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# Compressive creep of PZT ceramics: experiments and modelling

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#### Abstract

The compressive creep behaviour of a soft PZT has been studied. The effect of polarisation is measured and simulated by means of a viscoplastic-type model. The creep strain is linked to the amount of ferroelectric domains available to 90° ferroelastic switching. A temperature rise is found to reduce the anelastic strain but increase the creep strain itself.

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# 1. Introduction

In spite of response time as low as a few milliseconds, ferroelectric ceramics are known to be time-dependent. For example, the frequency of the applied electric field has a great influence on the poling cycles.<sup>1</sup> The response of a piezo-actuator to a step voltage is not constant and increases with time.<sup>2</sup> This rises some reliability problems in industrial applications. Therefore efforts have to be focused on experimental characterisation and modelling of the response of such materials to various solicitations, all the more as what really happens at the microscopic scale is not completely understood.

Furthermore, a mechanical stress is known to switch ferroelectric domains perpendicularly to its direction. Therefore some mechanically induced creep effects have been observed with perovskite-type bulk ceramics in compression and in tension.<sup>3–5</sup> This phenomenon disappears above the Curie temperature as the material is in a paraelectric phase.

In this paper, the influence of poling and temperature on the compressive creep behaviour of a soft PZT are highlighted experimentally. The measured responses of the specimens to complex solicitations are then simulated thanks to a viscoplastic-based model.

## 2. Experimental methodology

A commercial bulk soft ceramic is used in this study. PIC 255 is produced by PI Ceramic, Germany, and is indicated for actuators and high sensitive receivers applications. It is available in form of poled cylinders (15 mm height, 10 mm diameter). Some specimens have been depoled by heating treatment above Curie temperature.

Piezoelectric components may heat in use and temperatures may reach 80-100 °C. Thus experiments are performed at room-temperature (24 °C) and at 85 °C on non poled cylinders, which are maintained in a thermostatic box fixed on the testing machine.

A special device<sup>6</sup> has been designed to carry out compressive experiments. In order to insulate electrically the ceramic and to offer a better stress state in the sample, compliant layers made of glass/epoxy isotropic composite are used. Longitudinal strain is measured by means of an extensometric gauge glued in the central area of the cylinders. For poled ceramics, stress is parallel to polarisation and their electrodes are shortcircuited.

A compressive stress is applied during 2 h and released during 2 more hours. Loading speed has to be the highest possible (within the capabilities of the testing machine) and is taken to 20 MPa/s. Successive growing loadings (25, 50, 100 and 200 MPa) are applied to a same specimen. Experiments were repeated twice and offered very good repeatability.

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## 3. Experimental

# 3.1. Influence of polarisation

The anelastic strain is defined as the difference between the total strain and the elastic strain (the Young's modulus being taken to 110 GPa, which corresponds to a mechanically depoled material). The creep strain is the difference between the anelastic deformation at a given time and the initial anelastic deformation  $S_0^{\text{anel}}$ .

The experimental curves for poled or unpoled ceramics have a similar trend, although deformation levels are higher for the poled one (Fig. 1). Creep is of primary type and strain rate decreases with time. Andrade's power law can be used to quantify in a simple way this kind of behaviour. The creep strain  $S_c$  is defined according to the time t

$$S_{\rm c} = At^{1/q}$$

where A is a constant parameter and q depends on the stress level. The higher q is, the smaller the strain will be. Results are presented in Table 1. Another empirical law (logarithmic for instance) should have been chosen to characterise the evolution of the creep strain according to time. Nevertheless a drawback of this type of approach is that it does not take into account the history of the material. Therefore, only the coefficient measured during the first step of loading (25 MPa) can be compared to other values of the literature.

Independently from the polarisation state of the sample, creep is maximal for weak stresses, under 50 MPa in absolute value. Final  $S_c$  reaches then 0.065% for poled ceramic and only 0.030% for the non poled one. But

creep strain does not increase under higher stresses and becomes even negligible under 200 MPa. In addition, values of exponent q are very close for the two types of specimens.

It is possible to plot this strain according to the ratio of  $90^{\circ}$  switched domains, which was computed from ferroelastic strain levels measured thanks to other testings<sup>6</sup> (Fig. 2). Creep strain is high when ferroelastic activity is maximum, that is under 50 MPa. When the amount of domains available to the ferroelastic process saturates, creep stagnates.

Recovery strain can be interpreted as the amount of ferroelectric domains which switch back to their original position when stress is released (Fig. 3). For the unpoled ceramic, it increases according to the ratio of  $90^{\circ}$  switched domains. For the poled PZT, the recovery strain is apparently linked to the creep deformation.

