

L. KOZIELSKI*, M. ADAMCZYK*

MAGNETIC AND PIEZOELECTRIC PROPERTIES OF MAGNETOELECTRIC LAMINATE TRANSFORMER

MAGNETYCZNE I PIEZOELEKTRYCZNE WŁAŚCIWOŚCI MAGNETOELEKTRYCZNEGO TRANSFORMATORA WARSTWOWEGO

Modern electronic devices development steady trends toward miniaturization and multifunctionality. The application of such advanced devices is often realised by superimposing two different effects. In recent years, there was great interest shown in multiferroic materials in form laminated heterostructures taking advantage of mentioned superposition. In these stacked structured electric and magnetic properties are strain coupled leading to the possibility of controlling either magnetic polarization by application of electric fields or electric polarization by means of applied magnetic fields.

In this paper novel construction of piezoelectric transformer was proposed with electric current induced magnetic field generation, without any coil or wires. This magnetolectric laminate Piezoelectric Transformer from one layers of investigated piezoelectric material and second layer of commercial magnetostrictor was fabricated. This construction exhibits many interesting and unusual piezomagnetic properties and a demonstration of output voltage gain controlling via the application of magnetic field across the new heterostructure is presented.

Keywords: Piezoelectric Transformer (PT), Magneto Electric (ME) effect, PBZT, PLZT

Rozwojowi nowoczesnych urządzeń elektronicznych nierozłącznie towarzyszy dążenie do ich miniaturyzacji. W przypadku obecności uzwojeń w takich aplikacjach, trend ten to przejawia się w ich eliminacji poprzez zastosowanie nowych rodzajów przekształcania energii.

W artykule została przedstawiona i przetestowana praktycznie nowa konstrukcja transformatora piezoelektrycznego, również realizującego funkcję inteligentnego czujnika pola magnetycznego, generującego napięcie wyjściowe proporcjonalne do natężenia tego pola. Zastosowano strukturę wykorzystującą materiał magnetostrykcyjny w podwójnej funkcji, do przewodzenia prądu i wytwarzania drgań mechanicznych, co pozwoli na polepszenie parametrów przetwarzania energii (m.in. małe straty układu, mały pobór mocy), zapewniając w ten sposób prosty sposób sterowania układem oraz obniżenie kosztów produkcji.

Drugim celem tego artykułu jest dobór efektywnego materiału piezoelektrycznego do zastosowanie w sekcji wyjściowej proponowanej konstrukcji i finalne sprawdzenie jego działania.

1. Introduction

The magnetolectric (ME) effect as a change of electric polarization under magnetic field, or electric field induced magnetization was the focus of significant research interests recently. Theories of multiferroic phase interactions have been developed along with the progress in experimental studies, especially accounting for their coexistence and strong coupling between ferroelectricity and incommensurate magnetism. Scientific publications authors explained a paths from magnetic field induced local distortions to global ones by interaction of different charge ordering forms [1]. Hence, the response in induced multifunctional system with ferroelectric and

ferromagnetic ceramics is manifested in the form of cooperative atomic displacements [2].

The materials with multiferroic effect have been investigated for a variety of compounds including manganites, magnetite, nickelates, etc.[3,4,5,6,7]. However, these composites have much problems in reliability, low electric resistivity and an insufficient magnetolectric voltage coefficient for practical applications. Unfortunately, the both effects are contradictive additionally, what practically means, that the transition metal d electrons, which are essential for magnetism, reduce the tendency for off-center ferroelectric polarization. Consequently, in most applications the Magnetolectric Effect (ME) is created via strain to an additional but intimately

* UNIVERSITY OF SILESIA, DEPARTMENT OF MATERIALS SCIENCE, 41-200 SOSNOWIEC, 2 ŚNIEŻNA STR., POLAND

connected layers of piezoelectric and magnetostrictive materials [8]. The good example of strain driven application is a Piezoelectric Transformer (PT) with magnetic feedback on gain generated by magnetically induced internal tensions [9]. This application, as a resonant device, is particularly efficient, because there is a giant enhancement in magnitude of ME coefficient at the resonance frequency [10].

