TRANSIENT ELECTROMECHANICAL RESPONSES OF FERROELECTRIC CERAMICS TO IMPULSIVE ELECTRICAL FIELDS

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Abstract—When a partially or thoroughly poled disc of a ferroelectric ceramic is subjected to a suddenly applied step electric field, it vibrates symmetrically. The vibrations of the disc depend on the mapitude of the applied field, the state of polarization of the disc and the direction of the field with respect to the direction of polarization. Here, we consider the boundary-initial value problem characterizing the transient responses of the disc and determine. in particular, its boundary mechanical displacement history, Qualitative comparisons are made with available experimental results.

I. INTRODUCTION

In this paper we examine the dynamic electromechanical responses of ferroelectric ceramics to impulsive electric fields. When a partially or thoroughly poled disc of a ferroelectric ceramic is subjected to a suddenly applied step electric field, it vibrates symmetrically. The vibrations of the disc depend on the magnitude of the applied field, the state of polarization of the ceramic disc and the direction of the field with respect to the direction of polarization. Here, we are particularly interested in the transient responses of the disc.

The constitutive relations for describing the dynamic electromechanical responses of ferroelectric ceramics are quite complex.[†] In addition to the usual dependences of the stress on the strain and the electric displacement on the electric field, the stress and the electric displacement also depend on the responses of the dipole moment and the consequences of domain switching. The responses of the dipole moment due to strain and electric field are both instantaneous and transient, whereas domain switching due to strain and electric field seems to be a purely transient process.

The problem we have in mind corresponds physically to a fairly thin disc which is large in lateral extent. Only the center circular regions of the surfaces of the disc are electroded. The disc is subjected to a suddenly applied step electric field. The mechanical displacement history of either of its surfaces and the electric displacement history may be determined experimentally. In the formulation of this problem we presume that the physical processes are one dimensional corresponding to conditions of uniaxial strain. We further presume that the strains which arise in the disc due to electromechanical interactions are not of sufficient magnitude to induce domain switching so that we may have domain switching due to the applied field alone depending on its magnitude and its direction.

2. THE GOVERNING EQUATIONS

The constitutive relations for the stress and the electric displacement relevant to our present problem are special cases of those given by Chen[l]. Basically, he presumes that the following conditions hold:

(i) In the virgin or thermally depoled state the mechanical properties of the ceramic are elastic and it behaves like a normal dielectric exhibiting no piezoelectric effect. In such a state the material of the ceramic is regarded to be isotropic and homogeneous.

(ii) The material of the ceramic is transversely isotropic with respect to the poling direction, say, the X_3 -direction and its mechanical, piezoelectric and dielectric properties depend on the number of aligned dipoles in this direction.

(iii) The transient responses of domain switching and of the dipole moment give rise to rate effects which affect the mechanical, piezoelectric and dielectric properties of the ceramic.

tSee Ref. (I].

In our present situation the axial direction of the disc is the X_3 -direction. Since the physical processes associated with the problem are one dimensional corresponding to conditions of uniaxial strain, we see that the only non-zero component of the mechanical displacement is u_3 and

$$
u_3 = u_3(X_3, t). \tag{2.1}
$$

Let N_3 denote the number of aligned dipoles in the X_3 -direction. It is given by

$$
N_3 = N_3{}^p + N_3{}^r,\tag{2.2}
$$

where N_3^p is the number of aligned dipoles of permanently switchable domains, and N_3^r is the number of aligned dipoles of non-permanently switchable domains.