## 3.2. Influence of temperature

According to the pyroelectric effect causing a shrinkage of the remnant polarisation value, PZT specimens contract with increasing temperature. By subtracting this thermal contraction strain (equal to -0.077%), anelastic strain levels for non poled ceramics are found

Table 1

Creep parameters according to the polarisation of the ceramic

Stress (MPa)	Non poled		Poled	
	$S_0^{\mathrm{anel}}$	q	$S_0^{\mathrm{anel}}$	q
25	0.126	95	0.146	31
50	0.248	59	0.325	34
100	0.445	166	0.601	193
200	0.595	588	0.657	557



Fig. 1. Influence of polarisation on the anelastic strain (room-temperature).



## Ratio of 90° switched domains

Fig. 3. Recovery strain according to ferroelastic activity (room-temperature).

to be less important at 85 °C than at room temperature (Fig. 4).  $S_0^{\text{anel}}$  values at high temperature are thus proportional with a ratio of 2/3 to those measured at 24 °C, whatever the applied stress may be (Table 2). Secondly, the exponent *q* increases clearly with temperature. This means that the reorganisation of the ferroelectric domains inside the ceramic grains is assisted by heating. The recovery strain is hardly affected by these conditions.

## 4. Modelling

#### 4.1. Description of the model

In order to simulate the material response, a phenomenological model<sup>6</sup> is presented in its one-dimen-

sional formulation. To describe the non-linear electromechanical behaviour, two independent variables are used: the irreversible strain  $S_{33}^{ir}$  and the irreversible polarisation  $P_3^{ir}$ . Physically, the macroscopic internal variables must be viewed as averages of the corresponding microscopic quantities. The total strain and polarisation can thus be split into:

$$S_{33} = S_{33}^{\text{rev}} + S_{33}^{\text{tr}}, \tag{1}$$

$$P_3 = P_3^{\rm rev} + P_3^{\rm ir}, (2)$$

where the reversible components are given by the classical linear equations of piezoelectricity. The irreversible strain has to be decomposed in two terms, a remnant



Fig. 4. Influence of temperature on the anelastic strain.

Table 2Creep parameters according to the temperature

Stress (MPa)	24 °C		85 °C	
	$S_0^{\text{anel}}$	q	$S_0^{\text{anel}}$	q
25	0.126	95	0.100	68
50	0.248	59	0.201	47
100	0.445	166	0.332	68
200	0.595	588	0.411	376

strain  $S_{33}^{\text{rem}}$  and a depolarisation (or ferroelastic) strain  $S_{33}^{\text{dp}}$  produced by mechanical loading.  $S_{33}^{\text{rem}}$  is directly connected to the remnant polarization by:

$$S_{33}^{\text{rem}} = S_{\max}^{\text{rem}} \frac{|P_3^{\text{ir}}|}{P_{\max}}.$$
(3)

 $P_{\text{max}}$  represents the maximum value of the polarization in a material subjected to a high electric field and  $S_{\text{max}}^{\text{rem}}$  is the maximum value of the strain reached for  $|P_3^{\text{ir}}| = P_{\text{max}}$ .

The evolution laws of the two internal variables are chosen to be time-dependent and are written according to their conjugated variable (the mechanical stress  $T_{33}$  for  $S_{33}$  and the electric field  $E_3$  for  $P_3$ ):

$$\dot{S}_{33}^{\rm dp} = \dot{S}_0^{\rm dp} \left\langle \frac{\left| T_{33} - \alpha_{33}^{\rm m} \right| - T_0}{N^{\rm m}} \right\rangle^{n^{\rm m}} \frac{T_{33} - \alpha_{33}^{\rm m}}{\left| T_{33} - \alpha_{33}^{\rm m} \right|} \tag{4a}$$

$$\dot{P}_{3}^{\rm ir} = \dot{P}_{0}^{\rm ir} \left\langle \frac{|E_{3} - \alpha_{3}^{\rm e}| - E_{0}}{N^{\rm e}} \right\rangle^{n^{\rm e}} \frac{E_{3} - \alpha_{3}^{\rm e}}{|E_{3} - \alpha_{3}^{\rm e}|}$$
(4b)

The superscripts m and e indicate respectively the parameters associated with the mechanical and the electrical variables. The internal variables  $\alpha_{33}^{\text{m}}$  and  $\alpha_{3}^{\text{e}}$  are non-linear kinematic hardening variables and  $E_0$  and  $T_0$  are constant parameters corresponding to the activation thresholds of the irreversible variables from which one may understand the use of the MacCauley brackets ( $\langle x \rangle = x$  if x > 0 and  $\langle x \rangle = 0$  otherwise).

