Hydrostatic Piezoelectric Coefficient d_h of PZT Ceramics and PZN-PT and PYN-PT Single Crystals

L. BURIANOVA,[1*] P. HANA,[1] S. PANOS,[1] E. FURMAN,[2] S. ZHANG[2] & T.R. SHROUT[2]

[1]*Technical University of Liberec, Department of Physics and International Center for Piezoelectric Research, Halkova 6, CZ-461 17 Liberec 1, Czech Republic*

[2]*Materials Research Institute, The Pennsylvania State University, University Park, PA 16802, USA*

Abstract. The hydrostatic piezoelectric coefficient d_h of $Pb(Zr_xTi_{1-x})O_3$ ceramics (PZT), and of $Pb(Zn_{1/3}Nb_{2/3})O_3$-$PbTiO_3$ and $Pb(Yb_{1/2}Nb_{1/2})O_3$-$PbTiO_3$ (PZN-PT, PYN-PT, respectively) single crystals with compositions near to the morphotropic phase boundary (MPB) have been measured using a dynamic hydrostatic method. The effects of DC electric field and static component of hydrostatic stress on d_h of PZT ceramics, PZN-PT and PYN-PT single crystals were studied. Changes of the piezoelectric hydrostatic coefficients d_h caused by an electric field (DC bias) were observed along with pressure and temperature dependencies. The measurement of the hydrostatic piezoelectric coefficient d_h seems to be promising for investigation of intrinsic (single domain) and extrinsic (domain-walls) contributions to piezoelectric behavior of single crystals and ceramic materials.

Keywords: PZT ceramics, PZN-PT and PYN-PT relaxors, morphotropic phase boundary, hydrostatic piezoelectric coefficients

1. Introduction

It is well known that perovskite solid solutions have been intensively investigated because of their excellent piezoelectric and dielectric properties. They consist of simple cubic structure at high temperatures and variety of phases with polar states at lower temperatures. Among these materials are the ferroelectric $Pb(Zr_xTi_{1-x})O_3$ (PZT) solid solutions that present such properties in the morphotropic phase boundary (MPB) where rhombohedral and tetragonal phases coexist practically independently of temperature. Excellent piezoelectric properties were found for relaxor-based single crystals such as $Pb(Zn_{1/3}Nb_{2/3})O_3$-$PbTiO_3$ and $Pb(Yb_{1/2}Nb_{1/2})O_3$-$PbTiO_3$ (PZN-PT, and PYN-PT, respectively) near MPB [1–4]. This is the basis for practically all transducers and other piezoelectric devices.

*To whom all correspondence should be addressed. E-mail: lidmila.burianova@vslib.cz

Recently, considerable research has been focused on the piezoelectric d_{33} and d_{31} parameters and dielectric properties of PZT bulk ceramics and thin-films. The temperature dependence of piezoelectric coefficients of PZT ceramics using resonance method was reported in [5]. The effect of hydrostatic stress on piezoelectric coefficients of PZT was published in [9]. Besides d_{33} and d_{31}, no much attention was given to study the hydrostatic piezoelectric coefficient d_h variation under different influences (as DC electric field, temperature, pressure). Thus, in our previous papers [6–8], the methods for determining PZT material parameters as piezoelectric coefficients d_{33}, d_{31}, and hydrostatic piezoelectric coefficients d_h, g_h were described. In this article our work deals with d_h measurements by a dynamic hydrostatic method combining a static and variable superimposed pressure. The effect of a DC electric field and temperatures on the hydrostatic piezoelectric coefficients d_h of PZT ceramics as well as relaxor-based single crystals as PZN-PT and PYN-PT were investigated and described.

2. Hydrostatic Piezoelectric Coefficients

The determination of the dynamic hydrostatic coefficients utilizes a poled sample along z-axes under hydrostatic pressure $pT_{ii} = -p$, all other components of stress tensor equal to zero). Simultaneously, the applied electric field $E_3 = 0$, i.e. a short-circuit condition.

The polarisation P_3 on the sample induced by hydrostatic pressure p [7], is

$$
\begin{aligned}
P_3 &= (d_{31} + d_{32} + d_{33}) \cdot (-p) \\
&= (d_{33} + 2\, d_{31}) \cdot (-p) = d_h \cdot (-p),
\end{aligned} \quad (1)
$$

where $d_{i\lambda}$ are piezoelectric coefficients, $d_{31} = d_{32}$, and d_h is hydrostatic coefficient.

