European Congress on Computaional Methods in Applied Sciences and Engineering
ECCOMAS 2004
P.Neittaanmäki, T.Rossi, K.Majava, and O.Pironneau (eds.)
R.Owen and M.Mikkola (assoc. eds.)
Jyväskylä, 24-28 July 2004

MODELING AND SIMULATION OF PIEZOCERAMIC MATERIALS USING MICROMECHANICAL APPROACH

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Key words: Micromechanical modeling, Piezoelectricity, Probability, Electromechanical loading.

Abstract. *Piezoceramics are becoming more and more common among smart materials, applications of which are nowadays encouraged by scientist and engineers. Although they are so excellent in usage, they have some drawbacks in high performance applications. Domain switching (ferroelastic and ferroelectric) is the main reason for nonlinearities of piezoceramic materials. External excessive electromechanical loads (mechanical stress and electric field) are driving forces for domain switching. In this study, nonlinear behavior of tetragonal perovskite type piezoceramic materials is theoretically simulated using a micromechanical model. This model consists of many different grains which form the bulk material. Uni-axial loading is applied in the simulations. The calculations which are based on a linear constitutive model and nonlinear domain switching model are performed at each grain. All grains are assumed to be statistically random oriented in the beginning. The behavior of piezoelectric materials under constant compressive stress which is applied in the direction of the electric field is also investigated. The response of the bulk ceramics is predicted by averaging the response of individual grains. It is assumed that a crystalline switches if the reduction in potential energy of the polycrystal exceeds a critical energy barrier per unit volume of the material. Due to intergranular effects domain switching may occur in reality even for those grains, for which the critical energy level is not reached. This effect is modeled by introducing a probability for switching as a function of the actual energy level related to the critical energy level. With the help of this probability functions, it is possible to model the nonlinearity even in a small electromechanical loading range. The effect of different domain switchings (90° or 180° domain switching for tetragonal perovskite structure) due to energy differences, different probability functions, different statistical random generators and material parameters are analysed. The results of simulations are compared with experimental data from literature.*

1. INTRODUCTION

Recently, piezoelectric materials became a highly interesting part of smart materials due to their various commercial engineering applications such as active vibration and noise control, precision positioning & cutting, common rail system with piezo control in automobile industry, microactuators and sensors and ferroelectric random access memories (FRAM). Piezoceramic materials are divided into two groups that are called soft and the hard according to its substitutional elements. The main difference between the soft piezoelectric material and the hard one is that the former has a higher piezoelectric constant than the latter. $BaTiO_3$, PZT and PLZT are well known examples of piezoceramic materials which have a perovskite type tetragonal microstructure (Fig.1). An extensive basic knowledge of piezoceramic and ferroelectric materials are given by Jaffe et al. in [1].

Ferroelectric materials have a spontaneous polarization which can change its direction when loaded with high electromechanical loading when the temperature is under the Curie temperature. The Curie temperature is the border below which the material changes its geometric structure (cubic element to tetragonal element) (Fig.1). At lower loading conditions, piezoceramic materials behave linearly. The main nonlinearity in piezoelectric materials is the result of domain switching which is the change of the direction of the spontaneous polarization in the microstructure under high electromechanical loadings. Various types of experiments are performed by scientists in order to understand the non-linear behavior of ferroelectric and piezoelectric materials. In these experiments, the piezoceramic sample is usually subjected to a cyclic electric and mechanical loading [2-6]. During the experiments, the magnitude of the electric field or mechanical stress is slowly increased and decreased at a low rate (0.01-1Hz) for observing hysteresis loops. These experiments can give appropriate information about the effect of the composition of some PLZT materials, concentrations of elements, phase changes from tetragonal to rhombohedral or rhombohedral to tetragonal structures and types (soft and hard) of the piezoceramic material. The influence of loading rate on the electric displacement and strain is observed by Zhou et. al [7] using different loading frequencies in experiments.

