A Mathematical Model for Polarization Dynamics in a BTS 15 Ceramics

Original research article

Abstract

In this paper, a ferroelectric structure consisting of barium stannate titanate piezoelectric ceramic is used for mathematical modeling. The study of the dynamics of the structure requires the poling process that makes the ceramic active. This may be explained by the time-dependent nonlinear polarization behavior. The model results in a nonlinear second order ordinary differential equation of one degree of freedom. Analytical exact solutions have been obtained for three regimes following the specific value of the damping parameter. Numerical simulations of these solutions by optimization with Matlab package showed excellent agreement in comparison with measurements and a mean square error of order of 10^{-5} is obtained.

Keywords: polarization; piezoelectricity; BTS ceramics; mathematical model. 2010 Mathematics Subject Classification: 53C25; 83C05; 57N16

1 Introduction

The knowledge of the following physical phenomena called piezoelectricity, pyroelectricity, and ferroelectricity goes back over a hundred years. In 1917, Just Hauy first observed qualitatively electrical phenomenon resulting from the action of a mechanical stress on some crystals. In 1880, theoretical and experimental study of this phenomena was undertaken by the brothers Pierre and Jacques Curie who are credited with the discovery of the direct piezoelectric effect [(5)]. The following year, the reverse piezoelectric effect was theoretically stated by Lipman, in the same year, Pierre and Jacques Curie checked successfully. This inverse effect is manifested by a mechanical deformation caused by application of electric field. Thus the first industrial applications were made for the detection of ultasonic waves during the second world war. Since then progress is constantly growing. Then, the first piezoelectrity

materials were discovered, the most known is barium titanate ceramics. The effects of the piezoelectric can only be observed on insulating body. In power applications, the ceramic running high stresses have characteristic phenomena of non-linear behavior. To understand the origin of nonlinearities, it must be remembered that the existence of a piezoelectric effect in ceramics is due to a non-linear polarization process. This is justified by the fact that the areas of the structure that were referred during the polarization process are under high mechanical or electrical stresses tend to resume their original direction, these shifts being irreversible and appearing a precise order. Upon application of an electric field to a ferroelectric material, the molecules or atoms of which it consists are deformed such that the centroids positive and negative charges that make up no longer coincident. In spite of its drawback of low Curie temperature, (BaTiO₃)-based pizoelectric materials can be considered as an excellent model system for exploring the physics of piezoelectric materials [(9)]. Barium titanate $(BaTiO_3)$ is the most common ferroelectric oxide in the perovskite ABO_3 structure, which is used as various electronic devices such as capacitors, thermistors, transducers and non volatil memories in semiconductor industries because of its dielectric and ferroelectric properties. The electric and dielectric properties can be modified [(33)] by doping with various isovalent cations on both A (Ba) and B(Ti) sites (see (33) and references therein). SnO_2 was doped into $(BaTiO_3)$ to form barium titanate stannate $(Ba_xTi_{1-x}Sn_xO_3)$, short for BTS. Study of the pyroelectric behavior of $(Ba_xTi_{1-x}Sn_xO_3)$ piezo-ceramics [(25)] showed the thermal square wave method single-frequency applied to samples with a tin gradient of $0.075 \le x \le 0.15$. As a result, because of the polarization dependence of the piezoelectric coefficient the technique used enables evaluation of the impact of ceramic fabrication on the expected piezoelectric response [(25)]. In this paper x = 0.15 is used which corresponds to $BaTi_{0.85}Sn_{0.15}O_3$ or BTS15. This is a binary solid solution system composed of ferroelectric barium titanate and non-ferroelectric barium stannate. Both of them are of perovskite structures [(33)]. In [(31)] advancements a made and are favorable for pyroelectric device applications. The authors studied porous $BaSn_{0.05}Ti_{0.95}O_3$ (BTS 5) ceramics prepared by sintering compacts consisting of BTS and Poly as pore former. An important result has been revealed is dielectric constant decreases and loss increases with porosity. The main applications resulting from ferroelectric materials, are: the realization of high dielectric permittivity capacitors, the realization of piezoelectric sensors and ultrasonic generators using polarized poly crystalline samples, the provision dielectric amplifiers and modulators, the realization of thermoelectric energy converters, the realization of DRAM (Dynamic Random Access Memories) and NVFeRAM (Non Volatile Random Access Memories ferroelectric) [(1)].

