Modeling strain and dielectric hysteretic type dependences in polycrystalline ferroelectrics by methods of two-level continuum

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Abstract. A one-dimensional model is represented for electrical and mechanical response of polycrystalline ferroelectric material when exposed to electric field and coaxial him mechanical stress great intensity. The equations describing the residual polarization and deformation are obtained in the form of equations with differentials, which can also be regarded as finite-difference equations. The reversible or induced components par with residual or nonreversible components of polarization and deformation were introduced. The combination of these components allows us to construct the dielectric and deformation hysteresis loops. It is shown that for a specific choice of the model parameters, it is possible to describe the experimental curves not only qualitatively but also quantitatively. Model can be used in finite element analysis to describe the irreversible processes of polarization and depolarization of ferroelectric ceramics.

1. Introduction
It is well known that impact of the electric fields and mechanical stresses on ferroelectric polycrystalline sample causes therein the deformation and polarization. If the external forces are of sufficient magnitude, then begins an irreversible process associated with the appearance of an irreversible or residual deformation and polarization, and nonlinear response of the material. The physical basis of the appearance of residual polarization is an integral characteristic of the vectors of spontaneous polarization. In unpolarized state in each crystal the vectors of spontaneous polarization can have any direction, differing from each other at fixed angles. For example, in a perovskite type ferroelectric material these angles are 180 and 90 degrees. Each crystallite is oriented also arbitrarily. Therefore, in the unpolarized state in a representative volume the set of all vectors of spontaneous polarization is distributed evenly in all directions. The resulting polarization of the representative volume is equal zero. Application of an electric field leads to the fact that the vectors of spontaneous polarization turn in the direction of the electric field as far as it allows the crystal structure of the ferroelectric. As a result, the total polarization of the representative volume is already different from zero. In this case ordinary say that the process of polarization is begins. Impact of mechanical stress can also lead to the turns of the vectors of spontaneous polarization that in the final result affects on the polarization of a representative volume. Mechanical stress, being a tensor of second rank, can both promote and inhibit of polarization. In the latter case we speak about the depolarization ceramic by mechanical stresses. In this regard, the main objects of the experimental studies are the dependencies between the external loads in the form of electric fields and mechanical...
stresses on the one hand, and arising internal parameters of the deformation and polarization, on the other hand. In irreversible processes, such relationships are presented in the form of deformation and dielectric hysteresis loops. For circular processes the response of material is described by the big hysteresis loop. For stepwise increasing and decreasing external loads is observed a small hysteresis loop. The process of appearance of the residual polarization by the electric field is called polarization. In homogeneous electric field the ceramics is brought as generally to a state of saturation polarized state, i.e. a state where a further increase of the electric field do not alter the polarization state. Application, for example, a mechanical compressive stresses can depolarize ceramics partially or completely. The appearance of polarization in a representative volume leads to polar direction and change electrical and mechanical properties of the material. As a result, the class of material anisotropy is changed. If the material is not polarized, it is isotropic, both with respect to the electrical and mechanical properties. Polarized ceramics is locally transversely isotropic material with the axis of isotropy coinciding with the direction of the residual polarization. Based on the above we can conclude that the external electric field and mechanical stress can alter polarized and deformed state, i.e. change the class of local anisotropy of the material. Therefore, the modeling of nonlinear behavior irreversible process of polarization and depolarization such materials are of considerable interest for engineering and technological developments. To problems of construction mathematical models of nonlinear irreversible polarization at recently been given more attention. We note some directions of modeling such problems. In [1-4] presented mathematical models to describe the polarization based on the mathematical theory of plasticity. In [5-9] given the averaging methods by which one can construct a hysteresis dependence. Another focus is the method of micro-mechanical model in which a representative volume is considered as a set of unit cells, where the vector of spontaneous polarization can take place their position in a certain direction. The transition from one position to another is given by certain geometric or energetic criteria. These models are presented in [10-17]. Closely adjacent to this the model is described in [18-21]. Here the density distribution of the vectors of spontaneous polarization based on Boltzmann statistics was introduced. Further the energy balance between the energy needed to breaking the locking mechanisms of domain walls and work of external electric fields was formulated. More general list of published works in these directions can be found in [22, 23]. Our model is closest to the last of the models. But it has significant differences concerning the simultaneous action of both electrical and mechanical fields. Besides this we have a completely different view on induced polarization and strain components. This approach allowed us to describe the strain and dielectric small hysteresis loops not only qualitatively but also quantitatively.