There are two different approaches that have been frequently used in order to simulate the behavior of piezoelectric and ferroelectric materials. Micromechanical modeling is the one for which the simulation based on a physical approach of the material. Phenomenological modeling is the other approach that mostly relies on a thermodynamical basis for simulation. In micromechanical models, usually a bulk piezoelectric material is assumed which is composed of a large number of randomly oriented grains or microstructures. A uniform macroscopic load is applied to the bulk material. The linear constitutive equations and the domain switching under the effect of the applied electric field and stress are the main criteria for calculations within each element. An energy equation is used for the threshold of switching. Simple analytical averaging is implemented to get the macroscopic response of the bulk material to the external applied load. A similar micromechanical model is proposed by Hwang et. al. [2]. They used a Preisach hysteresis model for describing electric displacement and strain simulation of each grain of the PLZT (Lead lanthanum zirconate titane). The results are given and compared with

experiments which were performed for a PLZT 8/65/35. Chen et. al. [22] propose microscopic constitutive relations of single crystal ferroelectric materials for predicting the properties of polycrystalline ferroelectrics based on monocrystalline ferroelectrics by adding an internal variable in the theoretical constitutive equations and domain volume fractions. The differences between monocrystal and polycrystal materials result in some mismatching in the simulation.

A model for polarization switching in order to predict hysteresis and butterfly curves under electromechanical loading in polycrystalline tetragonal ferroelectric ceramics is presented by Hwang et. al. [8]. They added some additional material parameters to the domain switching energy barrier equation for getting best matching to experimental results that were measured before. Different electrical and mechanical loads were used in the simulations. The limitation in approximating the gradual process of switching is the main drawback of the model. Lu et. al. [6] used an orientation distribution function which is the basic difference from another approach while simulating the perovskite type of ferroelectric ceramics under various electromechanical loadings. Another model proposed by Chen and Lynch [9] give more attention to some other phenomena such as effects of tetragonal and rhombohedral phases, different energy levels associated with domain switching criteria of these phases, intergranular effects between grains and phase changes from rhombohedral to tetragonal, as well as tetragonal to rhombohedral. They also apply a linear saturation model in their calculations for matching simulations with experimental curves.

Some different phenomenological (macroscopic) constitutive models which are usually based on a thermodynamical framework are developed to predict the behavior of ferroelectric materials. Because of having less time requirement for calculations, phenomenological models are more advantageous than micromechanical models. A simple phenomenological model for ferroelectric materials is presented by Kamlah et. al. [10, 11]. They simulated basic dielectric hysteresis, butterfly hysteresis, ferroelectric hysteresis, mechanical depolarization and polarization induced piezoelectric curves. Certain internal variables are introduced in the model for representing the history dependence of the material. Kamlah et. al [12, 13] presented an interesting phenomenological model which contains macroscopically history dependent nonlinearities, dielectric, butterfly hysteresis due to switching, thermo-electromechanical coupling properties and time dependent effects in the simulations. Another phenomenological constitutive model for ferroelectric switching under multi-axial mechanical and electrical loadings is proposed by McMeeking et. al. [14].

In some applications of piezoelectric materials especially piezoelectric actuators are usually not only operated under electrical loading but also under mechanical stress. High mechanical loads can also effect the nonlinear characteristic of piezoceramic materials. Lynch [5] observed hysteresis and butterfly curves under various constant compressive stresses in the experiments. Lu et. al. [6] propose a model to simulate the behavior under fixed compressive stresses and compare the results with their experiments. Though they get good results, the smoothness of the curves is not predicted well. There are some other attempts to model the mechanical stress effect in the simulations. In these models different energy equations are used to simulate hysteresis and butterfly curves under fixed mechanical stress [9, 17, 20].

This paper presents a new micromechanical modeling of ferroelectric and piezoelectric materials. Using probability functions in the energy equations for domain switching is the sig-

nificant difference of the model [21]. By using this probability function, intergranular effects can be assumed to be taken into account phenomenologically. The results of the simulations are shown in curves for the dielectric displacement versus electric field hysteresis curve under uniformly loaded cyclic electric field with and without constant pressure.