Measurement of the induced charge Q on the area A of the electrodes and pressure p permits calculation of the hydrostatic piezoelectric coefficient d_h.

The dynamic method was described in [7] by equation

$$
\frac{dQ}{dt} \frac{1}{A} = d_h \cdot \frac{dp}{dt}, \quad \text{where} \quad Q = Q_0 \sin \omega t. \quad (2)
$$

For the combined static and dynamic (with the sinusoidal shape) pressure excitation

$$
p = p_0 \sin \omega t + p_{static}, \quad (3)
$$

we can express the dynamic hydrostatic piezoelectric coefficient d_h as:

$$
d_h = \frac{I_0}{p_0 \omega A}, \quad (4)
$$

where I_0 is the amplitude of the current, p_0 is the amplitude of the dynamic pressure excitation, and ω is the angular frequency.

The hydrostatic piezoelectric coefficient d_h can be also calculated from the Eq. (1),

$$
d_h = d_{33} + 2d_{31}. \quad (5)
$$

Equation (5) was used for calculation of d_h values in our previous paper [6].

3. Experiment

The dynamic method of d_h measurement is based on dynamic oscillations of pressure around an appointed static pressure mastered by an auxiliary hydraulic piston at a frequency of about 0.5–1 Hz. The temperature control is realized by the PID temperature controller with a resistive heater and by the compressor cooler. The advantage of the described method is the elimination of pyroelectric effect and higher accuracy of the measurement. The possibility of an applied electric field influence on the device under test was realized. The block diagram of the experimental set up is shown in Fig. 1.

We studied PZT ceramics and PZN-PT, PYN-PT crystals. The PZT samples EPC856 and APC856 soft PZT ceramics with composition of $Pb(Zr_{58}Ti_{42})O_3$ were provided by American Piezoceramics International, Mackeyville, PA, USA. These samples were machined in disk-shaped (1 mm thick and diameter of 10 mm) and square plates $4 \times 4 \times 2$ mm^3.

The $Pb(Zn_{1/3}Nb_{2/3})O_3$-$PbTiO_3$ and $Pb(Yb_{1/2}\text{-}Nb_{1/2})O_3$-$PbTiO_3$ (PZN-PT, PYN-PT, respectively) single crystals were grown at the Pennsylvania State University, Material Research Institute, University Park, PA, USA. The used PZN-4.5% PT (001) and (111) cuts, and PYN-40% PT (111) cut were machined in rectangular plate-shaped samples with side length from 1.3 mm to 6.8 mm and thickness from 0.4 mm up to 0.6 mm. The measurements of the dynamic hydrostatic piezoelectric coefficient d_h were performed in the temperature range from 283 K to 433 K at a static pressure of 10 MPa and a dynamical pressure of 100 kPa.

All samples were electroded on both major surfaces and poled by the manufacturers.

4. Results and Discussion

The dependencies of the hydrostatic coefficient d_h of the soft PZT ceramics (EPC856) on DC electric field measured at various temperatures are displayed in Fig. 2. The typical relative decrease of d_h, caused by DC electric field change from zero to 700 kVm^{-1} was smaller than 20% at temperatures between 283 K and 423 K. The $d_h(E)$ increases to vicinity of Curie temperature ($T_C = 423$ K).

