Non-linear properties of PZT ceramics in the wide temperature range

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Abstract

The paper deals with measurements of non-linear properties of $Pb(Zr_xTi_{1-x})O_3$ (PZT) type piezoelectric ceramics in the composition near to the morphotropic phase boundary (MPB). The hydrostatic piezoelectric coefficient d_h was determined using the piezoelectric coefficients d_{33} and d_{31} obtained by resonance method. The temperature dependencies of d_{33} and d_{31} were compared with ones obtained by laser interferometry method.

The differential hydrostatic coefficient $d_{h,diff}$ of PZT ceramics has been measured using a dynamic hydrostatic method. The influence of a DC bias electric field on $d_{h,diff}$ coefficient of a PZT ceramics was studied. Changes of the hydrostatic piezoelectric coefficients $d_{h,diff}$ caused by a bias electric field were observed at various temperatures.
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1. Introduction

Non-linear properties and hysteresis of piezoelectric materials are undesirable in many applications. The description and experimental investigation of such properties are necessary for the design of piezoelectric devices such as actuators, sensors, resonators and transducers in general. Recently, considerable research has been focused on the investigation of nonlinearity of PZT bulk ceramics, PZN-PT relaxor-based single crystals, and thin-films.[1–3]

In $Pb(Zr_xTi_{1-x})O_3$ (PZT) ceramics, the nonlinearity and hysteresis originate from small movements of domain walls. As a result, all piezoelectric, elastic and dielectric properties, both linear and nonlinear, depend on the temperature. The temperature dependencies of piezoelectric coefficients d_{33} and d_{31} of PZT ceramics are referred by Zhang.[1] In the Zhang's paper, the variations of d_{33} and d_{31} for soft and hard types of piezoelectric ceramics were determined by resonance method in the temperature range from 15 to 300 K. In our previous paper[4] the temperature dependencies of the coefficients d_{33} and d_{31} of PZT ceramics were obtained by laser interferometry, using the method of the measurement of the sample strain induced by an applied electric field. Masys et al.[5] used the laser interferometry method for determination of the piezoelectric coefficients $d_{i\lambda}$ of PZT ceramics as a function of the amplitude of the electric field and frequency.

The presented paper deals with resonance method of measurement of the piezoelectric properties of the hard PZT ceramics in a wide temperature range. The temperature dependencies of piezoelectric coefficients d_{33}, d_{31}, and hydrostatic coefficient d_h were determined. The non-linear behaviour such as influence of the bias field on the differential hydrostatic coefficients was investigated at various temperatures.

2. Experiment

The temperature dependencies of the piezoelectric low-field properties were measured using the "IEEE Standard on Piezoelectricity" resonance technique.[6] The tested samples were put in the temperature chamber Saunders 4220A and connected to the measuring set outside the chamber. The precise network analyzer Agilent E5100A was used to determine the serial and parallel resonance frequencies. The

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static capacitance C_0 was measured using an HP 4263B RLC meter at the frequency 1 kHz. The temperature in the chamber was set in the range from -40 to $150\,^\circ$C with the step of $10\,^\circ$C. At each temperature, the samples were stabilized for about 5 min. To avoid the generation of the pyroelectric charge, the sample was short-circuited during the heating and cooling. The error, caused by the accuracy of measurement system and temperature drift, was determinated from the measured data and was about 1%.

We studied the PZT ceramics samples of APC841 (hard) and APC856 (soft) types, provided by American Piezoceramics International, Mackeyville, PA, USA. For the APC841, the d_{31} coefficient was measured on thin bars, electroded on both major surfaces and poled in the direction of their thickness. The dimensions of the bar-shaped samples were $4\,\text{mm} \times 15\,\text{mm} \times 1\,\text{mm}$. The d_{33} coefficient was measured on the bars with the square cross section. The bars electroded at the ends were $2\,\text{mm} \times 2\,\text{mm} \times 10\,\text{mm}$ in size.

The hydrostatic piezoelectric coefficient d_h can be calculated from the equation:

$$d_h = d_{33} + 2d_{31}. \tag{1}$$

The piezoelectric coefficients d_{33} and d_{31} of APC841 were measured both by the resonance and the interferometry methods. The single beam interferometry was provided in the temperature range from 150 to 325 K.[4] The temperature control of the laser interferometer methods was realized using an optical helium closed cycle cryostat Oxford Instruments. The dimensions of the plate-shaped samples measured by laser interferometry were $4\,\text{mm} \times 4\,\text{mm} \times 3\,\text{mm}$.