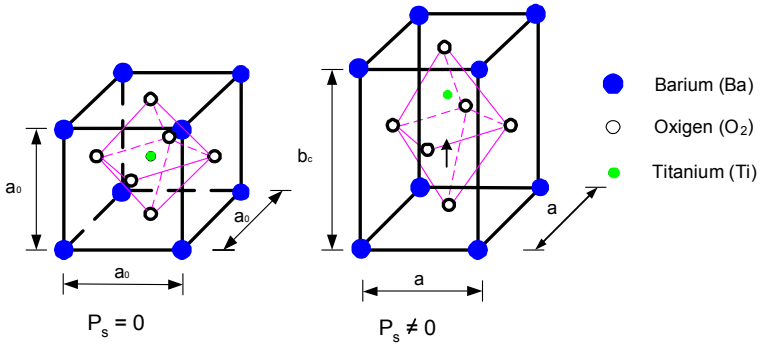

Figure 1: Lattice structure of cubic and tetragonal elements ($BaTiO_3$)

2. MICROMECHANICAL MODEL DESCRIPTION AND SIMULATION RESULTS

Microstructures of ferroelectric materials exist as simple cubic elements if the operating temperature is above the Curie temperature. This phase is called the paraelectric phase. Orientations of atoms in the cubic elements give a zero net dipole moment in the paraelectric phase. When the temperature is reduced below the Curie temperature, the lattice structure of ferroelectric materials changes from cubic to a tetragonal or a rhombohedral structure. The new phase occurring by this transition is called the ferroelectric phase. Figure 1 shows the cubic and tetragonal microstructures. Because of the changed orientations of atoms in the tetragonal lattice structure, the net dipole moment is different from zero in contrary to the cubic element. The spontaneous polarization which can change the direction when the material undergoes electrical or mechanical loadings is the net dipole moment in the microstructure due to these new orientations of atoms in the ferreolectric phase. In addition to this, movements of atoms during phase transitions give a net strain which is called the spontaneous strain. In the microstructure, the spontaneous strain (S_s) that is coming from the transition from the cubic phase to the tetragonal phase depends on the lengths of the unit cells of both phases (b_c and a_0) and may be calculated by

$$S_s = \frac{(b_c - a_0)}{a_0} \tag{1}$$

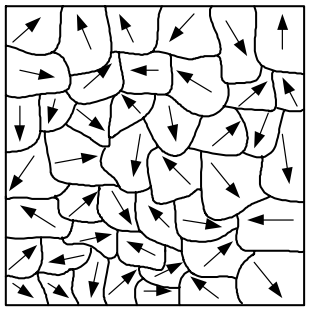

Figure 2a : Randomly oriented grains (unpoled case)

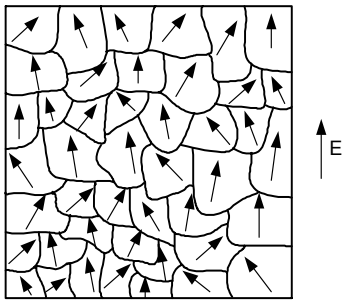

Figure 2b : Fully poled grains in electric field direction

Figure 2a shows randomly oriented grains of a typical bulk ferroelectric material for an unloaded case. Each grain has a different spontaneous polarization direction. The randomness of the direction of the spontaneous polarization for the unpoled case gives approximately zero polarization and strain of a bulk ferroelectric material. If the material is put under a high electric field like shown in figure 2b, all randomly oriented spontaneous polarization vectors are aligned in the loading direction. The electric displacement and strain values can be calculated for each grain of the microstructure both using constitutive equations and the spontaneous polarization values. The macroscopic polarization and the corresponding strain of the bulk piezoceramic material are calculated by adding analytically the average of the spontaneous values to the results obtained by the linear constitutive equations

$$P = P_s + d\,\sigma + \varepsilon\, E \tag{2}$$

$$S = S_s + s\,\sigma + d\, E \tag{3}$$

where P is the polarization, S is the strain, E is the electric field, σ is the mechanical stress, P_s is the spontaneous polarization, S_s is the spontaneous strain, ε is the dielectric constant, d is the piezoelectric constant and s is the elastic constant.