The main aim of this work was to find the performance-enhancing material for piezoelectric layer of proposed device. According the literature, the best reported candidates were PBZT and PLZT ceramics, due to the highest piezoelectric voltage coefficients, that can reach the level of 30 mV*m/N [11,12,13,14]. Therefore, the two prototypes were fabricated and transformation ratio (gain) experimentally proved. For the magnetic part of PT the TbDyFe (Terfenol-D) was chosen, the giant magnetostrictive alloy with very high energy density of 4.9–25 kJ/m³ (Etrema Products. Inc. Ames. I A).

The main advantage of investigated prototypes, in comparison to conventional magneto-electric devices is, that our ME transformer does not require secondary coils in order to change its output voltage. Additionally, it exhibits higher gain, defined as the output to input voltage ratio, and a wider bandwidth. Functionally moreover, it has an advantage of low input impedance, so that we can use low-voltage current drive for the magnetostrictive Terfenol-D input part and high output impedance for the PZT part. The combination of these advantages offers great potential for applications of this new type of magnetoelectric transformer.

2. Experimental

2.1. Sample preparation

Mixed oxide method was used for preparation of PLZT (Pb_{0,97}La_{0,03})(Zr_{0,52}Ti_{0,48})O₃ and PBZT (Pb_{0,84}Ba_{0,16})(Zr_{0,54}Ti_{0,46})O₃ ceramics, the compositions expected to have the highest piezoelectric charge coefficient g_{31} . Commercially available PbO, ZrO₂, La₂O₃, BaCO₃ and TiO₂ powders were used as starting materials (high purity (99%): lead (II) oxide (Sigma Aldrich, no.211907), lanthanum (III) oxide (Sigma Aldrich, no. L4000), barium carbonate (Sigma Aldrich, no. 11729), zirconium (IV) oxide (Sigma Aldrich, no. 230693), titanium (IV) oxide (Sigma Aldrich, no. 248576). The weighted powders for stoichiometric composition were mixed for 24h with zirconia balls (3 mm in diameters), as the grinding media and ethanol, as the solvent in a planetary mill Retch PM400. After milling, the resultant slurry was calcinated in a high-temperature furnace at 900°C for 2 h. Consequently, the calcined powders was

fed in the 23-mm in diameter stainless steel die and subjected to pressure to obtain of 1GPa to obtain a compact sample.

The final sintering of both samples had been undergone at 1250°C in 2 hours time using refractory furnace (Pyrotech 1300C, Pyrotech USA), equipped with temperature controller (Programmer 40/16SE). Five % excess of PbO was used to compensate the lead evaporation during calcination and sintering for PLZT ceramics at the elevated temperature.

From the obtained samples the rectangular in shape plates (20×10×0.5 mm) were cut out.

To evaluate electric spontaneous polarisation presence (zero field), the Piezoelectric Response Microscopy (XE-100 PSIA) was chosen and after that, the samples were poled in electric field of 3 kV/mm at temperature 140°C in silicon oil for creating oriented polarisations in longitudinal direction.

2.2. Material testing

The obtained PBZT ceramics sample density value was equal 6,8 g/cm³ (Archimedes method), whereas the PLZT value was equal 7,83 g/cm³. The AFM microscopy recorded topography of both materials, is presented in figure 1. The average grain size of polished and chemically etched samples, by the mixture of etching acids (HCl + HF), were equal 4,6 μm (Fig.1 a) and of 1,3 μm (Fig.1 c) for PBZT and PLZT samples, respectively (Lince software). The density of the ceramics tended to decreased with the increasing of grain size value, what corresponds with the PFM micrographs of the pellets surface of both ceramics.

Figures 1 b and d are presenting the PFM mode (Piezoresponse Force Microscopy) mapping of the ferroelectric domain polarization (potentials mapping) from the same area. As was mentioned above, application of such materials for efficient ME and for power conversion requires μm -sized domain structures with not high potential amplitude. It was estimated that about 25%–50% of the dielectric response at room temperature was from extrinsic sources [15]. The extrinsic contribution to the dielectric constant of PZT type materials was mainly attributed to 180° domain wall motion, which increased with grain size. For fine grained PZT, non-180° switching was negligible, suggesting that the pinning of non-180° domain walls is very strong. In studies on the direct and converse longitudinal piezoelectric coefficients of PZT ceramics as a function of driving either stress or electric field, it was found that the ferroelastic non-180° domain wall motion was limited. Thus extrinsic contributions to the piezoelectric response is small in fine grain PZT especially those under 1.5 μm in size. However, as the grains became bigger (1.5μm), nonlinear