Figure 3 shows the similar dependencies of d_h vs. DC electric field and temperature for the soft PZT ceramics (APC 856). The comparison of the d_h values obtained from resonance method and dynamic hydrostatic method shows, that the second method gives slightly smaller values by up to 10 %. The details are shown in the Table 1.
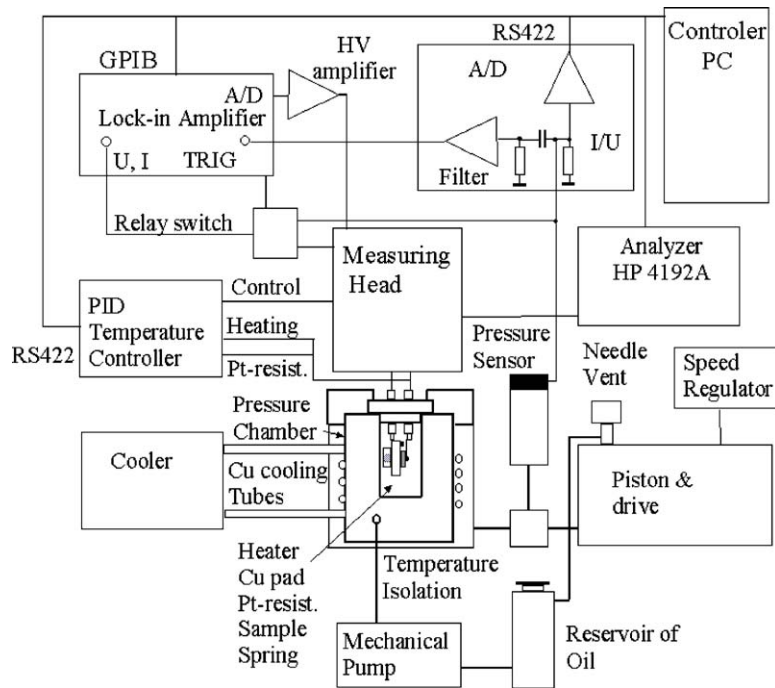
Fig. 1. A measuring set of high pressure dynamic method with the possibility of application of electric field onto sample.
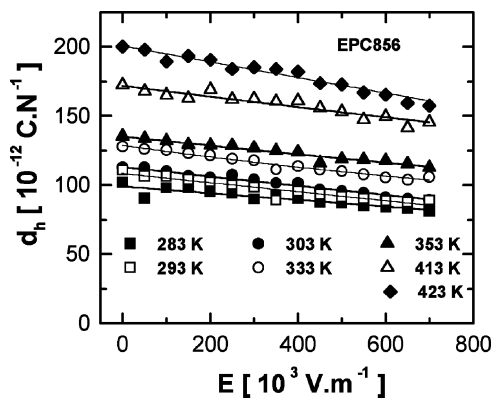

Fig. 2. Hydrostatic coefficient d_h of PZT ceramics (EPC 856) vs. DC electric field and various temperatures. Disk-shaped, 10 mm in diameter, 1 mm in thickness.

Fig. 3. Hydrostatic coefficient d_h of PZT ceramics (APC 856) vs. DC electric field and various temperatures. Plate-shaped, $4 \times 4 \times 2$ mm^3.

The temperature dependencies of the d_h coefficients at different DC electric field values applied to APC856 ceramics are shown in Fig. 4. A noticeable influence of the electric field on Curie temperature T_C position was exhibited.

Apparently, the phase transition is shifted to higher temperature by increasing the electric field. This is due
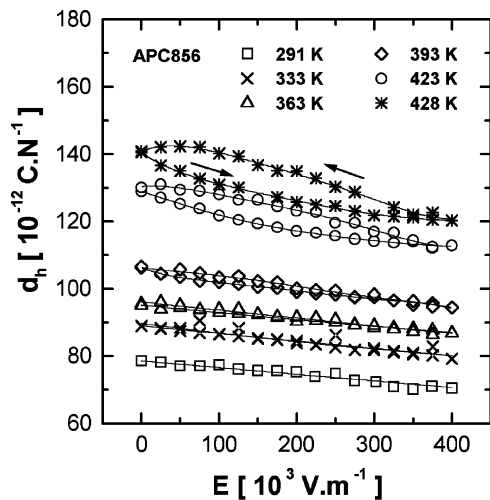
to the upkeep of polar state by the applied electric field which delays the phase transition ferroelectric-paraelectric to high temperature.

The samples of PZN-PT single crystals exhibit small variation of d_h with hydrostatic pressure in the pressure

Table 1. Hydrostatic piezoelectric coefficients d_h of PZT ceramics at room temperature (APC International, Mackeyville, PA, USA.)