The behavior of piezoceramic materials can not be described by linear constitutive equations when they are under high electromechanical fields. Domain switching in microstructures is the leading factor for the nonlinearity. There are six possible orientations for the polarization in the microstructure for a perovskite type tetragonal lattice element. For this reason, only two types of domain switching is possible which are 90° and 180° switching, see figure 3. Their names come from the angle of rotation which the central atom makes during switching with respect to its previous direction.

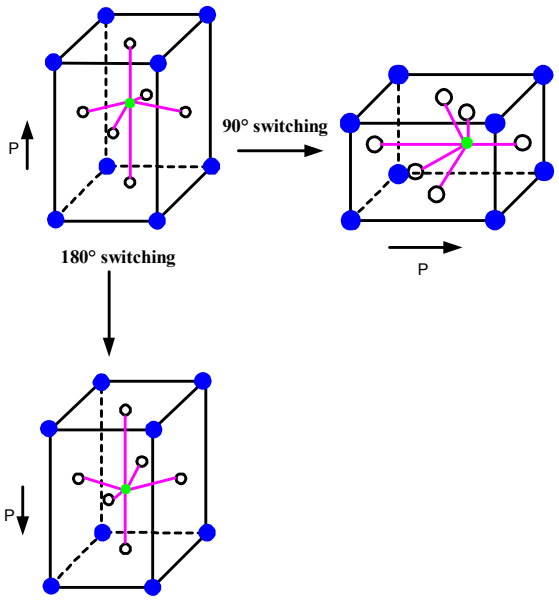

Figure 3: 90° and the 180° switching in the lattice structure of perovskite type tetragonal element

In our micromechanical model, we consider a bulk piezoceramic material having 1000 elements each of which is assumed to show the characteristics of an individual grain shown in figure 4. Each grain has random orientation in properties of polarization and strain. Randomness is given to every element by using Euler angles between 0 and 2π. In the simulation local coordinate systems for all grains and a global coordinate system are defined for transforming the values obtained by the calculations in the local coordinate system to global coordinates and vice versa. The relation between global coordinates $(x_g,y_g,z_g)^T$ and local coordinates $(x_l,y_l,z_l)^T$ is given by

$$(x_g,y_g,z_g)^T=R(\alpha,\beta,\gamma)\ (x_l,y_l,z_l)^T \tag{4}$$

The transformation matrix $R(\alpha,\beta,\gamma)$ can be calculated from Euler angles (α,β,γ) by a multiplication of subsequent three rotation matrices due to three rotation angles, see figure 5.

$$Rz(\alpha).Ry(\beta).Rz(\gamma) = \begin{bmatrix} \cos\alpha\cos\beta\cos\gamma-\sin\alpha\sin\gamma & \sin\alpha\cos\beta\cos\gamma+\cos\alpha\sin\gamma & -\cos\gamma\sin\beta \\ -\cos\alpha\cos\beta\sin\gamma-\sin\alpha\cos\gamma & -\sin\alpha\cos\beta\sin\gamma+\cos\alpha\cos\gamma & \sin\gamma\sin\beta \\ -\cos\alpha\sin\beta & \sin\alpha\sin\beta & \cos\beta \end{bmatrix} \tag{5}$$

Although it is clear that equally distributed Euler-angles do not give a uniform distribution of the polarization direction, equally distributed Euler-angles are more commonly used especially for micromechanical models in literature.

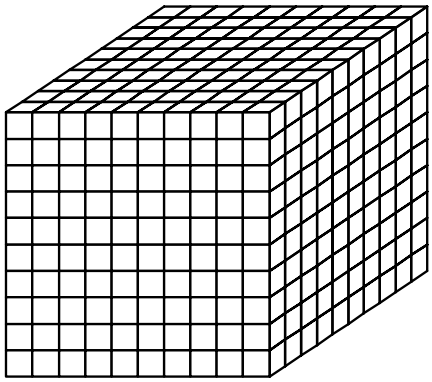

Figure 4: Micromechanical model in form of 1000 cubic elements

The external macroscopic load applied in the model is assumed to be equal to the load on each element of the bulk piezoceramic material. For getting a better fit of the simulations to the experimental data, some material parameters such as spontaneous polarization, spontaneous strain, piezoelectric and dielectric constants are chosen from literature. In order to simplify the calculations, dielectric and elastic constants are assumed to be isotropic and the same for all grains. On the other hand, the piezoelectric constant is assumed to be transversely isotropic. During the calculation, the value for the dielectric permittivity (ε) is taken to be $6.0\ 10^{-8}$ F/m, which is a typical value for PZT materials. The spontaneous polarization value is chosen as 0.3 C/m^2. At each grain, in addition to the piezoelectric constitutive equation, the

