.g. [2,3,4,5]. In this paper we present a thermodynamically consistent model for transversely isotropic ferroelectrics within the framework of the invariant theory, based on the coordinate invariant formulations proposed in [6,7].

2. Basic equations

The basic kinematic variable is the linear strain tensor, which is defined by the symmetric part of the displacement gradient, i.e. $\boldsymbol{\varepsilon}(\mathbf{x}) := 1/2(\text{grad}\mathbf{u} + \text{grad}^T\mathbf{u})$. The basic electric field variable, the electric field vector \mathbf{E} , is given by $\mathbf{E}(\mathbf{x}) := -\text{grad}\phi$, where ϕ is the electric potential. For the quasi-static case and neglected free

extrinsic charges the governing field equations are the equation of momentum $\text{div}\boldsymbol{\sigma} + \bar{\mathbf{f}} = \mathbf{0}$ and the Gauss equation $\text{div}\mathbf{D} = 0$. Here $\boldsymbol{\sigma}$ represents the symmetric Cauchy stress tensor, $\bar{\mathbf{f}}$ the given body force and \mathbf{D} the vector of electric displacements. The surface of the body is decomposed in mechanical $\partial\mathcal{B} = \partial\mathcal{B}_u \cup \partial\mathcal{B}_\sigma$ and electrical parts $\partial\mathcal{B} = \partial\mathcal{B}_\phi \cup \partial\mathcal{B}_D$, with $\partial\mathcal{B}_u \cap \partial\mathcal{B}_\sigma = \emptyset$ and $\partial\mathcal{B}_\phi \cap \partial\mathcal{B}_D = \emptyset$. The boundary conditions are defined by $\mathbf{u} = \bar{\mathbf{u}}$ on $\partial\mathcal{B}_u$ for the displacements, $\bar{\mathbf{t}} = \boldsymbol{\sigma}\mathbf{n}$ on $\partial\mathcal{B}_\sigma$ for the surface tractions, $\phi = \bar{\phi}$ on $\partial\mathcal{B}_\phi$ for the electric potential and $-\bar{\delta} = \mathbf{D}\mathbf{n}$ on $\partial\mathcal{B}_D$ for the electric charge.

3. Thermodynamic formulation

The basic assumption in the proposed model is the additive split of the strains $\boldsymbol{\varepsilon}$ and the vector of electric displacements \mathbf{D} into their reversible (\bullet^e) and remanent parts (\bullet^r), i.e.

$$\boldsymbol{\varepsilon} = \boldsymbol{\varepsilon}^e + \boldsymbol{\varepsilon}^r \text{ and } \mathbf{D} = \mathbf{D}^e + \mathbf{P}^r \quad (1)$$

where \mathbf{P}^r denotes the vector of remanent polarization. Let us now assume the existence of a thermodynamic potential $H = H(\boldsymbol{\varepsilon}, \boldsymbol{\varepsilon}^r, \mathbf{E}, \mathbf{P}^r)$, which acts as an electric enthalpy function. The observable variables are the total strains $\boldsymbol{\varepsilon}$ and the electric field \mathbf{E} whereas the remanent quantities describe the internal state of the material. The second law of thermodynamics yields $\mathcal{D} = \boldsymbol{\sigma} : \dot{\boldsymbol{\varepsilon}} - \mathbf{D}\dot{\mathbf{E}} -$

* Corresponding author. Tel.: +49 201 183 2701; Fax: +49 201 183 2680; E-mail: holger.romanowski@uni-essen.de

$\dot{H} \geq 0$, if we neglect thermal effects, which leads to the following inequality:

$$\begin{aligned} \mathcal{D} &= (\boldsymbol{\sigma} - \partial_{\boldsymbol{\varepsilon}} H) : \dot{\boldsymbol{\varepsilon}} - (\mathbf{D} + \partial_E H) \cdot \dot{\mathbf{E}} - \partial_{\boldsymbol{\varepsilon}'} H : \dot{\boldsymbol{\varepsilon}}' \\ &\quad - \partial_{\mathbf{P}'} H \cdot \dot{\mathbf{P}}' \geq 0 \end{aligned} \quad (2)$$