One example of these materials is ceramics based on barium titanate stannate solid solution, which is widely used in piezoelectric actuators and microsensors, tunable microwave devices, and thermistors. Practical applications require piezoelectric ceramics with not only uniform but also gradient chemical composition, ensuring a nonuniform distribution of dielectric, piezoelectric, and other physical properties in ceramic samples [(22)].

A major research activity for half a century was dedicated to perovskite materials having either type of structure properties, ferroelectric and piezoelectric or electrostrictive properties in a view of applications in electromechanical or ultrasonic devices [(22)]. In this domain, the majority of the work relates to lead-based perovskite. But in recent years, intense research has been conducted to study lead-free materials. In this paper, the propagation of electromagnetic wave in a structure consisting in a nonlinear ferroelectric multilayer structures is studied. These materials are characterized by the existence of spontaneous polarization and a demonstrated reorienting of the polarization (17). A ferroelectric material presents in the absence of external electric field, a polarization which can be reversed by the application of a electric field that exceeds a critical value called coercive field. Beyond this value all the material will be intrinsic dipoles preferentially oriented in the direction polarization. Several types of inorganic and organic materials have this property [(3)]. The modern definition of ferroelectric polarization can be found in some recent text books [(28)]. In ferroelectric materials, polarization manifests itself as a hysterisis loop. In the presence of an electric field the polarization of dipoles in a ferroelectric material is non-linear and causes a phenomenon known as electrical

hysteresis. A theoretical representation of polarization mechanism of the dipoles in a ferroelectric material is depicted in [(14)].

Based on the work done by Pientschke et al in [(23)], the present work is devoted to the study of the behavior of the time-dependent polarization in ferroelectric structure. In [(23)], optical poling techniques were used on the barium titanate doped with 15% Sn. Let's recall foremost that in such materials, the macroscopic polarization vanishes because of the randomly oriented domains. A poling process is necessary in order to impress the piezoelectric properties.

The propagation of a wave through a material produces changes in the spatial and temporal distribution of electrical charges as the electrons and atoms interact with the electromagnetic fields of the wave. The main effects of the forces exerted by the field on the charged particles is displacement of the valence electrons from their normal orbits. This perturbation creates electric dipoles whose macroscopic manifestation is the polarization [(30)]. Polarization is the central quantity that is used to explain the physics and behavior of dielectric material [(13)]. One can consider that the binding "core-electrons" are described by the model of the oscillator. Then the mathematical model developed in this work results in a nonlinear second order ordinary differential equation of one degree of freedom. It is well known that nonlinear problems having explicit exact solutions in terms of elementary standard functions are very limited in physical and engineering fields [(26)]. After a suitable change of variables, the differential equation obtained was solved analytically. Three different solutions were obtained, corresponding to three different regimes according to the values of the damping parameter. These regimes are the critically damped nonlinear response, the under-damped nonlinear oscillation, and the over-damped nonlinear response.

In this paper, we report the results from modeling the polarization dynamics for barium titanate stannate ceramics. The paper is organized as follows. The Section 1 is devoted to the mathematical model for the time-dependent polarization of the ferroelectric structure studied. Section 2 presents the analytical solutions of the nonlinear differential equation. Section 3 deals with the numerical approach and results and the last Section is the concluded part.

2 Mathematical model for the time-dependent polarization of the BTS 15 structure

Before we present the mathematical model, let's explain how a ferroelectric ceramic material is polarized. Ferroelectric ceramics are formed of grains and bond joints. Each grain is divided into areas whose dipole moments are oriented randomly in the absence of electric field. By applying an external electric field, a given temperature, the domains are oriented parallel to this field for a metallized ceramic plate on its sides. The ceramic polarizes and is made piezoelectric. When the material is heated, there is a temperature, so-called Curie temperature, which corresponds to a structural phase change of which is to consequently remove polarization. The material then passes the ferroelectric state to paraelectric state and this polarization remains stable in a given domain of temperatures [(20)].

