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Polarisation kinetics of electrically connected electroconductive ferroelectric multilayer structures

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Abstract

In this article, the polarisation kinetics of an electrically connected ferroelectric bimorph structure is described with an electric model by means of equivalent circuit analysis. This model results in a system of ordinary differential equations (ODEs) that can be solved numerically. A bimorph model structure consisting of wire-connected layers of barium–titanate–stannate ceramics was poled. The conductivities of the layers can be estimated by comparing of the measured electric field in the layers and the results of the poling model. In order to consider hysteretic effects, the Preisach model of hysteresis is included into the poling model. Within the ODEs, the slope of the hysteresis loop is needed. An algorithm for estimating it via the Preisach model was developed. This approach is based upon linear interpolation between adjacent grid points of the discretised electric field within the Preisach model. An example shows the capabilities of this method for considering poling, switching effects and depolarisation.

Keywords: Composites; Ferroelectric properties; $BaTiO_3$ and titanates; Capacitors; Preisach model

1. Introduction

In recent years developing and describing of functionally graded materials (FGM) have represented one focus of interest. In particular FGM with a gradient of piezoelectric properties are important due to their application for ultrasonic transducers or bending actuators.[1,2] In FGM consisting of ferroelectric ceramics, the macroscopic polarisation vanishes because of the randomly orientated domains. A poling process is necessary, in order to impress the piezoelectric properties. From a microscopic point of view, the poling process is a sophisticated problem resulting in the macroscopic well-known hysteresis of polarisation versus electric field. For describing and optimising the time-dependent poling behaviour, the macroscopic approach is adequate as long as there is a lack of first-principle calculations or they are too costly.

Multilayer systems can be regarded as model structures for FGM. An understanding of the poling process in a wide parameter range can be reached by an equivalent electric circuit analysis.[3] This method succeeded in the field of ferroelectric capacitors with their capabilities for ferroelectric-based non-volatile memories.[4,5]

This article presents the basics of equivalent circuit analysis. Knowing the electric properties of the single layers, the time-dependent electrical behaviour of the layers within the multilayer structure can be calculated. For that purpose, the derivation of the polarisation with respect to the electric field must be included into the model. Using the Preisach model for hysteresis, an algorithm for its estimation was developed.

2. Equivalent circuit analysis

In the following calculations a system consisting of two layers is regarded. Supposing the two different layers of the bimorph structure as homogeneous, an equivalent electrical circuit analysis according to Or[3] can be done by considering each layer as a RC-element. Each RC-element consists of a capacitor with ferroelectric and a resistance connected in parallel. These RC-elements are connected in serial (Fig. 1a).

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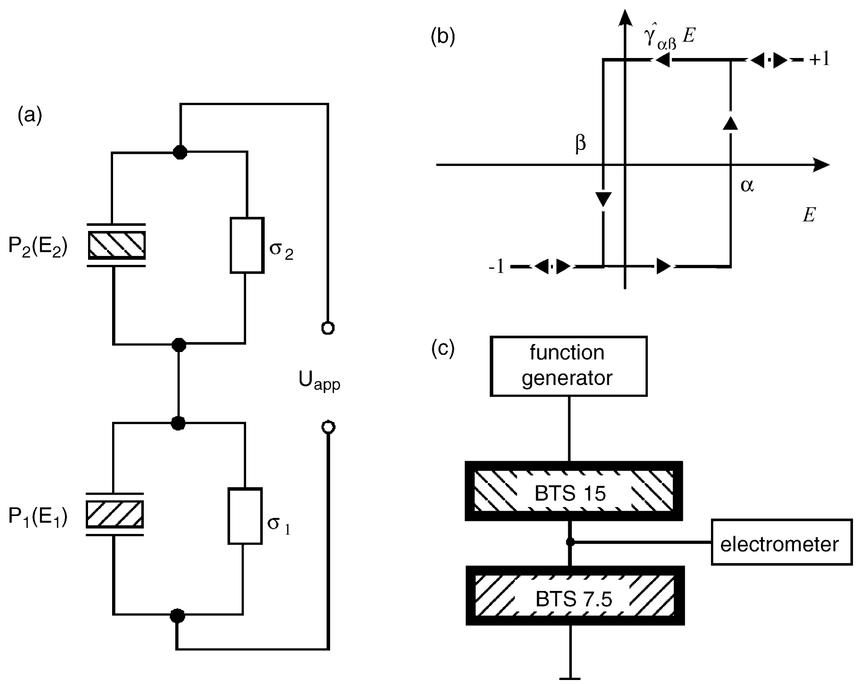