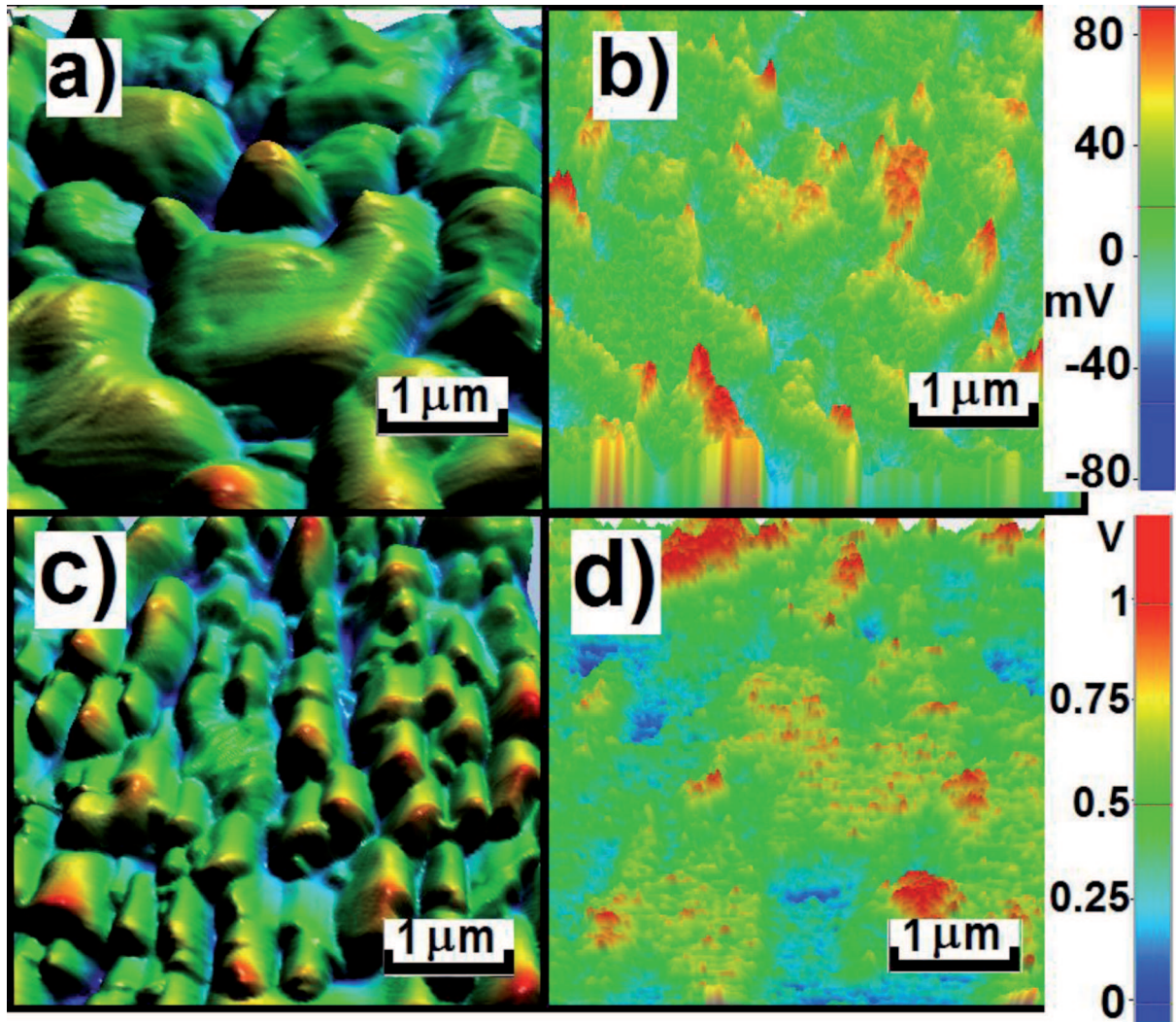


Fig. 1. Surface topography images of PBZT (a) and PLZT (c), consequently the ferroelectric domains pattern from the same area recorded in PFM mode (b) and (d), respectively

enhancement between the converse piezoelectric coefficient and the electric driving field was observed. This indicated, that the grain size is quite important factor accompanied by significant ferroelectric non-180° domain wall motion under external excitation for example via magnetic field or stress. We can also conclude this discussion, that the non-180° domain switching in large grained PBZT ceramics will be proportionally much easier and more significant than that in fine grained PZT.