2. Formulation of the problem and method of solution

2.1. Subject of investigation

We consider the representative volume of polycrystalline ferroelectric material (ceramics), which contains a many crystallites and each of the crystallites contains many domains. The walls of domains are fixed existing inhomogeneities presenting in the material. Without going into the physical nature, we say that there are mechanisms for fixing domain walls. And only the application of intense external loads can destroy these mechanisms, and rotate vectors of spontaneous polarization. It is required to construct a mathematical model that allowed us to determine the distribution of the integral characteristic of vectors of the spontaneous polarization, or in other words it is necessary to find the vector of the polarization and tensor of the strain for representative volume.

2.2. Stages of constructing a model

Assuming that the domain walls are not fixed and domains do not affect each other, we derived the expressions for the residual polarization and deformation from the action of Weiss electric field and mechanical stresses. This case is called an ideal. Let \( E_{\text{ef}} = E + \alpha P_0 \) - electric Weiss field. Quite similarly, we introduce \( \sigma_{\text{ef}} = \sigma + \beta \varepsilon_0 \) - Weiss mechanical field, assuming that the residual
deformation of a representative volume affects on mechanical stress on the micro level. The energy of electric dipole in Weiss fields can be represented as
\[ E = \varepsilon \sigma + P E \]
Therefore, according to the findings presented in [22], we write the expression for the limiting polarization and strain in a representative volume in the following form
\[
\varepsilon = \frac{\int e^{-U/\kappa T} \varepsilon_s dS}{\int e^{-U/\kappa T} dS}, \quad P = \frac{\int e^{-U/\kappa T} p_s dS}{\int e^{-U/\kappa T} dS},
\]
(1)
Then we calculate the energy required to rotate the domain, which consists of electrical and mechanical parts
\[
\Delta U = \int_{\Omega} \left[ k_1 \frac{dP_0}{dE} + k_2 \frac{d\varepsilon_0}{d\sigma} \right] d\Omega
\]
(2)
Next we calculate the work of the electric field and mechanical stresses and for a cyclic process established that part of the work that is dissipated by the hysteresis loss
\[
\Delta A = -\int_{\Omega} \varepsilon_0 d\sigma_0 d\Omega - \int_{\Omega} P_0 dE_0 d\Omega
\]
(3)
The image and likeness, we can specify the work needed to turn the domain in the ideal case
\[
\Delta A_\infty = -\int_{\Omega} \varepsilon_\infty d\sigma_\infty d\Omega - \int_{\Omega} P_\infty dE_0 d\Omega
\]
(4)
Using (1) - (3), we can set the energy balance, according to which the actual loss in the polarization process consist of losses in the ideal case, plus the energy costs required for breaking the locking mechanisms of the domain walls
\[
\Delta A = \Delta A_\infty + \Delta U
\]
(5)
Substituting (2) - (4) in (5) and taking into account that these relations must be satisfied for any representative volume and any increment of the effective field, we obtain two desired equations to determine the residual strain and polarization
\[
-\varepsilon_0 = -\varepsilon_\infty + k_2 \frac{d\varepsilon_0}{d\sigma_\infty}, \quad -P_0 = -P_\infty + k_1 \frac{dP_0}{dE_0}
\]
(6)
It is easy to see that the obtained relations are equations in differentials, which can also be regarded as a finite-difference equation. Included in these relations the ultimate strain and polarization in 1D case according to (1) can be written as
\[
\varepsilon_\infty = \varepsilon_s \int_{-1}^{1} e^{\eta(t)} \left( \frac{3}{2} s^2 - 1/2 \right) dt, \quad P_\infty = p_s \int_{-1}^{1} e^{\eta(t)} dt, \quad \varphi(t) = \frac{\varepsilon_s / p_s (\sigma_\infty (3/2 s^2 - 1/2) + E_\infty t)}{a}
\]
(7)
2.3. Method of solution
Examine, for example, the case when only the mechanical stress affects on a ceramic sample. Then \( E_\infty = 0 \). The solution of first equation of (6) is proposed to search for using the method of successive approximations. To do this, we rewrite it in finite differences. Let the increment of the mechanical stresses is \( \Delta \sigma \) and organize an iterative process as follows
\[
(\Delta \varepsilon_0)_m = \frac{\varepsilon_\infty \left( (\varepsilon_0)_m \right) - (\varepsilon_0)_m}{k_2} \Delta \sigma + \beta(\Delta \varepsilon_0)_m + (\varepsilon_0)_m + (\Delta \varepsilon_0)_m
\]
(8)
Once $\Delta \varepsilon_0$ will be found, we give a new increment of the mechanical stresses $\Delta \sigma$ and the process repeats as long as the stresses does not reach a predetermined value.