In this paper, it is more convenient to use the electrically neutral state as the reference state and we need the normal component of the stress T and the component of the electric displacement **D** in the X_3 -direction which in the standard condensed notation are given by[†]

$$
T_3 = (C_{11} + a_{333}N_3)S_3 + (b_{3i} + d_{3i3}N_3)\mu_{S_i} - e_{333}N_3E_3 - g_{333}N_3\mu_{E_3} + h_{33}^{\parallel} (N_3{}^p + \frac{1}{3}N_3{}^r) + \frac{2}{3}h_{33}^{\perp}N_3{}^r,
$$
\n(2.3)

$$
D_3 = e_{333}N_3S_3 + j_{3i3}N_3\mu_{S_i} + (\epsilon + \epsilon_{333}N_3)E_3 + (\delta + \delta_{333}N_3)\mu_{E_3} + k_{33}N_3, \tag{2.4}
$$

where $S_3 = \partial u_3/\partial X_3$ is the normal component of the strain S and E_3 is the non-zero component of the electric field E in the X_3 -direction and μ_s and μ_E are, respectively, the transient response of the dipole moment due to strain and electric field. The constitutive relations (2.3) and (2.4) characterize the departures of the stress and the electric displacement from the electrically neutral state. In addition to the constitutive relations, we have the rate laws

$$
b_{3i}\mu_{S_i} + \alpha b_{3i}\mu_{S_i} = \beta_{b33}(N_3)S_3,\tag{2.5}
$$

$$
d_{3i3}\mu_{S_i} + \alpha d_{3i3}\mu_{S_i} = \beta_{d333}(N_3)S_3, \tag{2.6}
$$

$$
j_{3i3}\mu_{s_i} + \alpha j_{3i3}\mu_{s_i} = \beta_{j333}(N_3)S_3,
$$
 (2.7)

$$
\dot{\mu}_{\mathbf{E}_3} + \alpha \mu_{\mathbf{E}_3} = \beta_{\mu_{\mathbf{E}_3}}(N_3) E_3, \tag{2.8}
$$

where α is a constant and β_{b33} , β_{d333} , $\beta_{\mu E3}$ and β_{j33} are, in general, functions of N_3 ; and for the number of aligned dipoles, we have the following:

(i) If sgn $N_3^P = \text{sgn } \beta_E^P / \alpha_E^P$ and $|N_3^P| \geq |\beta_E^P / \alpha_E^P|$, then

$$
\dot{N}_3{}^p = 0,\tag{2.9}
$$

otherwise

$$
\dot{N}_3{}^p + \alpha_{\mathbf{E}}{}^p(E_3)N_3{}^p = \beta_{\mathbf{E}}{}^p(E_3),\tag{2.10}
$$

where $\alpha_{\mathbf{E}}^p$ and $\beta_{\mathbf{E}}^p$ are known functions;

(ii) If sgn $N_3' =$ sgn $\beta_E'/(r\alpha_E^P)$ and $|N_3'| \geq |\beta_E'|(r\alpha_E^P)|$, then

$$
\dot{N}_3{}' + \alpha_r N_3{}' = 0,\tag{2.11}
$$

where α_r is a constant, otherwise

$$
\dot{N}_3' + r\alpha_E^{\ \rho}(E_3)N_3' = \beta_E'(E_3),\tag{2.12}
$$

where r is a constant and $\beta_{\rm E}$ is a known function.

tSee [\,2).

In the constitutive relations (2.3) and (2.4), C_{11} and ϵ are the instantaneous elastic constant and the instantaneous dielectric constant of the ceramic in the electrically neutral state. a_{333} gives the transient change to C_{11} due to domain switching. $b_{3i}\mu_{S_i}$ together with the rate law (2.5) give the transient changes to $C_{11} + a_{333}N_3$ due to the transient response of the dipole moment, whereas d_{3i} _{j_{4}} together with the rate law (2.6) give the transient change to a_{333} due to the transient responses of the dipole moment and domain switching. e_{333} is the transient piezoelectric constant due to domain switching, whereas g_{333} and $j_{3i3} \mu_S$, together with the rate laws (2.7) and (2.8) give the transient changes to e_{333} due to the transient responses of the dipole moment and domain switching. In addition, we require that

$$
g_{333}\beta_{\mu_{\mathbf{E}_3}} = \beta_{133}.\tag{2.13}
$$

This condition ensures the compatibility of the piezoelectric coupling terms. h_{33}^1 and h_{33}^2 give the additional stresses due to domain switching. δ together with the rate law (2.8) give the transient change to ϵ due to the transient response of the dipole moment. ϵ_{333} is the transient change to ϵ due to domain switching, whereas δ_{333} together with the rate law (2.8) give the transient changes to ϵ_{333} due to the transient responses of the dipole moment and domain switching. Finally, k_{33} gives the additional electric displacement due to domain switching.