Fig. 1. Equivalent circuit for the investigated multilayer structure (a), elementary hysteresis operator (b) and bimorph model structure used in the experiment (c).

The dielectric displacement in each layer is

$$D_k = \varepsilon_0 E_k + P_k, \quad k = 1, 2, \tag{1}$$

where ε_0 is the permittivity of free space, E is the electric field strength and P is the electric polarisation.

The current density j is composed of the conductive current and the displacement current. It is constant through each of the layers, namely

$$j = j_1 \overset{!}{=} j_2 = \sigma_1 E_1 + \dot{D}_1 = \sigma_2 E_2 + \dot{D}_2. \tag{2}$$

The conductivity σ is as a first approximation supposed to be constant. In general, the polarisation P depends on $E(t)$ and t. If the polarisation responds to the electric field much faster than it is changed by other effects, the explicit time-dependence can be neglected. Thus, the derivation with respect to time is

$$\frac{\mathrm{d}P(E(t))}{\mathrm{d}t} = \frac{\partial P}{\partial E}\frac{\partial E}{\partial t} = P'(E)\dot{E}(t). \tag{3}$$

The relation between polarisation and electric field will be discussed in the next section.

Eqs. (2) and (3) yield one differential equations, namely

$$\sigma_1 E_1 + \varepsilon_0 \dot{E}_1 + P_1' \dot{E}_1 = \sigma_2 E_2 + \varepsilon_0 \dot{E}_2 + P_2' \dot{E}_2. \tag{4}$$

For determining the two unknown $E_k(t)$, one further equation is necessary. Because the RC-elements are connected in serial, the sum of the impressed voltages on each layer is equal to the applied voltage U_{app} on the multilayer structure. Differentiating the corresponding electric fields with respect to time results in

$$\dot{E}_{app} = \frac{\dot{U}_{app}}{d} = \frac{d_1}{d}\dot{E}_1 + \frac{d_2}{d}\dot{E}_2, \tag{5}$$

where E_{app} is the applied electric field, d is the thickness of the multilayer structure and d_k is the thickness of the kth layer.

If E_1 and E_2 do not vary in their dependency on time, Eq. (4) shows saturated behaviour, namely

$$\frac{\sigma_1}{\sigma_2} = \frac{E_2}{E_1}. \tag{6}$$

Eq. (6) is approximately valid, too, if the polarisations are in the saturated range and the conductive currents are great in comparison with $\varepsilon_0 \dot{E}_k$ in both layers.

A charge density q is formed between the layers, if the dielectric displacements D of the particular layers are different, namely

$$q = D_2 - D_1. \tag{7}$$

By knowing the conductivities, the applied electric field (with respect to time) and the hysteresis loop of each layer, the electric field and the polarisation within each layer, the interfacial charge density and the current density through the multilayer structure can so be calculated with respect to time.

3. Preisach fitting of the hysteresis

Saturated hysteresis loops can described well by relations using the tanh-function.[4,5] Such models are not based on completed models of hysteresis. So the nonsaturated hysteresis loops are merely assumed to be rather arbitrarily scaled saturated loops.

A more flexible model for describing the complete dielectric hysteretic behaviour without considering first-principle calculations is preferable. The Preisach[6] model provides such an abstract approach for calculation of hysteresis loops.[7] It can be applied to ferroelectrics.[8,9]

The Preisach model is based on the weighted superposition of elementary hysteresis operators.