Similarly, consider the case where the electric field is set and mechanical stress is absent. Let the electric field got the increment $\Delta E$. Here also, as in the previous case, it is possible to organize the following iterative process:

$$
(\Delta P_0)_m = \frac{P_x[(P_0)_m] - (P_0)_m}{k_1} \Delta E + \alpha (\Delta P_0)_{m-1} \quad (P_0)_m = (P_0)_{m-1} + (\Delta P_0)_m
$$

(9)

3. Results and Discussion

3.1. Deformations hysteresis loop

A constructed mathematical model based on the iterative process (7) allows us to build residual deformations for any kind of uniaxial tensile or compressive stress. However, the experimental detachable deformation hysteresis loops include both residual and elastic deformation. Therefore, in our model we must to introduce the elastic component of deformation. We require that the elastic strain is been proportional to the applied stress i.e. $\Delta \varepsilon = c_2 \sigma$. Then the full strain will be $\varepsilon = \varepsilon_0 + \varepsilon_0$. As a result, we have 5 parameters: $\varepsilon_0$, $\varepsilon_0$, $\beta$, $k_2$, $c_2$, which we can dispose so that the calculated according to our model and experimental hysteresis loops are coincided. The most difficult is the problem of describing small hysteresis loops when mechanical stresses increase and then decrease, and on a new step their value is becoming more and more. As experimental data, we took the results of [24], where there are small deformations and dielectric hysteresis loops for one type of ceramic. Figure 1 shows the experimental deformations hysteresis loops taken from this work. After that, the nearest auxiliary problem was the task of choosing the model parameters that the calculated curves are coincided with the curves of the experiment. It should be noted that the problem of the choice of parameters is not trivial and represents itself the ill-posed coefficients problem. Fortunately for our operator can say that incorrectness practically does not show itself. This means that in our model manages to select parameters by direct sorting of valid values. I.e. in this case, there is the following trend: small changes of the parameters give little change the hysteresis curves. Following the numerical experiments, we obtained the following values of the model parameters $\varepsilon_0 = 0.01$, $\beta = 5 \cdot 10^7$ $m^2/N$, $a_2 = 2.5 \cdot 10^7$ $N/m^2$, $k_2 = 2.5 \cdot 10^8$ $N/m^2$, $c_2 = 0.75 \cdot 10^{-14}$ $m^2/N$.
Small deformations hysteresis loop, calculated with help of our model, one can see in Figure 2. Comparing Figures 1 and 2, we can say that these curves coincide not only qualitatively but also quantitatively in some areas.

3.2. Dielectric hysteresis loop

Quite similarly we can construct dielectric hysteresis using relations (8). Here also we must introduce a component of the reversible polarization, which, together with the residual, give us a full polarization of the sample. This model also contains five parameters. For compliance of the experimental and calculated curves it is necessary to select these parameters in a certain way. For example, we take as experimental results the results of studies published in [25] which presented in Figure 3. Numerical experiments have shown that if the parameters of the model got values

\[ P_s = 0.43 \text{C/m}^2, \alpha = 7.3 \cdot 10^6 \text{Vm/C}, a_i = 9.8 \cdot 10^6 \text{V/m}, k_1 = 4.9 \cdot 10^5 \text{V/m}, c_1 = 1.4 \cdot 10^{-8} \text{C/Vm} \]

we can get small dielectric hysteresis loops as shown in Figure 4.

Comparing Figures 3 and 4, we can say that these curves coincide not only qualitatively but also quantitatively in some areas.