Since Eq. (2) has to be fulfilled for all possible thermodynamic processes, we obtain the following constitutive equations for the stresses and electric displacements:

$$\boldsymbol{\sigma} = \partial_{\boldsymbol{\varepsilon}} H \quad \text{and} \quad \mathbf{D} = -\partial_E H \quad (3)$$

respectively. For the thermodynamic forces associated to the remanent quantities we introduce the abbreviations $\tilde{\boldsymbol{\sigma}} := -\partial_{\boldsymbol{\varepsilon}'} H$ and $\tilde{\mathbf{E}} := -\partial_{\mathbf{P}'} H$, which lead to the reduced dissipation inequality:

$$\mathcal{D} = \tilde{\boldsymbol{\sigma}} : \dot{\boldsymbol{\varepsilon}}' + \tilde{\mathbf{E}} \cdot \dot{\mathbf{P}}' \geq 0 \quad (4)$$

In order to describe the evolution of the remanent variables the existence of a dissipation potential is assumed, which is expressed as a continuous, convex scalar-valued function of the flux variables $\dot{\boldsymbol{\varepsilon}}'$ and $\dot{\mathbf{P}}'$. Applying a Legendre–Fenchel transformation leads to a corresponding potential, that can be formulated in terms of the dual quantities. Therefore we introduce a switching surface $\Phi(\tilde{\boldsymbol{\sigma}}, \tilde{\mathbf{E}}) \leq 0$, formulated in terms of the dual variables. Expecting that the principle of maximum remanent dissipation can be seen as a generalization of the principle of maximum dissipation, we construct the Lagrangian functional:

$$\mathcal{L}(\tilde{\boldsymbol{\sigma}}, \tilde{\mathbf{E}}, \lambda) = -D(\tilde{\boldsymbol{\sigma}}, \tilde{\mathbf{E}}) + \lambda \Phi(\tilde{\boldsymbol{\sigma}}, \tilde{\mathbf{E}}) \quad (5)$$

with the Lagrange multiplier λ . The optimization conditions $\partial_{\tilde{\boldsymbol{\sigma}}} \mathcal{L} = 0$, $\partial_{\tilde{\mathbf{E}}} \mathcal{L} = 0$ and $\partial_{\lambda} \mathcal{L} = 0$ lead to the associated flow rules of the remanent variables:

$$\dot{\boldsymbol{\varepsilon}}' = \lambda \partial_{\tilde{\boldsymbol{\sigma}}} \Phi(\tilde{\boldsymbol{\sigma}}, \tilde{\mathbf{E}}) \quad \text{and} \quad \dot{\mathbf{P}}' = \lambda \partial_{\tilde{\mathbf{E}}} \Phi(\tilde{\boldsymbol{\sigma}}, \tilde{\mathbf{E}}) \quad (6)$$

and the loading/unloading conditions $\lambda \geq 0$, $\Phi(\tilde{\boldsymbol{\sigma}}, \tilde{\mathbf{E}}) \leq 0$ and $\lambda \Phi(\tilde{\boldsymbol{\sigma}}, \tilde{\mathbf{E}}) = 0$. It should be noted that the normality rule is sufficient to satisfy the second law of thermodynamics.

4. Invariant formulation and polynomial basis

The governing constitutive equations have to represent the material symmetries of the considered body. Therefore the representation theorem of isotropic tensor functions is used, in order to formulate the explicit invariant constitutive equations. The values of the enthalpy function have to be invariant under all transformations \mathbf{Q} of the material symmetry group \mathcal{G}_{ii} , i.e.