The elementary hysteresis operator $\hat{\gamma}_{\alpha\beta}(E)$ depends on input electric field E (Fig. 1b). They are shaped rectangularly and characterised by a upper (α) and a lower (β) coercive field strength ($\alpha \geq \beta$). Their output is an elementary polarisation given by

$$\hat{\gamma}_{\alpha\beta}E = \begin{cases} +1, & \text{for } E > \alpha \\ -1, & \text{for } E < \beta \end{cases}. \qquad (8)$$

For electric fields between the limiting coercive fields, the last output of the elementary hysteresis operator is retained. The resulting hysteresis is constructed as a superposition of elementary hysteresis operators, according to

$$P = \iint_{\alpha \geq \beta} \mu(\alpha, \beta)\hat{\gamma}_{\alpha\beta}E \, \mathrm{d}\alpha \, \mathrm{d}\beta, \qquad (9)$$

where $\mu(\alpha,\beta)$ is a weighting function, that defines the shape of the resulting hysteresis loop. The output of the elementary hysteresis operator $\hat{\gamma}_{\alpha\beta}$ depends on its previous output, which must be stored. So, a discretisation in regard to the electric field is necessary for numerical calculation and the integrals in (9) become sums.

Moreover, not the polarisation itself is needed but the derivation $P'(E)$. Therefore an algorithm for estimating $P'(E)$ on the discrete grid of electric fields was developed using linear interpolation between the two adjacent grid points with higher or lower electric field. Because only the grid points are included in the hysteresis calculation, the grid points already passed through are included in the storage process of the model. The polarisation of the other grid point is calculated without storage of the associated elementary polarisations.

If the electric fields changes between increasing and decreasing and vice versa, first of all a step to the grid point in the old direction is done with storage of the elementary polarisations, in order to take account of the hysteresis. Then, steps in the resulting polarisation can occur. They become smaller, if the discretisation is chosen finer.

4. Results

The system of ordinary differential Eqs. (4) and (5), is solved numerically via the classical fourth-order Runge–Kutta method.

The model structure (Fig. 1c) was composed of two layers consisting of barium-titanate-stannate $Ba(Ti,Sn)O_3$ with 7.5 mol% tin (BTS 7.5) and 15 mol% tin (BTS 15), respectively. Each of them was 0.59 mm thick and they were connected with wires. A voltage increasing within 1 second from 0 up to 1.5 kV and then leaving constant was applied. The voltage in one layer was measured with a high-impedance electrometer Keithley 6514.

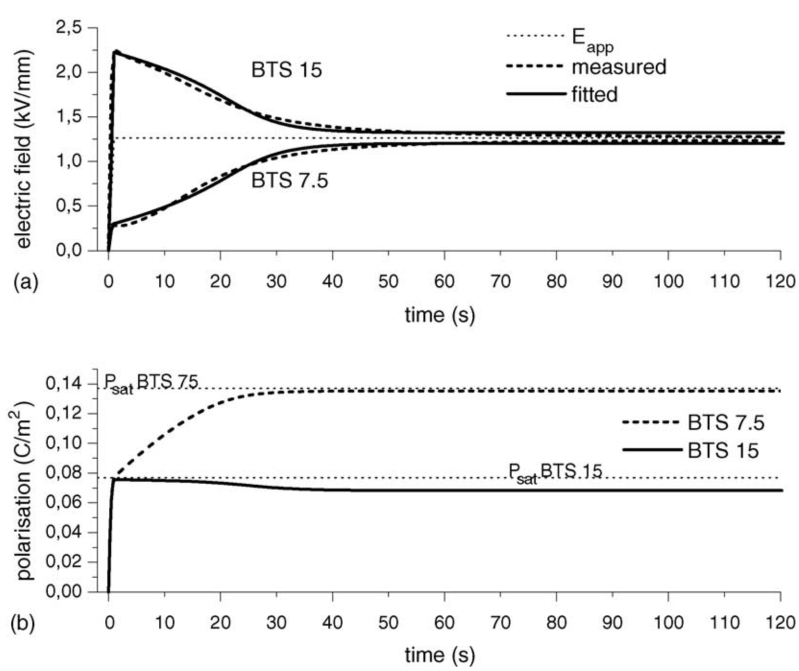

Fig. 2. Time-dependence of the measured and the modelled electric field (a) in the bimorph model structure, modelled polarisation behaviour (b) in the BTS 7.5 and the BTS 15 layer in comparison with the saturated polarisations.