The results show that the model allows us to construct depending strain and dielectric hysteresis only qualitatively. Quantitative coincidence occurs only in certain areas of curves. Most good agreement is obtained for large values of the load. For loads slightly different from zero you can see only qualitative coincidence. In addition, portions of the curve, where the increasing load is replaced by decreasing the experimental curves are rounded but calculated curves have sharp peaks. A next discrepancy is observed in the areas of increasing and decreasing load. For the experimental curves this parts of curves are very close to each other, but for the calculated curves markedly significant discrepancy. Explanation for all of this is may be next fact: when we create a model, we used statistics of Boltzmann distribution, which allow us to construct the density distribution of the domains in the fields of applied loads. In all probability, the exponential distribution is not quite accurately describes the real picture of the behavior of domains in intense fields. This is especially noticeable for loads of small value. In this connection the question of constructing mathematical models to describe the polarization processes remains relevant.

4. Conclusion

A one-dimensional model to describe the nonreversible processes of polarization and depolarization of polycrystalline ferroelectric materials was created. Model is based on statistical distribution law in the domain of intense electric and mechanical fields. Using the energy balance between the work of the external loads, actual losses and energy to rotate the domains we built energy relation, from which the
equations for the residual strain and polarization in the differentials are obtained. Considering these equations as equations in finite differences we proposed a method of successive approximations to solve them. Introducing in model a reversible strain and polarization components as a quantity proportional to the corresponding loads, we built hysteretic relationship between mechanical stress and strain and between the polarization and electric field. If external loads vary at a circular law of maximum intensity, then we obtain large deformations and dielectric hysteresis loops. If the load increases and decreases incrementally with gradually increasing amplitude, one can obtain the small hysteresis loop. Shown that the model has a set of parameters and we can dispose them of at their discretion. Numerical experiments were performed and the results were compared with experimental data published earlier. It is shown that the optimal choice of parameters can give the coincidence between the small hysteresis curves not only qualitative but also quantitative. The deficiencies of models also were noted and were pointed out on their probable causes. The constructed model can be used in finite element analysis ferroelectric polarization processes.

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References
[1] McMeeking R M, Landis C M 2002 Int. J. Eng. Sci. 40 1553
[2] Huber J E, Fleck N A 2001 J. Mech. Phys. Solid 49 785
[3] Landis C M 2002 J. Mech. Phys. Solid 50 127
[4] Huber J E, Fleck N A, Landis C M, McMeeking R M 1999 J. Mech. Phys. Solid 47 1663
[5] Marutake M J 1956 Phys. Soc. Japan 11 807
[6] Eshelby J D 1957 Proc. R. Soc. Lond. A 241 376
[7] Aleshin V I, Luchaninov A G 2002 Ferroelectrics 266 111
[8] Dunn V L 1995 J. Appl. Phys. 78 1533
[9] Dunn M L, Wiencke H A 1997 Int. J. Solid. Struct. 34 3571
[10] Cheng J, Wang B, Du S 1999 Acta Mech. 138 163
[11] Hwang S C, Lynch C S, McMeeking R M 1995 Acta Metall. Mater. 43 2073
[12] Chen X, Fang D N, Hwang K C 1997 Acta Mater. 45 3181
[13] Huo Y, Jiang Q 1997 Smart Mater. Struct. 6 441
[14] Fotinich Y, Carman G P 2000 J. Appl. Phys. 88 6715
[15] Zhang Z K, Fang D N, Soh A K 2006 Mech. Mater. 38 25
[16] Sun C T, Achuthan A 2001 Domain switching criteria for piezoelectric materials. Proc. SPIE, Smart Structures and Materials
[17] Suo Z, Kuo C M, Barnett D, Willis J R 1992 J. Mech. Phys. Solids. 40 739
[18] Jiles D C, Atherton D L 1986 J. Magn. Magn. Mater. 61 48
[19] Smith R C, Hom C L 1999 SPIE Conference on Mathematics and Control in Smart Structures. SPIE: Newport Beach, CA. March 1-4, 3667 150
[20] Smith R C, Ounaies Z 2000 J. Intellig. Mater. Syst. Struct. 11 62
[21] Hom C L, Shankar N 1998 IEEE Trans. Ultrason., Ferroelectr. Frequency Control. 45 (2) 409
[22] Belokon A V, Skaliukh A S 2010 Mathematical modeling of irreversible processes of polarization FIZMATLIT, Moscow (Russian addition).
[23] Skaliukh A S 2012 in: I A Parinov (Ed.), Piezoelectric materials and devices, Nova science publishers, Inc. New York
[24] Salten M, Schneider G A, Knoblauch V, McMeeking R M 2005 Int. J. Solid. Struct. 42 3953