The field equations appropriate to our problem are balance of linear momentum

$$
\frac{\partial T_3}{\partial X_3} = \rho \ddot{u}_3,\tag{2.14}
$$

where ρ is the mass density of the ceramic and Gauss' law for a charge free body

$$
\frac{\partial D_3}{\partial X_3} = 0. \tag{2.15}
$$

Formula (2.15) implies that D_3 is independent of X_3 so that

$$
D_3 = D_3(t). \t\t(2.16)
$$

3. FORMULATION OF THE PROBLEM

To begin with, let us define a poled state via the relations

$$
T_3 = 0, S_3 = S_R, E_3 = 0, D_3 = D_R, N_3 = N_3^P = N_R. \tag{3.1}
$$

A poled state is an equilibrium state and is attained after the removal of the switching field. Clearly, innumerable poled states may exist. A thoroughly poled state is that for which $|N_{\rm R}|$ is maximum. There are two such states and the associated numbers of aligned dipoles may be normalized, viz.

$$
N_R = \pm 1. \tag{3.2}
$$

For our present purposes, it is sufficient to regard β_{133} , β_{4333} , β_{1333} and $\beta_{\mu_{13}}$ of the rate laws (2.5) - (2.8) as constants. It follows directly from the constitutive relations (2.3) and (2.4), and the conditions (3.1) that

$$
S_R = \frac{-h_{33}^2 N_R}{C_{11} + a_{333} N_R + \beta_{b33}/\alpha + \beta_{d333} N_R/\alpha},
$$
(3.3)

$$
D_R = e_{333} N_R S_R + \beta_{\beta 33} / \alpha N_R S_R + k_{33} N_R. \tag{3.4}
$$

Substituting these results into (2.3) and (2.4), we have

$$
T_3 = (C_{11} + a_{333}N_3)\bar{S}_3 + \gamma_{b3} + \gamma_{d33}N_3 - e_{333}N_3E_3 - g_{333}N_3\mu_{E_3} + (C_{11} + a_{333}N_3 + \beta_{b33}/\alpha + \beta_{d333}N_3/\alpha)S_R + h_{33}^{\parallel} (N_3^{\rho} + \frac{1}{3}N_3^{\prime}) + \frac{2}{3}h_{33}^{\perp}N_3^{\prime},
$$
\n(3.5)

$$
\bar{D}_3 = e_{333}N_3\bar{S}_3 + \gamma_{133}N_3 + (\epsilon + \epsilon_{333}N_3)E_3 + (\delta + \delta_{333}N_3)\mu_{E_3} \n+ \{(e_{333} + \beta_{1333}/\alpha)S_R + k_{33}\}(N_3 - N_R),
$$
\n(3.6)

where

$$
\bar{S} = S_3 - S_R, \bar{D}_3 = D_3 - D_R, \tag{3.7}
$$

$$
\gamma_{b3} = b_{3i}\mu_{S_i} - \beta_{b33}/\alpha S_R, \qquad (3.8)
$$

$$
\gamma_{433} = d_{3i3} \mu_{S_i} - \beta_{4333} / \alpha S_R, \tag{3.9}
$$

$$
\gamma_{133} = j_{313} \mu_{S_1} - \beta_{1333} / \alpha S_R. \tag{3.10}
$$

Further, the rate laws $(2.5)-(2.8)$ become

$$
\dot{\gamma}_{b3} + \alpha \gamma_{b3} = \beta_{b33} \bar{S}_3 \tag{3.11}
$$

$$
\dot{\gamma}_{d33} + \alpha \gamma_{d33} = \beta_{d333} S_3, \tag{3.12}
$$

$$
\dot{\gamma}_{j33} + \alpha \gamma_{j33} = \beta_{j333} S_3, \tag{3.13}
$$

$$
\dot{\mu}_{\mathbf{E}_3} + \alpha \mu_{\mathbf{E}_3} = \beta_{\mu_{\mathbf{E}_1}} E_3. \tag{3.14}
$$

Formulae (3.5) and (3.6) are more convenient for our present purposes in that they characterize the departures of the stress and the electric displacement from a poled state rather than the electrically neutral reference state.