nonlinear switching model is also implemented. The domain switching at each grain is determined by using the electromechanical energy criteria

$$E\Delta P+\sigma\Delta S \geq 2P_0E_c. \qquad (6)$$

where E and σ are electric field and mechanical stress, E_c is the coercitive electric field, P_0 is the critical value of the spontaneous polarization that depends on the material, ΔP and ΔS are the polarization and strain change during domain switching correspondingly. For the first part of the simulation, the mechanical stress is taken constant and zero. Thereby, on the left side of the inequality, the stress part becomes zero. According to this criteria, domain switching occurs if the change of energy is higher than a certain critical level. The same energy levels are assumed for 90° and 180° domain switching during the simulations. Therefore, the hysteresis curves have sharp edges as can be seen in figure 6. This is not observed in experiments due to intergranular effects. Thus a probability for switching is introduced which allows a switching even for energy levels which are below the critical energy level. The probability is a function of the actual energy related to the critical energy level. For energy levels above the critical value the probability is one which means that the polarization direction is given. With this model nonlinearities may be explained even in small electromechanical loading range. This is the new and important part of the research.

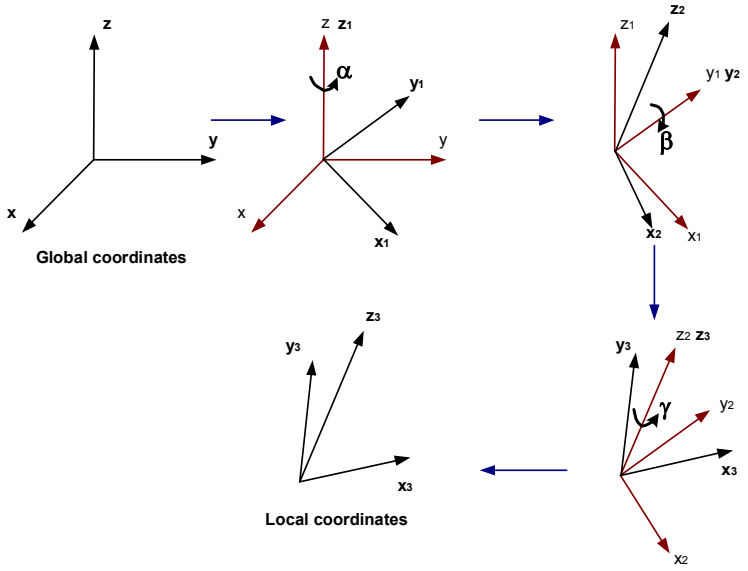

Figure 5: Coordinate transformation by using Euler angles ZYZ convention

The probability function is given by equation (7). The probability function for domain switching is related to some degree (n) of the polynomial function of the energy of microstructure (U) and critical energy (U_c)

$$p(E, \sigma) = (U/U_c)^n \qquad (7)$$

In this equation p is the probability for switching which is varying between 0 to 1 according to the applied electric field. In the simulations, the value for n is chosen as three, four and five. The difference between simulated curves with probability criteria and without probability is significant, see figures 6 to 8.

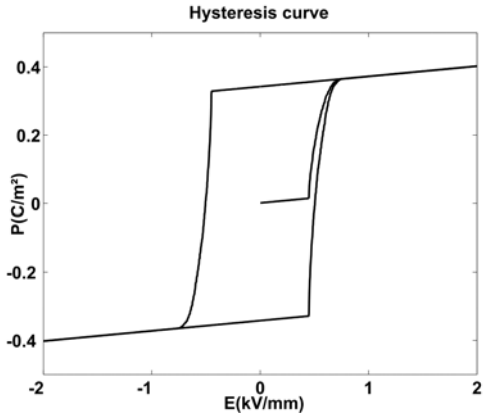

Figure 6: Hysteresis curve without probability function.