We present an analysis of the contrast for the detection of ferroelectric domains at surfaces of PBZT (Fig.1 b) and PLZT (Fig.1 c) ceramics. The domain contrast can be attributed to mechanisms of the electrostatic tip-sample interaction at the domain boundaries caused by surface charges [16]. A careful analysis of the movement of the cantilever with respect to grain orientation allows a clear attribution of the observed PBZT domain contrast to the micrometers size (smaller the same contrast areas) and low driving forces (high PLZT potential

of the level of 1V whereas for PBZT ceramics it is on the level of several tens of mV). Thus the high domain mobility offered by PBZT ceramics seems to be highly desired.

2.3. XRD characterization of PBZT and PLZT samples

The x-ray measurements patterns of investigated materials were obtained by PANalytical X'Pert Pro Diffractometer and later analyzed by the program X'Pert High-Score Plus. Presented in Figure 2 recorded diffraction patterns are characteristic for pseudo regular perovskites structure. According our XRD investigation, there were coexistence of tetragonal P4mm and rhombohedral R3c space groups for both compositions (Fig. 2). This fact confirms experimentally, that we had chosen proper material candidates of the highest piezoelectric coefficients, hence the best performance of PZT type ceramics exist

TABLE 1

Sintering Atmosphere	a [Å] (R3c)	c [Å] (R3c)	a [Å] (P4mm)	b [Å] (P4mm)	c [Å] (P4mm)	$\delta = \frac{c}{a}$ distorsion
PBZT	5.758928	14.17603	4.056225	4.056225	4.11479	0.0144
PLZT	5.75318	14.24959	4.07233	4.07233	4.11144	0.0096