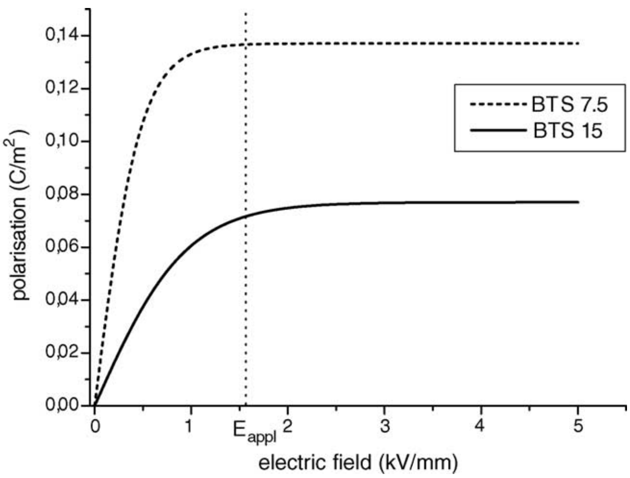

Fig. 3. Virgin loops of BTS 7.5 and BTS 15 fitted with tanh-functions.

The results converted in electric fields are shown in Fig. 2(a). The conductivities of both layers were estimated by fitting with the poling model. The $P(E)$ relation was introduced into the model by fitting the measured virgin-loop with a tanh-function (Fig. 3). Using them, the poling model (4) and (5) was solved repeatedly with varying the conductivities of both layers. The deviations between fitted and measured data on discrete time-points were minimized. Keeping in mind the saturation behaviour (6) and using a step-size controlled Runge–Kutta algorithm, this “try and error”-method is rather fast. As results, the conductivities are approximately $1.9 \times 10^{-9}\ \Omega^{-1}\ \mathrm{m}^{-1}$ in BTS 15 and $2.1 \times 10^{-9}\ \Omega^{-1}\ \mathrm{m}^{-1}$ in BTS 7.5.

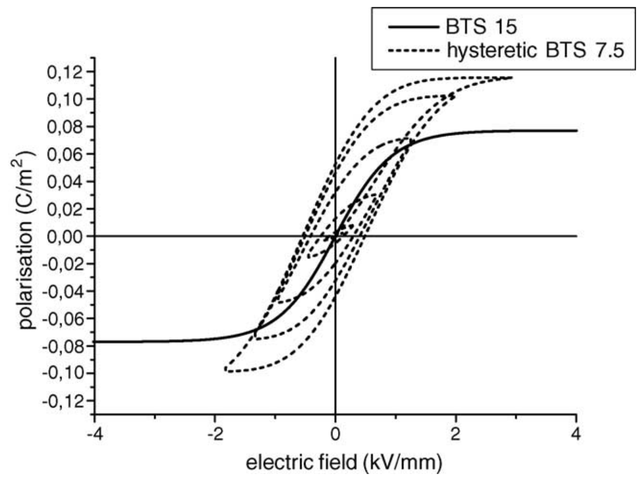

Fig. 5. Demonstration of the equivalent circuit analysis including the Preisach model. Hysteresis loops for the process of poling and depolarisation as shown in Fig. 4. The hysteresis loop of the hysteretic BTS 7.5 is arbitrary excepting the value of the saturated polarisation.