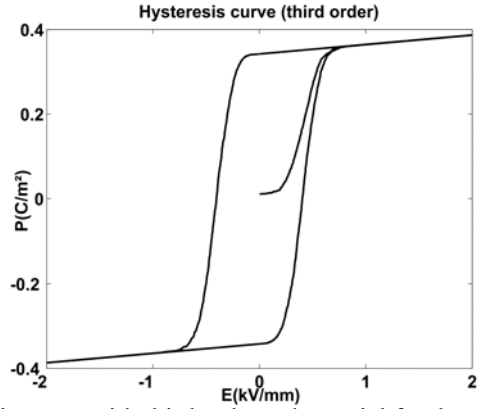

Figure 7: Hysteresis curve with third order polynomial for the probability function.

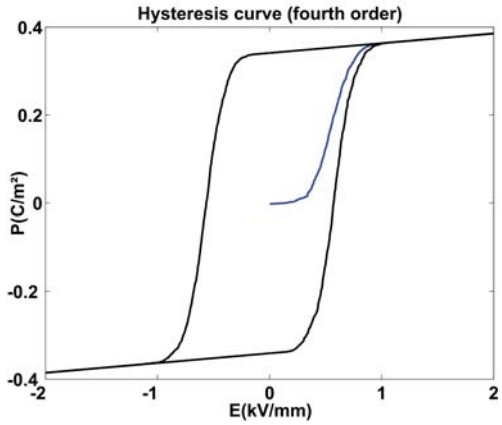

Figure 8: Hysteresis curve with fourth order polynomial for the probability function.

A comparison of the simulation with experimental data which was measured by Lu et. al. [6] is given in figure 9. The simulated results obtained with a probability function fit better with experimental results than those curves which were simulated without using a probability criteria. During the simulation a cyclic, uni-axial electric field is applied with a maximum ab-

solute value of 2 kV/mm. For the comparison, the spontaneous polarization is assumed to be 0.2 (C/m^2) which is nearly the same as in experimental data. The starting point for the first cycle is at zero electric field for the unpoled ceramic. Euler transformations and averaging of all grains are used to get the macroscopic response of the bulk material to the applied electro-mechanical loads.

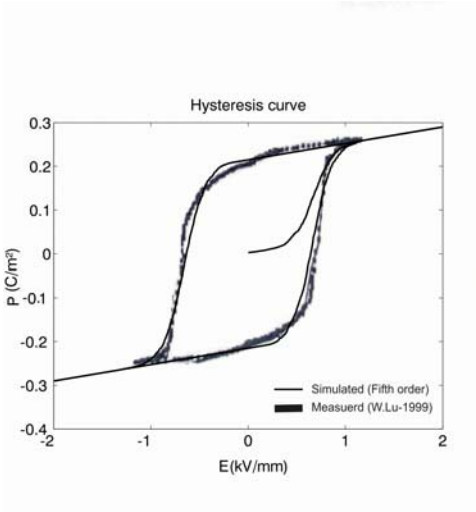

Figure 9: Comparison between simulation (fifth order polynomial for the probability function) and measured data.

In the following, the behavior of piezoelectric materials is investigated if a constant compressive stress is applied in direction of the electric field. The experimental results which are given in literature show that if a fixed high compressive stress is applied during cyclic electrical loading of piezoceramic materials, there are three basic differences which occur in the electric displacement versus electric field curve. First, the coercitive field which is the threshold for domain switching decreases. The reason is the reduction of energy required for switching for the first 90° switching due to the stress distribution inside the material. On the other hand, this stress distribution in the microstructure of the piezoelectric material increases the energy required for the second 90° switching (Fig. 10). Therefore, the total bandwidth of switching is enlarged for the case of compressive stress compared to the no stress case. The bandwidth of switching is linearly increasing with applied compressive stress. We take one unit cell of piezoelectric material after applying a fixed compressive stress and a cyclic electric field. The stresses ($\sigma_1>\sigma_3>\sigma_2$) and electric fields ($E_1>E_3>E_2$) are assumed like in Fig. 10 in local coordinates (X_1, X_2, X_3) of the lattice structure. For some microstructures these stresses can be tensile because of local directions. So that the second term of the left side of the energy equation in (6) can become positive for the first 90° switching of piezoelectric material. Then three switching relations occur as in (8) for lower electric field levels. When the critical energy is achieved for the second direction of the lattice structure, 90° switching occurs. After switching, with increasing electric field, the inequality energy relation for the second 90° switching is expected to pass the energy threshold. Now, a higher electric field is re-