With regard to the problem we have in mind, we presume that the measured electric displacement history $\bar{D}_3(t)$ is given and the solution of the problem entails the simultaneous solution of balance of linear momentum (2.14) with the constitutive relations (3.5) and (3.6) and of the rate laws (2.9) - (2.12) and (3.11) - (3.14) . The boundary-initial conditions of the problem are

$$
u_3(X_3, 0) = S_R X_3, \quad \dot{u}_3(X_3, 0) = 0,
$$

\n
$$
T_3(0, t) = 0, \quad u_3(l/2, t) = 0,
$$

\n
$$
N_3^P(0) = N_R, \quad N_3^r(0) = 0,
$$

\n
$$
\gamma_{b3}(0) = 0, \quad \gamma_{d33}(0) = 0,
$$

\n
$$
\gamma_{b3}(0) = 0, \quad \mu_{E_3}(0) = 0,
$$

\n(3.15)

where l is the thickness of the disc. In the following section we present specific numerical results of the solution of this problem.

4. SOLUTION OF THE PROBLEM

Here, we present the results of the solution of the problem. To this end, we consider the axial responses of a hot pressed ferroelectric ceramic PZT $65/35$ [†] disc with area 1.0×10^{-4} m²

tThe composition of PZT 65/35 is $Pb_{0.99}Nb_{0.02}(Zr_{0.65}Ti_{0.35})_{0.98}O_3$.

and thickness 4.29×10^{-3} m. The material properties with respect to the electrically neutral state are taken to bet

$$
C_{11} = 1.654 \times 10^{11} \text{ Pa},
$$

\n
$$
|a_{333}| = 8.559 \times 10^{10} \text{ Pa with sgn } a_{333} = - \text{ sgn } N_3,
$$

\n
$$
\beta_{b33}/\alpha = -7.875 \times 10^9 \text{ Pa},
$$

\n
$$
|\beta_{d333}/\alpha| = 0.4076 \times 10^{10} \text{ Pa with sgn } \beta_{d333} = \text{sgn } N_3,
$$

\n
$$
e_{333} = 17.09 \text{ C/m}^2,
$$

\n
$$
g_{333} = \beta_{\mu33}/\alpha = -2.773 \text{ C/m}^2,
$$

\n
$$
\beta_{\mu_{E_3}}/\alpha = 1.0,
$$

\n
$$
|h^{\parallel} = 5.657 \times 10^7 \text{ Pa with sgn } h^{\parallel} = - \text{sgn } (N_3^P + \frac{1}{3} N_3^P),
$$

\n
$$
|h^{\perp}| = 1.151 \times 10^8 \text{ Pa with sgn } h^{\perp} = - \text{sgn } N_3^P,
$$

\n
$$
\epsilon = 2.966 \times 10^{-9} \text{ F/m},
$$

\n
$$
|\epsilon_{333}| = 1.655 \times 10^{-9} \text{ F/m with sgn } \epsilon_{333} = - \text{sgn } N_3,
$$

\n
$$
\delta = 1.483 \times 10^{-9} \text{ F/m},
$$

\n
$$
|\delta_{333}| = 0.8273 \times 10^{-9} \text{ F/m with sgn } \delta_{333} = - \text{sgn } N_3,
$$

\n
$$
k_{33} = 0.3397 \text{ C/m}^2,
$$

\n
$$
\alpha = 1.25 \times 10^6 \text{ s}^{-1}.
$$

In Figs. 1 and 2 we exhibit, respectively, the properties of the functions $\alpha_{\mathbf{E}}^P$ and $\beta_{\mathbf{E}}^P/\alpha_{\mathbf{E}}^P$. These functions concern the permanently switchable domains; and, as for the non-permanently switchable domains, we let $\alpha_r = 0.2 \text{ s}^{-1}$, $r = 0.5$ and $\beta_E' / (r \alpha_E^p) = 0.14 \beta_E^p / \alpha_E^p$.