The dynamic hydrostatic method was used to determine the differential hydrostatic coefficient $d_{h,\text{diff}}$. This method was described in our previous paper.[7] The experimental set up of the $d_{h,\text{diff}}$ coefficient measurement under bias field is schematically given in Fig. 1. The differential hydrostatic coefficient $d_{h,\text{diff}}$ is defined by the equation:

$$d_{h,\text{diff}} = \frac{I_0}{p_0 \omega A}, \tag{2}$$

where I_0 is the amplitude of the current, p_0 is the amplitude of the dynamic pressure excitation, A is area of the electrode, and ω is the angular frequency.[7]

The measurements of the $d_{h,\text{diff}}$ coefficient were performed under constant bias field in the temperature range from the room temperature up to $200\,^\circ$C at a static pressure of 10 MPa, the frequency and the amplitude of the dynamical pressure excitation were 1 Hz and 100 kPa, respectively. The hard APC841 and soft APC856 disc-shaped samples were 1 mm thick and 12 and 15 mm in diameter, respectively. All samples were electroded and poled by the manufacturers.

3. Results and discussion

The temperature dependencies of the piezoelectric coefficients d_{33} and $-d_{31}$ for the hard type ceramics APC841, measured by resonance method, are shown in Fig. 2. The temperature dependencies of both d_{33} and $-d_{31}$ were fitted by polynomial function of the third order. The piezoelectric coefficients d_{33} and $-d_{31}$ increase with the temperature in the range from -40 to $150\,^\circ$C. Our experimental results show the same character of the temperature dependencies of d_{33}, $-d_{31}$ as reported in.[1] The temperature dependencies of the piezo-
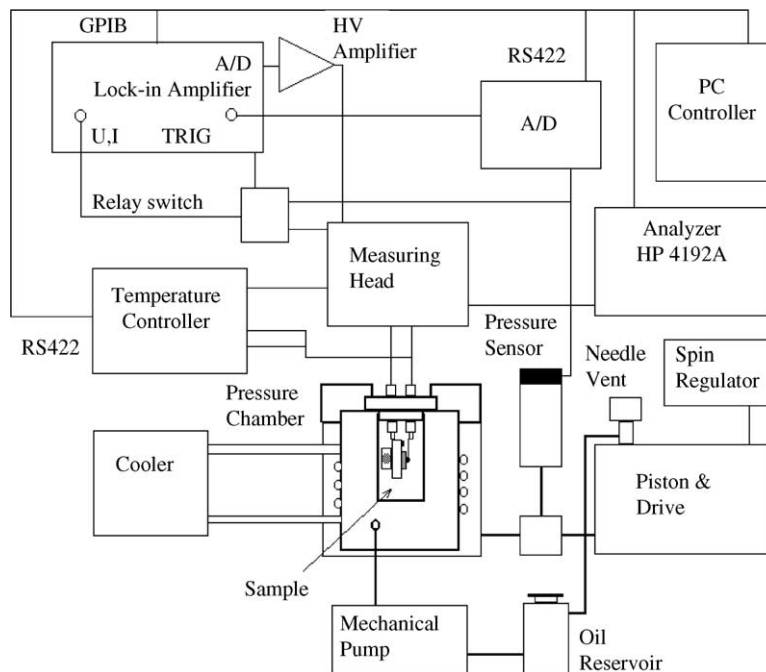

Fig. 1. A measurement set of the dynamic method with the possibility of the application of the bias electric field onto the sample.
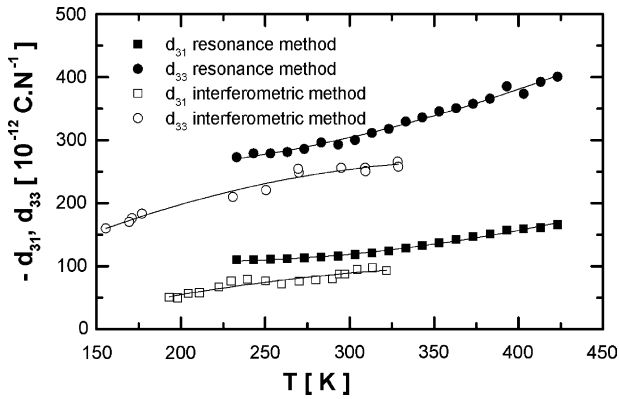
Fig. 2. The temperature dependence of the piezoelectric coefficients d_{33}, d_{31} of the PZT ceramics APC841 type; bar with the square cross section, 2 mm × 2 mm × 10 mm and bar-shaped, 4 mm × 15 mm × 1 mm (resonance method); plate-shaped, 4 mm × 4 mm × 3 mm (interferometric method).