As shown in Fig. 2(b), the poling process takes approximately one minute. The maximum polarisation within the BTS 15 layer is reached as soon as the full voltage applies on the bimorph model structure. At the beginning, the polarisation in the other layer is the same, according to (7), because the interface is not yet charged. This border case was modelled in previous works of our group as poling neglecting the conductivity.[10]

Later, the polarisation in BTS 15 decreases little and the polarisation of BTS 7.5 increases permanent. Because of the

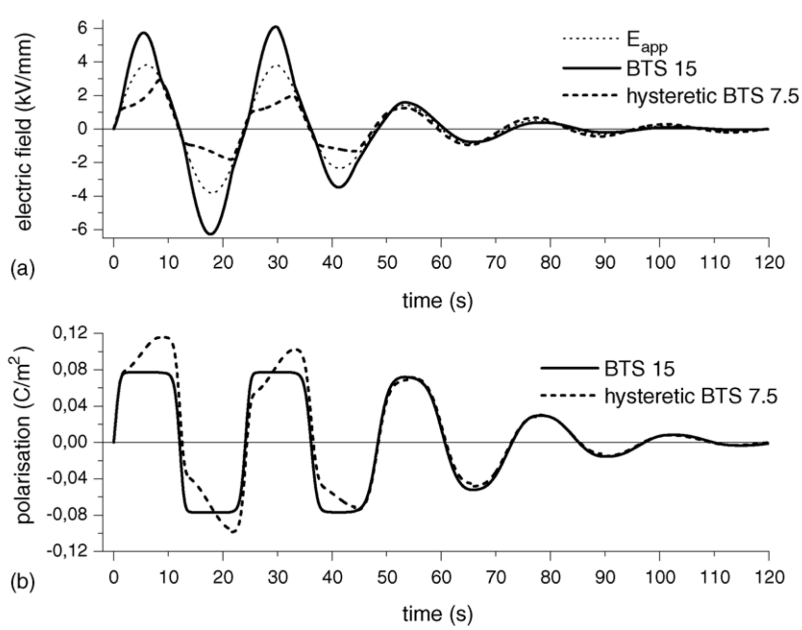

Fig. 4. Demonstration of the equivalent circuit analysis including the Preisach model. Poling and depolarisation by cycling with decaying amplitude of the applied electric field in a bimorph model structure. Time-dependence of electric fields (a) and polarisations (b). The underlying hysteresis loop of the hysteretic BTS 7.5 is arbitrary excepting the value of the saturated polarisation.

relaxor-like behaviour of BTS 15, hysteretic effects are small and can be neglected.

The response to arbitrary applied electric fields was calculated with the developed algorithm involving the Preisach modell. As a test structure, a bimorph consisting of BTS 15 and hysteretic BTS 7.5 in each case with conductivities $2 \times 10^{-9}\,\Omega^{-1}\,\mathrm{m}^{-1}$ was regarded. The same non-hysteretic virgin-loop as before was implemented for the BTS 15 layer. The modelled hysteresis loop of the hysteretic BTS 7.5 differs from the measured one. Even if the coercive field and the remanent polarisation are arbitrary values, the saturated polarisation is supposed to be the same as the measured one in BTS 7.5.

Fig. 4 shows the electric fields and the polarisations corresponding to a poling process with following depolarisation by cylcing with decaying amplitudes. The resulting hysteresis loops are shown in Fig. 5. These results demonstrate the capabilities of the developed poling model.

5. Conclusion

An electric equivalent circuit analysis was adopted, in order to calculate the polarization kinetics of an electrically connected bimorph model structure consisting of BTS 7.5 and BTS 15. By comparing the measured and the calculated response on an applied electric field, the conductivities of the layers were estimated as $2 \times 10^{-9}\,\Omega^{-1}\,\mathrm{m}^{-1}$. By using the Preisach model of hysteresis, partial polarisation and switching can be taken into account. The combination of the equivalent circuit analysis and the Preisach model of hysteresis can describe the polarisation kinetics of electrically connected ferroelectric multilayer systems for complex poling regimes.

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