The starting point of this study is the Newton's law of motion, for a particle of masse m and charge q:

$$m\frac{d^2u(t)}{dt^2} = F_{ext} - F_{int}$$
(2.1)

where u(t) is the displacement of the particle of charge q from equilibrium. F_{ext} is the applied exciting force and F_{int} is assumed to be [(26)]

$$F_{int} = k\phi(u) + b\frac{d\phi u(t)}{dt}$$

Assuming $\phi(u) = u$ we have:

$$m\frac{d^2\phi(u)}{dt^2} + b\frac{d\phi u(t)}{dt} + k\phi(u) = F_{ext}$$
(2.2)

If we set $F_{ext} = 0$, we have the well-known differential equation of motion for the displacement u(t) of the particle of charge q and mass m, from the equilibrium.

$$\frac{d^2u(t)}{dt^2} + \lambda \frac{du(t)}{dt} + \omega_0 u(t) = 0$$
(2.3)

with $\lambda = \frac{b}{m}$, and $\omega_0^2 = \frac{k}{m}$; ω_0 is the natural resonance frequency of the mass-spring system and λ is the damping factor. Eq.(2.3) is used to describe the damped linear oscillator of a single degree of freedom. This equation represents the dynamics of the motion of one dimensional viscoelastic system [(26)]. The studied material having viscoelastic behavior which is the case for most materials. Indeed the material can conserve energy and release energy after deformation. On the other hand, it also has the ability to dissipate energy. Such oscillatory viscoelastic dynamical system is intrinsically characterized at least by its stiffness, damping and inertia nonlinearities so the mathematical description of dynamics of the viscoelastic system must include these nonlinearities. The Eq. (2.3) should them be replaced, as mentioned in [(26)]; [(19)] by the generalized quadratically disspative Liénard type equation governing the dynamics of a viscoelastic system [(26)]; [(19)].

$$\phi'(u)\ddot{u} + \phi''(u)\dot{u}^2 + \lambda\phi'(u)\dot{u} + \omega^2\phi(u) = \frac{1}{m}F(t, u, \dot{u}, \ddot{u})$$
(2.4)

 $F(t, u, \dot{u}, \ddot{u})$ being the external exciting function and u(t) is the displacement variable. For mathematical reasons, we use $\phi(u) = u^l$, where l is the nonzero stiffness nonlinearity controlling parameter. Using the relationship between the force and the electric field and introducing the expression of $\phi(u)$, we have:

$$\dot{u} + (l-1)\frac{\dot{u}^2}{u} + 2\mu\dot{u} + \frac{w_o^2}{l}u = g(u)\frac{F(t)}{m}$$
(2.5)

with $g(u) = \frac{u^{1-l}}{l}$ and

$$F(t) = E(t)L_o^{l-1}$$
 (2.6)

A simple substitution in Eq. 2.5 gives:

$$\ddot{P} + (l-1)\frac{\dot{P}^2}{P} + 2\mu\dot{P} + \frac{w_0^2}{l}P = \frac{1}{l}E(t)P_s^{l-1}Ne_0P^{1-l};$$
(2.7)

where $e_0 = \frac{q}{m}$ is the charge per unit mass and $P_s = L_0 N q$ is the spontaneous polarization. We will restrict ourselves for this study to a constant field $E(t) = E_0$. Let's solve the following equation obtained:

$$\ddot{P} + (l-1)\frac{\dot{P}^2}{P} + 2\mu\dot{P} + \frac{w_0^2}{l}P = \frac{E_0}{l}Ne_0P_s^{l-1}P^{l-1}$$
(2.8)

3 Exact analytical solutions

By making a variable change $P = y^{\frac{1}{l}}$, the previous differential equation may be cast in the form:

$$\ddot{y} + 2\mu \dot{y} + w_0^2 y = N E_0 e_0 P_s^{l-1} \tag{3.1}$$

Let's solve foremost the homogeneous equation associated to the equation 3.1

$$\ddot{y} + 2\mu \dot{y} + w_0^2 y = 0 \tag{3.2}$$

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the solutions of this equation depends on the roots of the caracteristic equation:

$$r^2 + 2\mu r + 1 = 0 \tag{3.3}$$

The reduced discriminant Δ' of this equation gives us:

$$\Delta' = \mu^2 - \omega_0^2 \tag{3.4}$$

Three cases will be distinguished: $\mu > \omega_0$, $\mu = \omega_0$, and $\mu < \omega_0$ representing three regimes called respectively over damped, critically damped, under damped regimes. Let's then examine the solutions related to each of these regimes.