electric coefficients were determined with the accuracy of 2%. This is better than the accuracy of the laser interferometry measurement. Temperature dependencies of d_{33}, $-d_{31}$ coefficients of PZT type APC841 were measured by the interferometry method in the temperature range 150–325 K, see Fig. 2. However, the errors of d_{33} and d_{31} single-beam micro-interferometer measurements[4] at low temperatures are about 10%. The d_h coefficient calculated from Eq. (1) is much smaller than d_{33} ($d_{33} \approx -2d_{31}$). From these reasons, the influence of temperature on d_h coefficient has been determined only by resonance method. The temperature dependence of normalized hydrostatic coefficient d_h of the hard APC841 is shown in the Fig. 3. Any comparison between hard and soft materials is not provided, because the corresponding samples for soft material (as bar shaped samples for d_{33} measurement) were not at one's disposal.

Dynamic hydrostatic method[7] has been used to determine the differential hydrostatic coefficients $d_{h,diff}$ of PZT ceramics as a function of a bias electric field at various temperatures. Fig. 4 shows the bias field dependence of $d_{h,diff}$ coefficient of hard PZT ceramics of type APC841 at various temperatures. Bias electric field was applied along the poling direction to the poled hard PZT ceramics, in this case, we designate the bias field positive. A small decrease in $d_{h,diff}$ coefficient
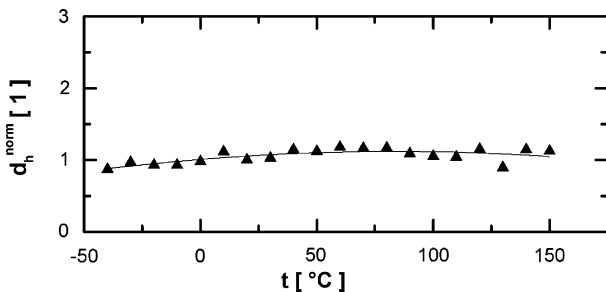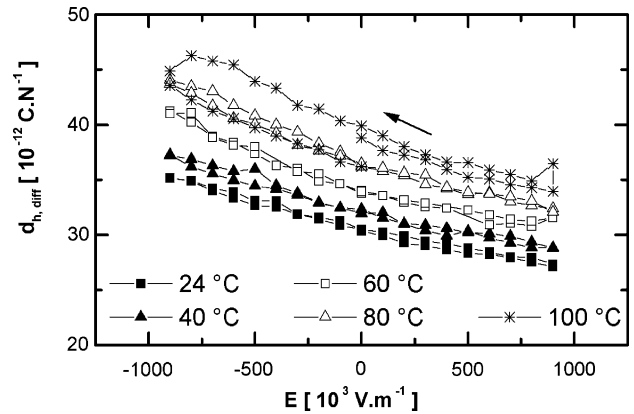

Fig. 4. The hydrostatic coefficient $d_{h,diff}$ of the APC841 disc-shaped, 12 mm in diameter, 1 mm in thickness vs. bias field at various temperatures.

was measured. The bias field applied along the poling direction probably contributes to the pinning of the domains. A positive bias field does not appreciably increase the total polarization.[5] The relative decrease of $d_{h,diff}$, caused by bias electric field in the range from 0 to 900 kV m^{-1}, was smaller than 10% at temperatures between 24 and 60 °C. When the bias was applied in the opposite direction, an increase in $d_{h,diff}$ coefficient was observed. A negative bias maybe causes a de-pinning of the domain walls. Thus, the extrinsic contribution to the piezoelectric response caused by domain walls motions increases. These dependencies were measured in the bias field range from 0 to ±900 kV m^{-1}, i.e. below coercive electric field. The $d_{h,diff}$ coefficient increases with increasing temperature. The hysteresis appears at the temperature 100 °C.