The numerical results of the solution of the problem are obtained by means of a one dimensional Lagrangian finite difference wave code, WONDY.[‡] This code is suitably modified to include the constitutive relations (3.5) and (3.6) , and the rate laws (2.9) – (2.12) and (3.11) – $(3.14).$

In Fig. 3, we exhibit the input electric displacement $\bar{D}_3(t)$ and the corresponding solution of

It should be mentioned that when the transient responses associated with $\mu_{\bar{z}_i}$ and $\mu_{\bar{z}_i}$ have reached equilibrium, these properties reduce to those given previously by Chen and Tucker[2] and that these properties are based on our best estimates of experimental results at this time.

‡Lawrence and Mason[3].

Fig. 3. Input electric displacement $\tilde{D}_3(t)$ and the solution of the boundary mechanical displacement $u_3(0, t)$.
In this case there is no domain switching.

Fig. 4. Experimentally measured boundary mechanical displacement in an experiment for which there is no domain switching.

Fig. 5. Input electric displacement $\bar{D}_3(t)$ and the solution of the boundary mechanical displacement $u_3(0, t)$. In this case there is domain switching.

Fig. 6. Experimentally measured boundary mechanical displacement in an experiment for which there is domain switching.

the boundary mechanical displacement $u_3(0, t)$. The initial condition of N_3^p is

$$
N_3{}^p(0) = N_R = 0.5.
$$

It suffices to point out that up to about 4.0×10^{-6} s the total number of aligned dipoles N_3 is approximately 0.5, i.e. the resulting electric field is not of sufficient magnitude to cause domain switching. Notice that the first maximum of the mechanical displacement is higher than the second maximum and that the minimum is different from zero. These features of the solution are due to a combination of different effects. Roughly speaking, that the minimum is different from zero is due to the mechanical relaxations arising from the rate laws (3.11) and (3.12) and that the first maximum is higher than the second maximum is due to the piezoelectric relaxations arising from the rate laws (3.13) and (3.14). The amplitudes of the boundary mechanical displacement depend on the total number of aligned dipoles N_3 in that it affects the values of piezoelectric coupling terms in the constitutive relations (3.5) and (3.6). The results obtained for different initial conditions of N_3 ^{*e*} are qualitatively the same but quantitatively different. The solution of the boundary mechanical displacement $u_3(0, t)$ is in qualitative agreement with that obtained experimentally some years ago for a specimen of atmospherically fired PZT 65/35. Unfortunately, there is not enough information for us to effect a quantitative comparison at this time. Nevertheless, the experimental result is shown graphically in Fig. 4.

In Fig. 5, we exhibit another input electric displacement $\bar{D}_3(t)$ and the solution of the boundary mechanical displacement $u_3(0, t)$. The initial condition of N_3^p is again 0.5. At about 4.0×10^{-6} s the total number of aligned dipoles N₃ is 0.56 so that domain switching has occurred. Now, the first maximum of the mechanical displacement is lower than the second maximum. This is because the increasing number of aligned dipoles results in increasing piezoelectric coupling, and this is sufficient to overcome the effects of piezoelectric relaxations alluded to earlier. In Fig. 6, we exhibit the experimental result of the boundary mechanical displacement of an atmospherically fired PZT 65/35 specimen for which there is domain switching. The qualitative comparison of the numerical and experimental results is extremely good.

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REFERENCES

- J. P. J. Chen. Three dimensional dynamic electromechanical constitutive relations for ferroelectric materials. *lnl.* J. *Solids Struclures* IS. 1059-1067 (1980).
- 2. P. J. Chen and T. J. Tucker, One dimensional polar mechanical and dielectric responses of the ferroelectric ceramic PZT 65/35 due to domain switching, *lnl.* J. *Engng Sci.* 19. 147-158 (I98\).
- 3. R. J. Lawrence and D. S. Mason, Sandia Laboratories Report SC-RR-710284 (1971).