quired to overcome the negative effect of the compressive stress in the energy equation in direction 1. Therefore, stress in direction one discourages the second 90° switching.

$$E_2\Delta P+\sigma_2\Delta S \geq E_3\Delta P+\sigma_3\Delta S \geq E_1\Delta P+\sigma_1\Delta S \quad (8)$$

A second major effect of a constant compressive load is the reduction of remanent and saturation polarization values when the stress is applied. This phenomena is explained by the negative effect of the stress term in the constitutive equation (2). As seen in the constitutive equation (2), the magnitude of remanent and saturation polarization is decreasing linearly when increasing the applied mechanical stress.

The change in the piezoelectric coefficient is the third important difference when mechanical stress is applied. According to the experiments, there is no linear correlation between the applied stress and the piezoelectric coefficient [5].

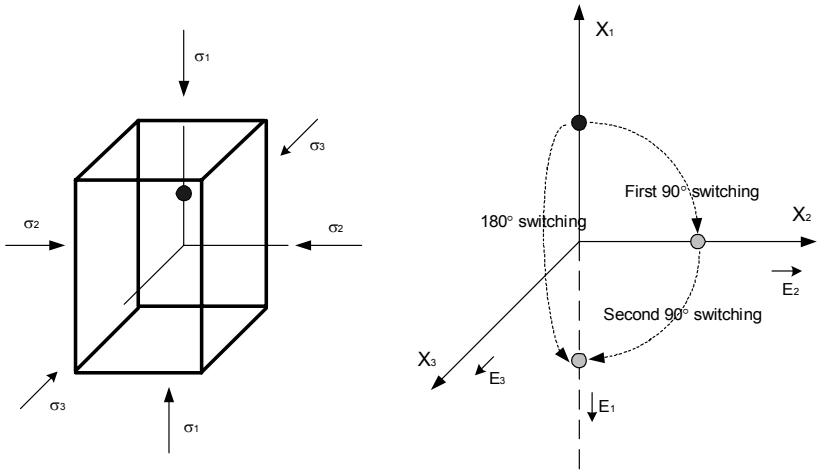

Figure 10: Lattice structure under constant compressive stress with electric field and switching possiblities

In our model, we have also tried to simulate the electric displacement versus electric field curve under fixed mechanical stress. In the simulations the constant stress values are chosen as 20 and 40 Mpa. The piezoelectric constant is assumed to behave transversly isotropic as mentioned before. The value of the piezoelectric constant in loading direction is taken to be $3.0\ 10^{-9}$ (m/V).

Figures 11-13 show the hysteresis curves with and without probability functions for piezoceramic materials that have a tetragonal perovskite type microstructure loaded under 20 Mpa constant compressive stress. Figure 11 demonstrates a hysteresis curve which was simulated without taking into consideration the probability function. It can be seen easily that, there are two steps near both the loading and the unloading coercitive electric field levels. This is explained by the different energy levels of types of domain switchings due to the effect of compressive stress. The first step during switching period is dominated by first 90° switchings which are encouraged by compressive stress. In a real experimental case, such a stepwise increasing is not so apparent. Therefore, the authors have used probability functions in the simulations in order to have a better matching with the experimental curve. Figures 12-13 have been obtained by using third and fourth order polynomials for the probability functions. Especially figure 12 matches the corresponding experimental curve. An increased switching period, and decreased overall polarization values are well aprroximated in these simulations.