$$H(\boldsymbol{\varepsilon}, \boldsymbol{\varepsilon}', \mathbf{E}, \mathbf{P}') = H(\mathbf{Q}\boldsymbol{\varepsilon}\mathbf{Q}^T, \mathbf{Q}\boldsymbol{\varepsilon}'\mathbf{Q}^T, \mathbf{Q}\mathbf{E}, \mathbf{Q}\mathbf{P}') \quad \forall \mathbf{Q} \in \mathcal{G}_{ii} \quad (7)$$

this reflects the geometrical and physical symmetries of the anisotropic solid. The main idea of the invariant theory is the extension of the \mathcal{G}_{ii} -invariant functions, see Eq. (7), to functions which are invariant under a larger group of transformations. Since we want to focus on transversely isotropic crystals, whose anisotropy class is characterized only by one preferred direction, we introduce the polar vector \mathbf{a} as an additional argument. Applying the so-called principle of isotropy of space, see e.g. [8], leads to the following representation of the enthalpy function:

$$\begin{aligned} H(\boldsymbol{\varepsilon}, \boldsymbol{\varepsilon}', \mathbf{E}, \mathbf{P}', \mathbf{a}) &= H(\mathbf{Q}\boldsymbol{\varepsilon}\mathbf{Q}^T, \mathbf{Q}\boldsymbol{\varepsilon}'\mathbf{Q}^T, \mathbf{Q}\mathbf{E}, \mathbf{Q}\mathbf{P}', \mathbf{Q}\mathbf{a}) \\ &\quad \forall \mathbf{Q} \in \mathcal{O}(\ni) \end{aligned} \quad (8)$$

which is the definition of an isotropic tensor function with respect to the whole set of arguments. The invariance group \mathcal{G}_{ii} of the electro-mechanically coupled solid can be obtained by the so-called Principle of the Superposition of Symmetries and we define $\mathcal{G}_{ii} := \{\mathbf{Q} \in \mathcal{O}(3), \mathbf{Q}\mathbf{a} = \mathbf{a}\}$. For a detailed discussion of this topic see e.g. [9]. In order to formulate the electric enthalpy function we need a finite set of invariants, which builds the so-called polynomial basis. In the following we are not interested in the derivation of the whole basis and refer to Spencer [10], Wang [11] and Liu [9], in the context of electro-mechanical coupled systems we refer to Schröder et al. [6] and the references therein. For the finite set of vectors and tensors that is given by the symmetric tensors $\boldsymbol{\varepsilon}$, $\boldsymbol{\varepsilon}'$, the vectors \mathbf{E} , \mathbf{P}' and the preferred direction \mathbf{a} , with $\|\mathbf{a}\| = 1$, the basic and mixed invariants of interest are:

$$\begin{aligned} I_1 &:= \text{trace}[\boldsymbol{\varepsilon} - \boldsymbol{\varepsilon}'], \quad I_2 := \text{trace}[(\boldsymbol{\varepsilon} - \boldsymbol{\varepsilon}')^2], \\ I_4 &:= \text{trace}[(\boldsymbol{\varepsilon} - \boldsymbol{\varepsilon}')(\mathbf{a} \otimes \mathbf{a})] \\ I_5 &:= \text{trace}[(\boldsymbol{\varepsilon} - \boldsymbol{\varepsilon}')^2(\mathbf{a} \otimes \mathbf{a})], \quad J_1 := \text{trace}[(\mathbf{E} \otimes \mathbf{E})], \\ J_2 &:= \text{trace}[(\mathbf{E} \otimes \mathbf{a})] \\ K_1 &:= \text{trace}[(\boldsymbol{\varepsilon} - \boldsymbol{\varepsilon}')(\mathbf{E} \otimes \mathbf{a})], \quad \bar{N}^P := \mathbf{P}' \cdot \mathbf{a} \end{aligned} \quad (9)$$

Now the enthalpy function H is formulated in terms of these invariants and it should be noted that polynomial functions in elements of the polynomial basis are invariant under all transformations $\mathbf{Q} \in \mathcal{O}(3)$.

5. Model problem

In this paper the underlying thermodynamic potential is assumed to consist of five parts:

$$\begin{aligned} H &= H_1(\boldsymbol{\varepsilon}, \boldsymbol{\varepsilon}') + H_2(\mathbf{E}) + H_3(\boldsymbol{\varepsilon}, \boldsymbol{\varepsilon}', \mathbf{E}, \bar{N}^P) + \\ &\quad H_4(\mathbf{E}, \bar{N}^P) + H_5(\bar{N}^P) \end{aligned} \quad (10)$$

whereas the explicit expressions formulated in terms of the polynomial basis are:

$$\begin{aligned} H_1 &= \frac{1}{2}\lambda I_1^2 + \mu I_2 + \alpha_1 I_5 + \alpha_2 I_4^2 + \alpha_3 I_1 I_4 \\ H_2 &= \gamma_1 J_1 + \gamma_2 J_2^2 \\ H_3 &= [\beta_1 I_1 J_2 + \beta_2 I_4 J_2 + \beta_3 K_1] \frac{1}{P_s} \bar{N}^P =: \omega \bar{N}^P \\ H_4 &= -J_2 \bar{N}^P, \quad H_5 = f(\bar{N}^P) \end{aligned} \quad (11)$$

The coefficients $\{\lambda, \mu, \alpha_1, \alpha_2, \alpha_3\}$ characterize the mechanical, $\{\gamma_1, \gamma_2\}$ the dielectric and $\{\beta_1, \beta_2, \beta_3\}$ the piezoelectric material properties within the invariant formulation. In the linear piezoelectric range (neglecting H_4 and H_5 and setting \bar{N}^P equal to the maximum value of polarization), this parameters can directly be identified with their coordinate-dependent representation, see [6]. For $f(\bar{N}^P)$ we choose:

$$f(\bar{N}^P) = \frac{1}{c} \left[\bar{N}^P \operatorname{Artanh} \left(\frac{\bar{N}^P}{P_s} \right) + \frac{1}{2} P_s \ln \left(1 - \left(\frac{\bar{N}^P}{P_s} \right)^2 \right) \right] \quad (12)$$

where P_s is the maximum value of the remanent polarization and the parameter c affects the slope of the hysteresis-curves. For this specific model the explicit forms of the stresses and electric displacements appear with Eqs (3) as follows:

$$\begin{aligned} \boldsymbol{\sigma} &= (\lambda I_1 + \alpha_3 I_4) \mathbf{1} + 2\mu \boldsymbol{\varepsilon} + \alpha_1 [\mathbf{a} \otimes \boldsymbol{\varepsilon} \mathbf{a} + \boldsymbol{\varepsilon} \otimes \mathbf{a}] \\ &\quad + (2\alpha_2 I_4 + \alpha_3 I_1) \mathbf{a} \otimes \mathbf{a} \\ &\quad + [\beta_1 J_2 \mathbf{1} + \beta_2 J_2 \mathbf{a} \otimes \mathbf{a} + \frac{1}{2} \beta_3 (\mathbf{E} \otimes \mathbf{a} + \mathbf{a} \otimes \mathbf{E})] \frac{1}{P_s} \bar{N}^P \\ \mathbf{D} &= -2\gamma_1 \mathbf{E} - 2\gamma_2 J_2 \mathbf{a} - [(\beta_1 I_1 + \beta_2 I_4) \mathbf{a} + \beta_3 \mathbf{a} \boldsymbol{\varepsilon}] \frac{1}{P_s} \bar{N}^P + \mathbf{P}^r \end{aligned} \quad (13)$$

Here $\mathbf{P}^r = \bar{N}^P \mathbf{a}$ is the remanent polarization with respect to the polarization axis. With Eq. (13), the reduced dissipation inequality Eq. (4) takes the form:

$$-(\partial_{\boldsymbol{\varepsilon}^r} H_1 + \partial_{\boldsymbol{\varepsilon}^r} H_3) : \dot{\boldsymbol{\varepsilon}}^r - (\partial_{\bar{N}^P} H_3 + \partial_{\bar{N}^P} H_4 + \partial_{\bar{N}^P} H_5) \cdot \dot{\bar{N}}^P \geq 0 \quad (14)$$