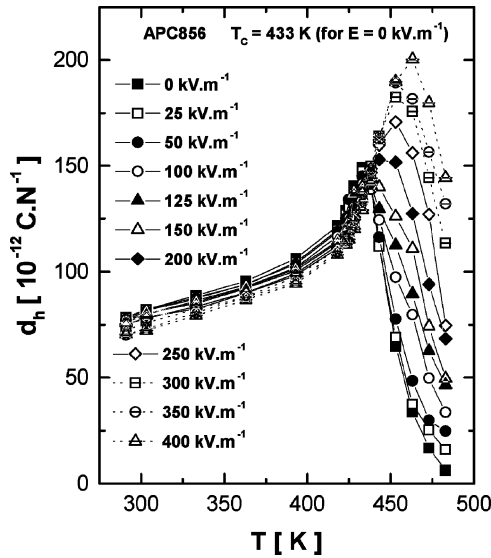|		PZT (APC856)	PZT (EPC856)
Resonance method	d_{33} [10^{-12} C·N^{-1}]	558	591
	$-d_{31}$ [10^{-12} C·N^{-1}]	236	237
	d_h [10^{-12} C·N^{-1}]	86	117
Dynamic hydrostatic method	d_h [10^{-12} C·N^{-1}]	79	111

Fig. 4. Temperature dependencies of hydrostatic coefficient d_h of PZT ceramics (APC 856) at various DC electric field. Plate-shaped, $4 \times 4 \times 2$ mm^3.

Fig. 5. Hydrostatic coefficient d_h of PZN-4.5%PT (001) cut and PZN-4.5%PT (111) cut vs. hydrostatic pressure. Plate-shaped, $6.8 \times 6.8 \times 0.48$ mm^3, $3.2 \times 3.2 \times 0.6$ mm^3, respectively.

Fig. 6. Hydrostatic coefficient d_h of PZN-4.5%PT (001) cut vs. DC electric field at various temperatures. Plate-shaped, $2.0 \times 4.2 \times 0.46$ mm^3.

range of zero to 70 MPa. Decrease of d_h value does not exceed 5% of initial values as shown in Fig. 5.

In Fig. 6 is plotted the hydrostatic coefficient d_h of PZN-4.5%PT (001) cut vs. DC electric field at various temperatures. We can observe a change of the shapes of curves at temperatures above 373 K and low DC electric fields. We suppose that it is connected with the phase transition from ferroelectric phase to paraelectric phase. In vicinity of Curie temperature $T_C = 423$ K the value of d_h is very low at zero DC bias. For low electric field values we can observe a sharp peak of the coefficient d_h and its consequent exponential decay with increasing bias. The peak position shifts toward higher applied electric field values with increasing tempera-
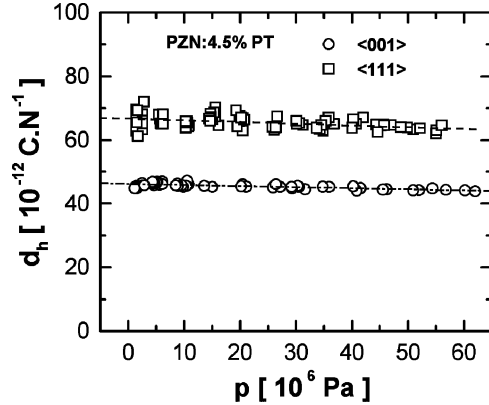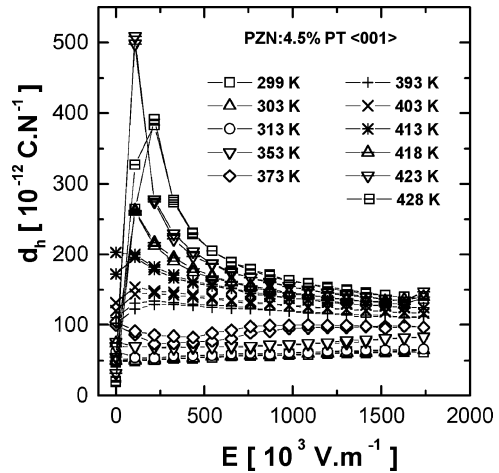
tures. Furthermore, the peak magnitude undergoes a decrease with increasing electric field. The piezoelectric hydrostatic coefficient peak could be due to the domain subdivision which takes place when the temperatures approach T_C. Thus, an increase of temperature engages an increase in the domain density. The influence of electric field on domains (domain reorientation) will depend on their density, and consequently, an increase in domain density will need a high electric field for domain alignement. This could explain the peak shift to high electric fields when the temperature increases [10, 11]. The exponential decay of right
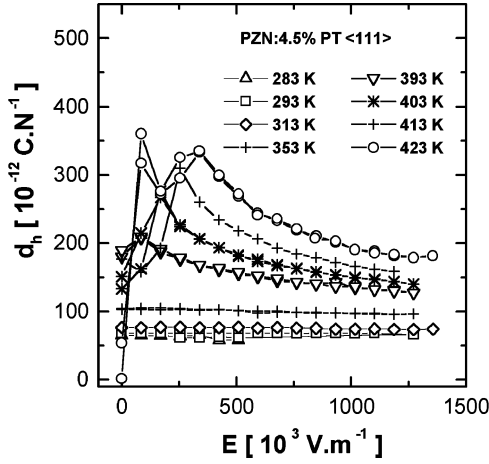
Fig. 7. Hydrostatic coefficient d_h of PZN-4.5%PT (111) cut vs. DC electric field at various temperatures. Plate-shaped, $3.2 \times 3.2 \times 0.59$ mm^3.
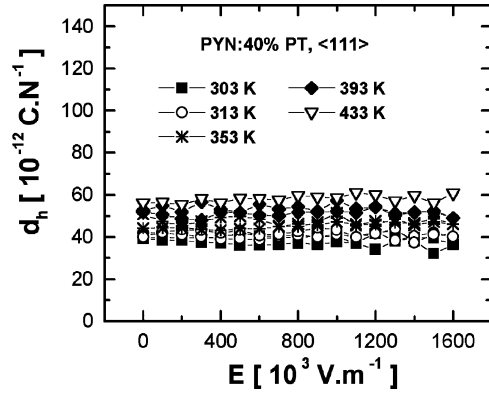