3.1 Exact analytical solution for critically damped nonlinear response: $\mu = \omega_0$

This case corresponds to the following solution:

$$y(t) = \left(E_1 + E_2 t\right) e^{-ut} + \alpha/\omega_0^2;$$
(3.5)

where E_1 and E_2 are arbitrary real constants; and the constant $\alpha = NE_0e_0P_s^{l-1}$. Using initial conditions: $P(0) = P_0$ and $\dot{P}(0) = q_0$ the response of the system is the time dependent polarization, given by:

$$P(t) = \left[\left[P_0^l - \frac{\alpha}{w_0^2} + \left(P_0^l (\frac{lq_0}{P_0} + \mu) - \frac{\mu\alpha}{w_0^2} \right) t \right] e^{-\mu t} + \frac{\alpha}{w_0^2} \right]^{\frac{1}{t}}$$
(3.6)

3.2 Exact analytical solution for under damped nonlinear response $\mu < \omega_0$

The solutions are given by:

$$y(t) = e^{-\mu t} \left(D_1 \cos w_d t + D_2 \sin w_d t \right) + \alpha / \omega_0^2$$
(3.7)

where $\alpha = NE_0e_0P_s^{l-1}$. Use made of initial conditions $P(0) = P_0$ and $\dot{P}(0) = q_0$ gives:

$$P(t) = \left[\frac{\alpha}{w_0^2} + \left[\left(P_0^l - \frac{\alpha}{w_0^2}\right)\cos(w_d t) + \frac{1}{w_d}\left[P_0^l\left(\frac{lq_0}{P_0} + \mu\right) - \frac{\alpha\mu}{w_0^2}\right]\sin(w_d^t)\right]e^{-\mu t}\right]^{\frac{1}{t}}$$
(3.8)

with

$$\omega_d = \sqrt{\omega_0^2 - \mu^2}$$

3.3 Exact analytical solution for over damped nonlinear response : $\mu > \omega_0$

The solutions are given by:

$$y(t) = H_1 e^{-(u+w_d)t} + H_2 e^{-(u-w_d)t} + \alpha/\omega_0^2$$
(3.9)

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where H_1 and H_2 are arbitrary real constants. The final solution by making use of the initial conditions: $P(0) = P_0$ and $\dot{P}(0) = q_0$ gives:

$$P(t) = \left[\frac{\alpha}{w_0^2} + e^{-\mu t} \left[\left(P_0^l - \frac{\alpha}{\omega_0^2} - \frac{1}{2\omega_d} \right) \left[lq_0 P_0^{l-1} + \left(\mu + \omega_d \right) \left(P_0^l - \frac{\alpha}{\omega_0^2} \right) \right] \right] e^{-\omega_d t} + \frac{1}{2\omega_d} \left[lq_0 P_0^{l-1} + \left(\mu + \omega_d \right) \left(P_0^l - \frac{\alpha}{\omega_0^2} \right) \right] e^{\omega_d t} \right]^{\frac{1}{l}}$$
(3.10)

where the constants α and ω_d are given by: $\alpha=E_0Ne_0P_s^{l-1}$ and $\omega_d=\sqrt{\mu^2-\omega_0^2}$

4 Numerical approach and results

Let's recall that changes in polarization by the electric field appear in the form of a hysteresis cycle where we denote the presence of a coercive field, a polarization and a spontaneous polarization. One also studies the spontaneous polarization with respect to the temperature in ferroelectric materials where the essential parameter introduced is the Curie temperature; and the phase transitions have been highlighted. In our work, we analytically compute time-dependent polarization. The measurements presented in this paper have been performed by [(23)].

We used the method of optimization with the least squares technique in Matlab package. The problem is formulated as follows: what are the values of parameters that reproduce at best the experimental data? These parameters obtained following the specific value of the damping parameter are presented for each regime. From the outset, it is important to emphasize the shared characteristics of three systems and the characteristics that are particular to each regime. For three regimes, it is observed that for the time $\tau > 30s$, the experimental and the theoretical curves are coincident with the experimental curve slightly above. We observe there a plateau indicating an asymptotic value of the polarization: $P = 0.071C.m^{-2}$ of the BTS15 layer. When τ is varying between 0 and 30s, the variation of the polarization is very abrupt at the beginning and after decreasing to the value $\tau = 30s$.