According to the abbreviations given in Section 3, we recast this reduced inequality in $\bar{\boldsymbol{\sigma}} : \dot{\boldsymbol{\varepsilon}}^r + \bar{\mathbf{E}} \cdot \dot{\bar{N}}^P \geq 0$. Following McMeeking and Landis [4] this expression is reduced to $\bar{\mathbf{E}} \cdot \dot{\bar{N}}^P \geq 0$ by using the following constitutive assumption for the remanent strains:

$$\boldsymbol{\varepsilon}^r = \frac{\varepsilon_a^r}{P_s^2} \operatorname{dev}(\mathbf{P}^r \otimes \mathbf{P}^r) \quad (15)$$

where ε_a^r characterizes the maximum achievable remanent strain in direction of the polarization axis. This quadratic relationship between the remanent polarization and strains is a commonly reasonable assumption, see e.g. [12]. As a simple choice for the switching criterion we specify:

$$\Phi = \tilde{E}^2 - E_c^2 = (E - E^B)^2 - E_c^2 = (J_2 - E^B)^2 - E_c^2 \leq 0 \quad (16)$$

with $E^B := \partial \bar{N}^P H_3 + \partial \bar{N}^P H_5$, $E := -\partial \bar{N}^P H_4 = J_2$ and the coercive field strength E_c . For the loading case $\Phi = 0$, Eq (16) leads to the so-called back electric field:

$$E^B = \omega + f'(\bar{N}^P) \quad \text{with} \quad f'(\bar{N}^P) = \frac{1}{c} \operatorname{Artanh} \left(\frac{\bar{N}^P}{P_s} \right) \quad (17)$$

Resolving Eq. (17)₁ with respect to \bar{N}^P leads to a formula for the remanent polarization, governed by the strain and electric field quantities as the basic variables:

$$\bar{N}^P = P_s \tanh[c(E^B - \omega)] =: \tilde{f}(E^B - \omega) \quad (18)$$

6. Numerical examples

In the following examples the material parameters for the elastic stiffness tensor are chosen as $\mathbb{C}_{11} = 166$; $\mathbb{C}_{12} = 76.6$; $\mathbb{C}_{13} = 77.5$; $\mathbb{C}_{33} = 162$ and $\mathbb{C}_{44} = 42.9$ in units of $10^3 \text{ N}/(\text{mm}^2)$. The components of the piezoelectric tensor are set to $e_{31} = -4.4$; $e_{33} = 18.6$ and $e_{15} = 11.6$ in units of $10^{-3} \text{ N}/(\text{Vmm})$. The parameters for the dielectric tensor are set to $k_{11} = 1260 \cdot \epsilon_0 = 1.12$ and $k_{33} = 1420 \cdot \epsilon_0 = 1.26$ in units of $10^{-11} \text{ C}/(\text{Vmm})$, where $\epsilon_0 = 8.854 \cdot 10^{-15} \text{ C}/(\text{Vmm})$ is the permittivity of free space. For the corresponding values of the maximum achievable polarization and the coercive field we choose $P_s = 26 \cdot 10^{-8} \text{ C}/(\text{mm}^2)$ and $E_c = 1000 \text{ V}/(\text{mm})$, whereas the maximum remanent strain along the poling direction, see Eq. (15), is set to $\varepsilon_a^r = 0.001$. The calculated hysteresis loops for a simply supported bar under a cyclic varying electric field, see Fig. 1, are given in Fig. 2.

In the next example we consider a square plate ($20 \times 20 \text{ mm}$) with a centred hole (radius 3 mm) and the boundary conditions for the displacements and the time varying electric potential given in Fig. 3(a). Figure 3(b) illustrates the distribution of the electric potential at time $t = 3.0 \text{ s}$ within the specimen and Fig. 3(c) depicts the variation of the applied electric potential versus time. The preferred direction \mathbf{a} points in vertical direction and the parameter c is set to 0.0002. During the cyclic loading of the specimen, the polarization process is initiated in regions where the switching criterion Eq. (16) is identically fulfilled, i.e. $\Phi = 0$. Figure 4 depicts the