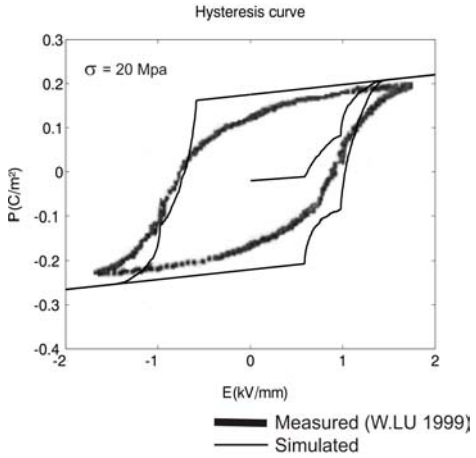

Figure 11: Hysteresis curve without probability function.

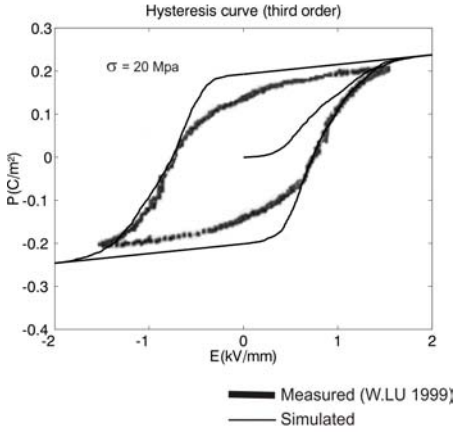

Figure 12: Hysteresis curve with third order polynomial for the probability function.

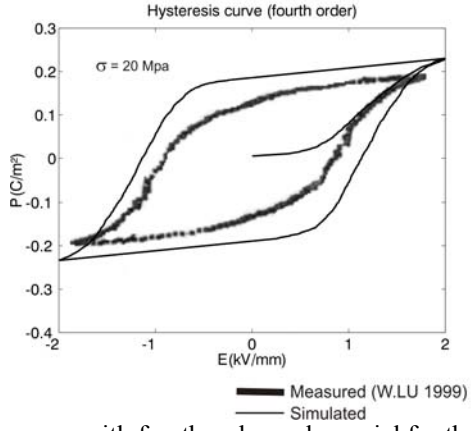

Figure 13: Hysteresis curve with fourth order polynomial for the probability function.

A higher mechanical loading case is also simulated and compared with experimantal data. Figure 14 shows the hysteresis curve with 40 Mpa constant compressive stress using a third order polynomial for the probability function. As the constant compressive stress is increased, the period of switching is enlarged which is aproximated well in the simulation. However, the magnitude of change of overall electric displacement has been underestimated.

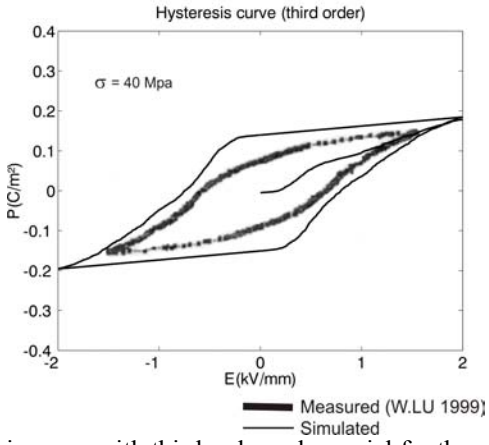

Figure 14: Hysteresis curve with third order polynomial for the probability function.

3 CONCLUSION

Properties of tetragonal perovskite type piezoceramic materials have been simulated by using a new micromechanical model under uni-axial high cyclic electric loading and fixed compressive stress. A piezoelectric linear constitutive model, and nonlinear domain switching with probability functions have been used in the model. The hysteresis curves, using probability functions do better match to the experimental hysteresis curves than simulation results in which probability functions have not been used. Piezoelectric materials under constant compressive stress have been deeply investigated in this paper. Hysteresis curves under different constant compressive stress values are also simulated. In the next step, the model will be implemented to get butterfly curves in which strain is plotted versus the electric field curve. In addition to this, intergranular effects between neighboring grains and time dependent properties of piezoelectric and ferroelectric materials will be also investigated in future.

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