Fig. 8. Hydrostatic coefficient d_h of PYN-PT (111) cut vs. DC electric field at various temperatures. Plate-shaped, $2.2 \times 1.3 \times 0.5$ mm^3.

side of the peak could be associated to a relaxation phenomenon stimulated by charge carrier screening in the material bulk due to the increase conductivity with temperatures. It was shown, in our experimental measurement, the value of d_h equaled to $47 \cdot 10^{-12}$ CN^{-1} for $T = 298$ K and $E = 0$ Vm^{-1}. This value is smaller than $d_h \approx 250 \cdot 10^{-12}$ CN^{-1}, which was calculated from Eq. (5) using measured d_{33} and d_{31} [1]. To put in evidence this difference further investigation of the effect of a DC electric field on domain engineering states and domain wall configuration is required.

The hydrostatic coefficient d_h of PZN-4.5%PT (111) cut vs. DC electric field at various temperatures is plotted on Fig. 7. In this case we can observe significantly different behavior for temperatures in vicinity of Curie temperature $T_C = 423$ K. The coefficient d_h reaches two maxima for low bias fields. Possibly it could be caused by presence of intermediate orthorhombic phase. The values of d_h at low temperatures up to 353 K show slight change. Experimental value of $d_h = 68 \cdot 10^{-12}$ CN^{-1} for $T = 298$ K and $E = 0$ Vm^{-1} is in good agreement with [1].

The hydrostatic coefficient d_h of PYN-40%PT (111) cut vs. DC electric field at various temperatures is plotted in Fig. 8. The relative change of d_h, caused by DC electric field of 1600 kVm^{-1} was smaller than 6% at temperatures between 303 K and 393 K. The d_h increases with increasing temperature up to $T_C = 523$ K.

5. Conclusion

The non-linear effects of DC bias electric field on the hydrostatic piezoelectric coefficient d_h for variable temperature in soft PZT ceramics, and PZN-PT and PYN-PT relaxor based single crystals were studied. The hydrostatic coefficient d_h was determined by the dynamic hydrostatic method. We believe that the discrepancy between the values of the d_h, determined by Eq. (5), and those measured directly (using dynamic hydrostatic method) is mainly caused by extrinsic contributions of domain walls. This hypothesis supports the significant contribution of domain walls motion observed and described in the paper [1] on domain engineered samples of PZN-4.5%PT single crystals, (001) cut.

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