Each of the non-linear kinematic variables is split up in two terms, a reversible component  $\alpha^{r}$  and an irreversible component  $\alpha^{ir}$ :

$$\alpha_{33}^{m} = \alpha_{33}^{rm} + \alpha_{33}^{irm} \text{ with } \alpha_{33}^{rm}(0) = \alpha_{33}^{irm}(0) = 0,$$
  

$$\alpha_{3}^{e} = \alpha_{3}^{re} + \alpha_{3}^{ire} \text{ with } \alpha_{3}^{re}(0) = \alpha_{3}^{ire}(0) = 0,$$
(5)

and their evolution laws are given by:

$$\dot{\alpha}_{33}^{rm} = b^{rm} \left( a^{rm} + \left| \alpha_{33}^{rm} \right| \right) \dot{S}_{33}^{dp}, \tag{6a}$$
$$\dot{\alpha}_{33}^{irm} = b^{irm} \left( a^{irm} \dot{S}_{33}^{dp} - \alpha_{33}^{irm} \left| \dot{S}_{33}^{dp} \right| \right),$$

$$\dot{\alpha}_{3}^{\text{re}} = b^{\text{re}} (a^{\text{re}} + |\alpha^{\text{re}}|) \dot{P}_{3}^{\text{rr}},$$
  
$$\dot{\alpha}_{33}^{\text{ire}} = b^{\text{ire}} \left( a^{\text{ire}} \dot{P}_{3}^{\text{ir}} - \alpha_{3}^{\text{ire}} \left| \dot{P}_{3}^{\text{ir}} \right| \right).$$
(6b)



Fig. 5. Comparison between experiment and modelling for a non poled ceramic.



Fig. 6. Comparison between experiment and modelling for a poled ceramic.

The other terms introduced in (4) and (6) are some constants of the model.

Furthermore, the piezoelectric coefficients  $d_{ij}$  depend on the initial polarisation. As a consequence, the coefficient  $d_{333}$  is written as a function of the irreversible polarization  $P_3^{ir}$ :

$$d_{333} = d_{333_0} + d_{333_{\max}} \frac{P_3^{\text{ir}}}{P_{\max}}.$$
 (7)

 $d_{333_0}$  (being possibly equal to zero) is the spontaneous piezoelectric coefficient in the absence of an induced polarisation.  $d_{333_{max}}$  is the maximum value of the piezoelectric coefficient induced when the polarisation is saturated to the value  $|P_k^{ir}| = P_{max}$ .

In addition, the material can loose its polarisation in an irreversible way when a compressive stress is increased beyond a threshold value. It means that the variables  $S_{33}^{dp}$  and  $P_3^{ir}$  are not completely independent as it has been assumed firstly. We introduce a depolarisation function  $f_{dp}$ :

$$f_{\rm dp} = 1 - \beta \left\langle -S^{\rm dp_{\rm C}} - S^{\rm dp}_0 \right\rangle \frac{\langle -T_{33} \rangle}{|T_{33}|} \text{ with } S^{\rm dp_{\rm C}} = S^{\rm dp}_{33}$$
(8)

where  $\beta$  is a constant of the model and  $S^{dp_c}$  represents the strain associated to the compressive stress causing depolarisation beyond the threshold value  $S_0^{dp}$ . Below this value,  $f_{dp}$  is equal to 1. This function is used for all terms depending on  $P_3^{ir}$ .

Finally, the total strain and polarisation can be written as:

$$S_{33} = \frac{T_{33}}{Y_{33}^{\rm E}} + \left( \left( d_{333_0} + d_{333_{\rm max}} \frac{P_3^{\rm ir}}{P_{\rm max}} \right) E_3 + S_{\rm max}^{\rm rem} \frac{|P_3^{\rm ir}|}{P_{\rm max}} \right) f_{\rm dp} + S_{33}^{\rm dp}$$
(9)

$$P_{3} = \varepsilon_{33}E_{3} + \left[ \left( d_{333_{0}} + d_{333_{\max}} \frac{P_{3}^{\text{ir}}}{P_{\max}} \right) T_{33} + P_{3}^{\text{ir}} \right] f_{\text{dp}} \quad (10)$$

## 4.2. Simulation of the creep behaviour

This formalism is used and programmed with Matlab software to simulate the behaviour of a poled and an unpoled material at room-temperature. Model parameters have been identified through other testings on PIC 255 cylinders.<sup>6</sup>

Experimental and simulated total strains are plotted according to time in Figs. 5 and 6 (experimental data are shifted of 0.4% for poled specimens, which corresponds to the initial longitudinal strain due to the poling process of the ceramic). Even if total strain levels are not very accurate, especially during the first two cycles (due to the necessity of taking  $T_0$  different from 0), the creep strain itself is quite subtly simulated. For example, the polarisation has an influence on the value of the calculated creep strain, which changes according to the applied stress as for real materials. Recovery strain is also well reproduced by the model.

## 5. Conclusion

In this paper, compressive creep testings have been carried out on a soft PZT. The influence of the macroscopic polarisation is highlighted on the anelastic and creep strain levels. The experimental curves are qualitatively reproduced by a viscoplastic phenomenological model, which shows the non-symmetry between creep and recovery deformations. Furthermore, an increase of 60 °C above room temperature modifies the creep behaviour of this soft PZT ceramic in an antagonistic way. On the one hand, the anelastic strain is reduced due to its pyroelectric properties. On the other hand, creep is assisted thermally. The same kind of study could be carried out on hard PZT, which are known to comprise pinning defects. These polarisation defects may influence the response of the material according to temperature.

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