Fig. 5 shows the bias field dependencies of $d_{h,diff}$ coefficient of the hard ceramics APC841 at higher temperatures. The $d_{h,diff}$ coefficient increases with the applied negative bias field until the field −500 kV m^{-1} at temperature 140 °C. A further increase of the negative bias has caused an opposite poling of the sample. The effect of the decrease of pinning appears, if the negative bias field is decreased. The pinning
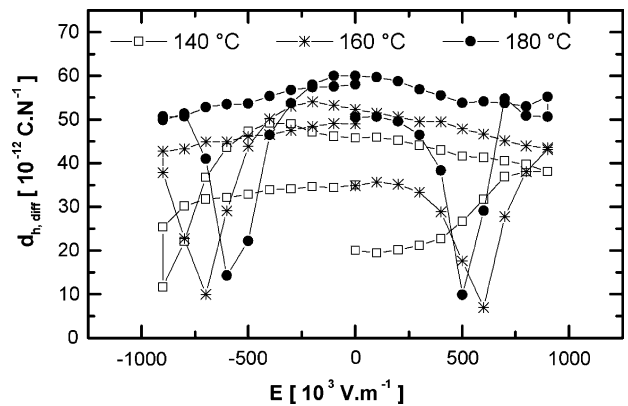

Fig. 3. The temperature dependence of the normalized hydrostatic coefficient d_h of the APC841; bar with the square cross section, 2 mm × 2 mm × 10 mm and bar-shaped, 4 mm × 15 mm × 1 mm (resonance method).

Fig. 5. The hydrostatic coefficient $d_{h,diff}$ of the APC841 disc-shaped, 12 mm in diameter, 1 mm in thickness vs. bias field at various temperatures (the temperatures are higher than 100 °C).
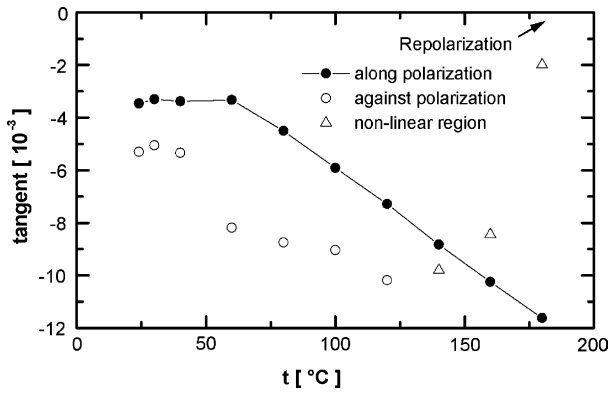
Fig. 6. The tangent of bias field dependence of $d_{h,diff}$ coefficient vs. temperature; APC841 disc-shaped, 12 mm in diameter, 1 mm in thickness.
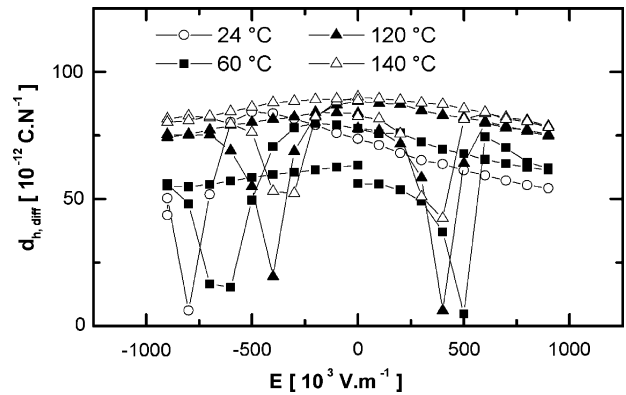

Fig. 7. The hydrostatic coefficient $d_{h,diff}$ of the APC856 disc-shaped, 15 mm in diameter, 1 mm in thickness vs. bias field at various temperatures.
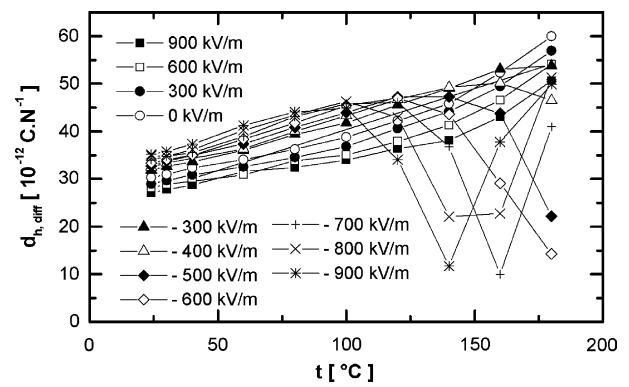