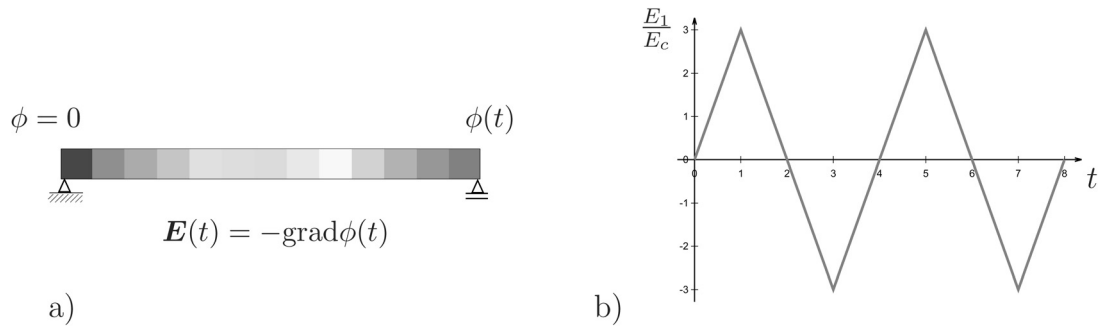


Fig. 1. a) Boundary conditions and relative distribution of the electric potential and b) variation of electric field versus time.

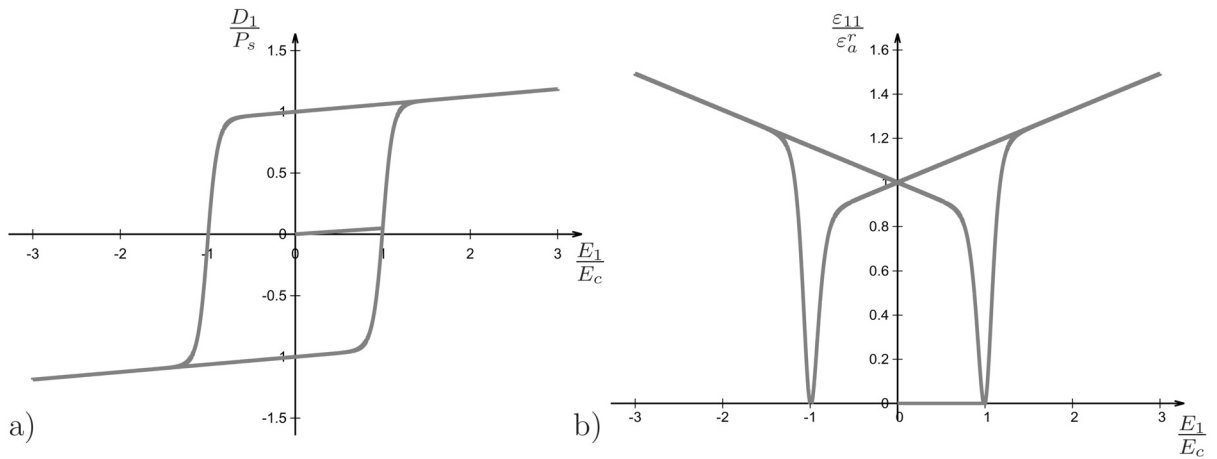


Fig. 2. Standardized hysteresis curves for $c = 0.008$: a) electric-displacement electric-field and b) strain electric-field (butterfly) hysteresis loop.

distribution of the remanent polarization for different time steps, where the maximum achieved polarization in preferred direction for $t = 1.0$ s is given in Fig. 4(a). The maximum achieved polarization in opposite direction for $t = 3.0$ s is illustrated in Fig. 4(b) and Fig. 4(c) depicts the distribution of the remaining remanent polarization in the absence of an electric field.

7. Conclusions

The main idea of the proposed model is an additive split of the strain tensor and the vector of electric displacements into reversible and remanent parts. In order to simplify the model we have made a constitutive assumption for the remanent strains, relating the remanent strains directly to the remanent polarization and we have neglected the topic of ferroelasticity and the effect of mechanical depolarization. Based on these assumptions we have presented a thermodynamically consistent

phenomenological model for an assumed transversely isotropic ferroelectric crystal, where the anisotropic behavior of the electro-mechanical coupled material has been captured by an invariant formulation of the constitutive equations.