The mean square error calculated numerically is equal to 10^{-5} , which allows us to say with these parameters, the built model is excellent, hence validating the model.

The various parameters optimized for the different regimes are as follows:

4.1 Aperiodic regime: $\mu > \omega_0$

Consider the case of the aperiodic regime see Figure 1. The parameters are:

N = 0.08179: number of particles per unit volume (density), $\mu = 1.049$: damping coefficient, l = 1.032: parameter hardening, $P_0 = 0.001185$: initial polarization, $q_0 = 0.1693$: initial polarization speed, $e_0 = 0.07377$: electric charge per unit mass, $\omega_0 = 0.3638$: natural frequency, $P_s = 0.2223$: saturation polarization E = 1.500: echelon excitation force, the mean square error obtained is: $mse = 1.1737 \times 10^{-5}$. The obtained results are shown in the Fig. 1. From this figure, we can see that the experimental plot is in full agreement with the theoretical plot.



Figure 1: Experiment and theoretical plots showing dependence of polarization with time in ferroelectric structure for strong damping. Constant electric field is applied at time t=0.

4.2 Critical regime: $\mu = \omega_0$

Consider the case of the aperiodic regime see Figure 2. The parameters are:

N = 0.3385: number of particles per unit volume (density), $\mu = 4.0714$: damping coefficient, l = 1.6627: parameter hardening, $P_0 = 0.00118$: initial polarization, $q_0 = 0.7255$: initial polarization speed, $e_0 = 0.1285$: electric charge per unit mass, natural frequency: $\omega_0 = 4.0714$, E = 1.500: echelon excitation force, $P_s = 5.6613$: saturation polarization, the mean square error obtained is: $mse = 1.4402 \times 10^{-5}$.

4.3 Pseudo-periodic regime: $\mu < \omega_0$

Consider the case where the aperiodic regime see Figure 3. The parameters are:

N = 1.448: number of particles per unit volume (density), $\mu = 2.448$: damping coefficient, l = 1/3: parameter hardening, $P_0 = 0.00118$: initial polarization, $q_0 = 0.3131$: initial polarization speed, $e_0 = 1.489$: electric charge per unit mass, natural frequency $\omega_0 = 2.483$, $P_s = 1.427$: saturation



Figure 2: Experimental and theoretical plots showing dependence of polarization with time in ferroelectric structure for critical damping. Constant electric field is applied at time t=0



polarization E = 1.500: echelon excitation force, the mean square error obtained is: $mse = 1.2622 \times 10^{-5}$.

Figure 3: Experiment and theoretical plots showing dependence of polarization with time in ferroelectric structure for for low damping. Constant electric field is applied at time t= 0.

We use nonlinear least square method as approach of optimization. The method of least squares is used to estimate the values of the unknown parameters. The biggest advantage of nonlinear least squares regression over many other techniques is the broad range of functions that can be fit. Although many scientific and engineering processes can be described well using linear models, or other relatively simple types of models, there are many other processes that are inherently nonlinear. One common advantage is efficient use of data. Nonlinear regression can produce good estimates of the unknown parameters in the model with relatively small data sets. Another advantage that nonlinear least squares shares with linear least squares is a fairly well-developed theory for computing confidence, prediction and calibration intervals to answer scientific and engineering questions. In most cases the probabilistic interpretation of the intervals produced by nonlinear regression are only approximately correct, but these intervals still work very well in practice. Even if all the optimization methods have advantages and disadvantages, it is useful to highlight here the robust advantages of the presented method.

5 CONCLUSIONS

A model for the simulation of the polarization dynamics in a ferroelectric material, the barium stannate titanate was presented. A detailed analytical solutions of the nonlinear differential equation was performed for different regimes of damping parameter. The simulations compared to measurements reveal that the model is satisfactory for optimum values of the obtained parameters. That is comforting because the mean square error obtained is of order of 10^{-5} . It is not worth denoting that the effects of electric field on the dynamical behavior of ferroelectric materials is of a capital interest and especially for large amplitude of externally applied field. That leads to strong non-linearity in ferroelectric materials. In future work we will consider a variable electric field and determine the response of the material.

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