Fig. 8. The temperature dependence of the hydrostatic coefficient $d_{h,diff}$ of the hard APC841 disc-shaped, 12 mm in diameter, 1 mm in thickness at various bias fields.

induced by the bias electric field decreases, resulting in the easier domain wall motion. As the consequence, the coefficient $d_{h,diff}$ increases. At higher temperatures, the repoling effect appears under lower bias field.

The bias field dependencies of $d_{h,diff}$ (along and opposite the poling direction) at various temperatures were fitted by linear function. The tangent of the bias field dependencies of $d_{h,diff}$ as a function of the temperature is shown on Fig. 6. We distinguish three cases: first, the bias field applied along polarization direction. We can observe an almost constant value of the tangent of these dependencies at temperatures up to 60 °C. Above 60 °C the magnitude of the tangent of bias field dependencies $d_{h,diff}$ started to increase linearly with temperature. This behaviour is valid for poled and repoled samples by applied positive bias field during cycling. Second, the bias field was applied opposite to the polarization direction but did not cause depoling and switching of polarization. Here, the behaviour of tangent of the bias field dependence of $d_{h,diff}$ was observed. Tangent increases with temperature till the value of $d_{h,diff}$ reaches the maximum at zero bias field. The dependence is not linear because the stabilizing effect of the bias field is not present. Third, processes inductive of depolarization and repolarization of the samples. The change of the tangent is governed probably by "softening" of the polarization during the process of depolarization. The zero value of tangent means, that the maximum of the piezoelectric coefficient $d_{h,diff}$ was reached.

The dependencies of $d_{h,diff}$ coefficients of the soft PZT ceramics of the type APC856 on the bias electric field at various temperatures are displayed in Fig. 7. The $d_{h,diff}$ coefficient shows the increase, if the negative electric field is applied, till the samples start to be depoled. The samples are depoled at about $-800\,\text{kV}\,\text{m}^{-1}$ at room temperature. The further increase in the negative bias causes a repoling of the sample. In the whole cycle, we obtain the typical "butterfly" shape of the bias dependencies.

To compare of the main piezoelectric properties of the hard APC841 and soft APC856 materials, the experimental investigations of $d_{h,diff}$ as function of electric field were performed.

The results are shown and described in the Fig. 5 (APC841) and Fig. 7 (APC856). It is obviously from the comparison of these dependencies, that nonlinearities like "butterfly" shape dependencies appears for smaller electric fields (related to the smaller temperatures) for APC856. The lower coercive field is characteristic for the soft ceramics in comparison with the hard one.

Fig. 8 shows the temperature dependence of the hydrostatic coefficient $d_{h,diff}$ of the hard APC841 at various bias fields. A noticeable influence of the temperature on coercive field value was observed. A significant minimum of the temperature dependence of $d_{h,diff}$ was observed. In the minimum of the temperature dependence of $d_{h,diff}$, the corresponding bias represents the magnitude of the coercive field E_c. From the Fig. 8 we see, that this coercive field decreases with increasing temperature. Extinction of spontaneous polarization near the temperature of the phase transition should naturally appear for zero or a small positive bias.

4. Conclusion

Some of non-linear piezoelectric properties of the hard and soft ceramics (APC841 and APC856) were experimen-

tally investigated by resonance, laser interferometry, and dynamic hydrostatic method. The piezoelectric coefficients d_{33} and d_{31} of the hard poled ceramics APC841 were measured by resonance method, and were used for calculation of the hydrostatic coefficient. We found, that the resonance method gives the more precise results as compared to laser interferometry method, especially at low temperatures. The typical constant bias field dependencies experimentally investigated by dynamic hydrostatic method were studied also in the temperature range from 24 to 200 °C. The significant shifts of the temperature dependencies of the differential hydrostatic coefficient $d_{h,diff}$ are shown. The phenomenon of polarisation and possible influence of pinning and extrinsic contributions in the hard ceramics (APC841) are discussed. The noticeable influence of the temperature on coercive field value was exhibited both for hard and for soft materials.

Acknowledgements

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