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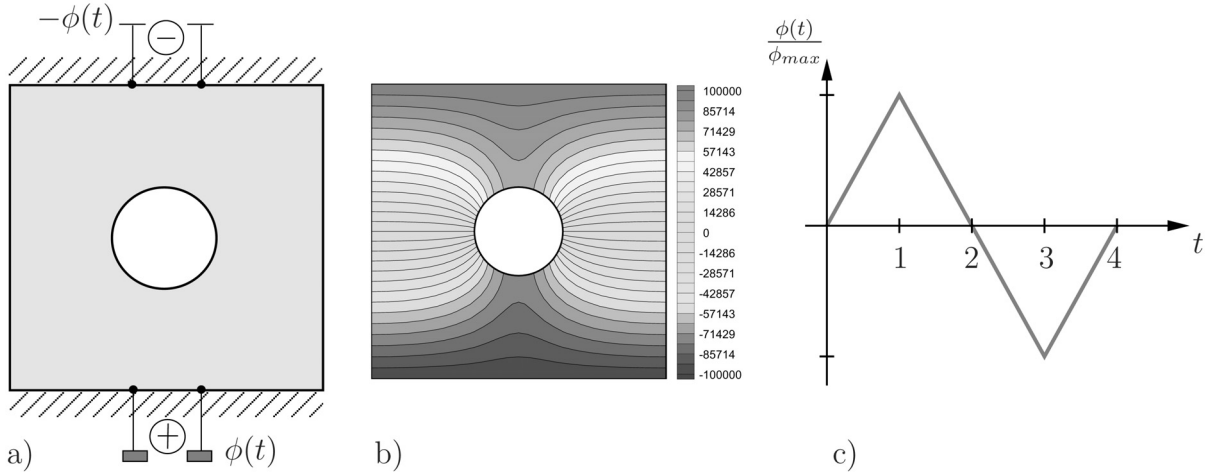


Fig. 3. Square plate with a centered hole: a) system and boundary conditions; b) distribution of the electric potential ϕ in [V] for $t = 3.0$ s and c) varying electric potential at the upper boundary versus time.

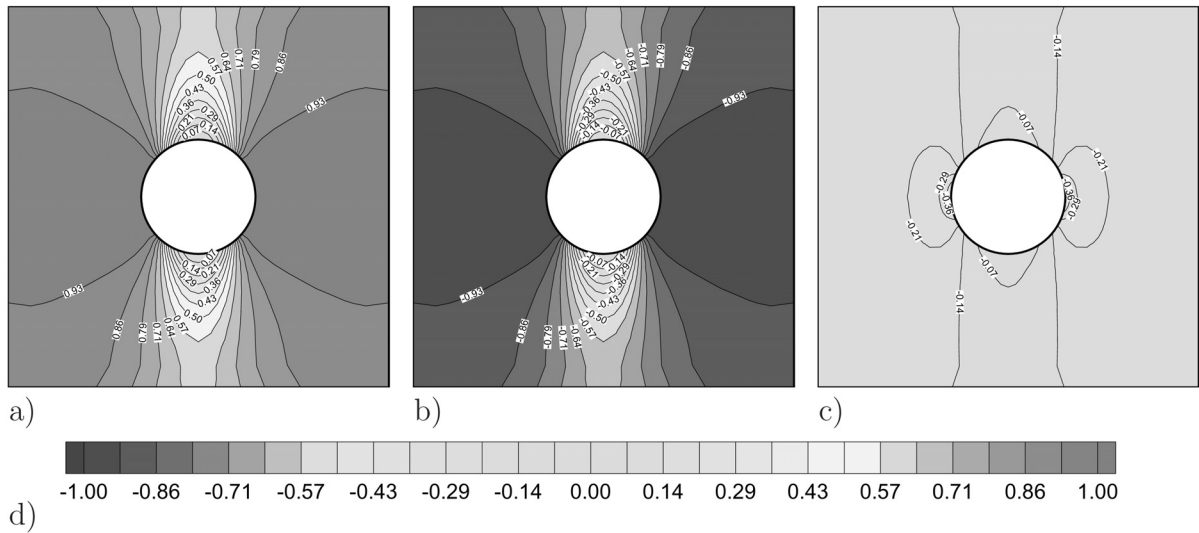


Fig. 4. Distribution of $\bar{N}^P = \mathbf{P}^f \cdot \mathbf{a}$ divided by P_s for: a) $t = 1.0$ s; b) $t = 3.0$ s; c) $t = 4.0$ s; d) contour legend for the standardized values.

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