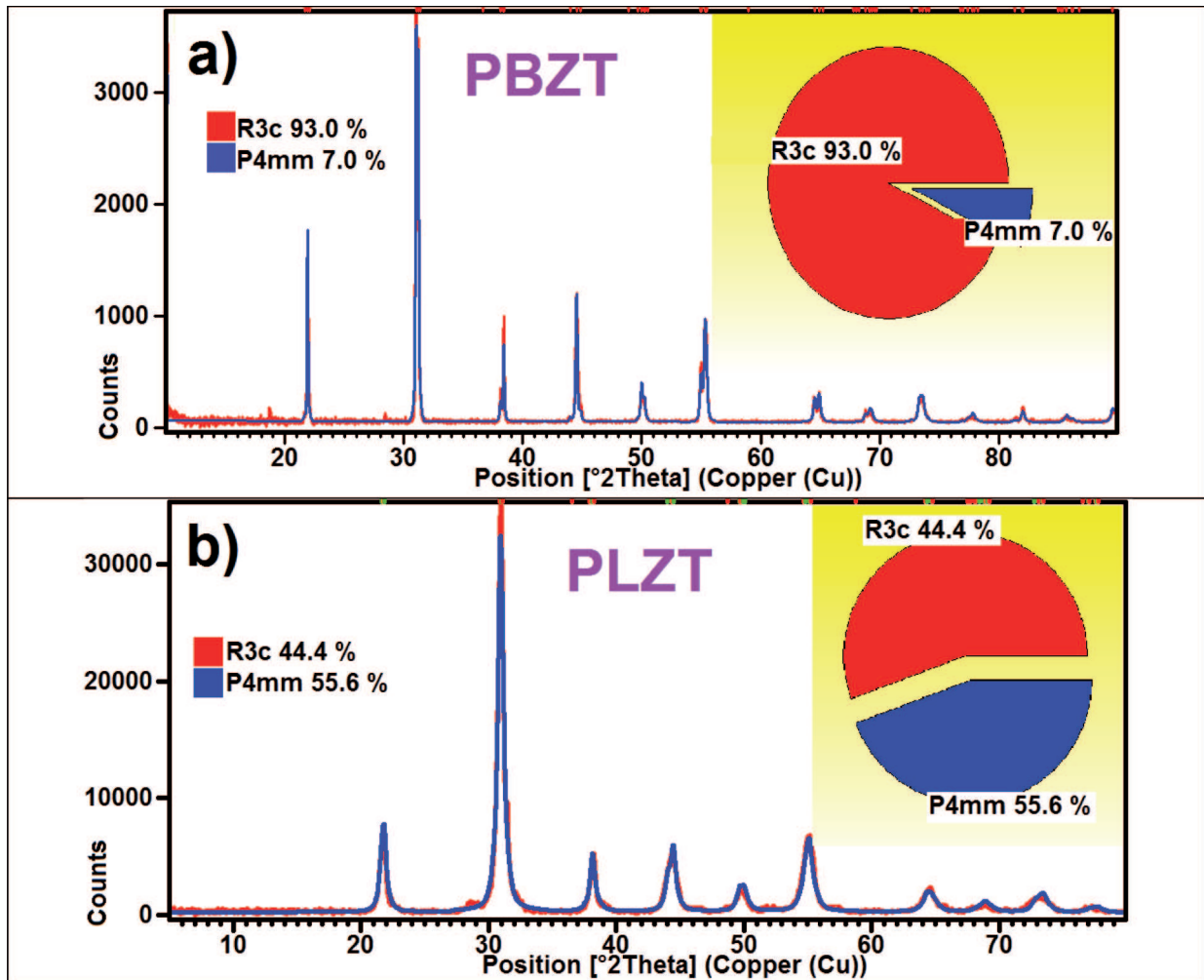


Fig. 2. XRD diffraction pattern and the phase analysis revealed coexistence of tetragonal P4mm and rhombohedral R3c space groups in PBZT and PLZT ceramics

around the morphotropic phase boundary (MPB). From the measured and calculated profiles the crystallite size was derived using PseudoVoigt function. The lattice parameters are presented in Table 1.

2.4. Dielectric properties evaluation of PBZT and PLZT samples

At room temperature, there is large differences observed for the dielectric constants of the investigated PBZT and PLZT, namely the PBZT dielectric constant overweight almost 10 times PLZT value. The samples increase in dielectric constant and dielectric loss factor

value if their temperature departs from room temperature through heating. However, the temperature coefficients of capacitance as well as Curie temperatures are different. Eventually the dielectric constants of all these samples converge to maxima at the value of 270°C (Fig. 3a) and 345°C (Fig. 3b) for PBZT and PLZT, respectively.

The results from Fig. 3 indicate that although the PLZT ceramics have much different dielectric constants at room temperature due to the difference in grain size, they seems to have similar loss tangent contributions. A close look at the dielectric loss factor at maximal temperature range showed that there is still a weak depen-

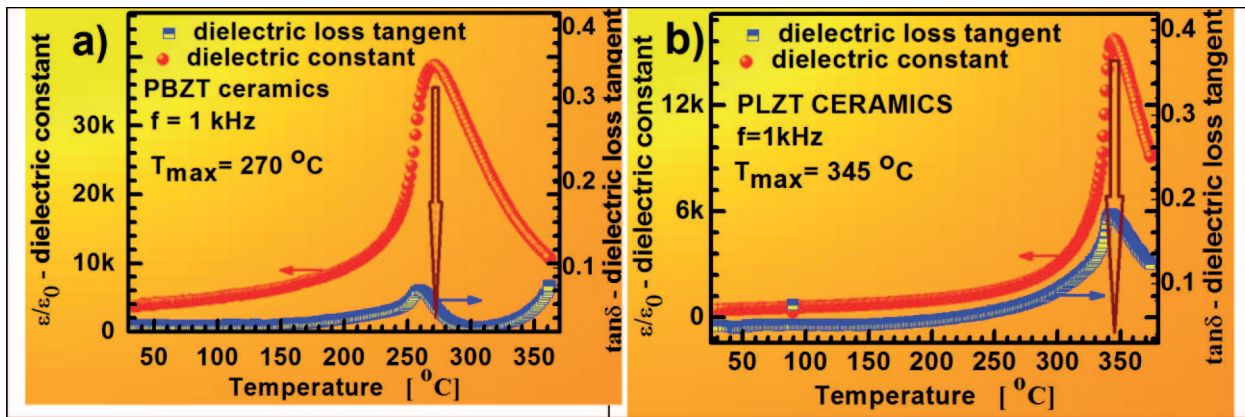


Fig. 3. The recorded values of phase transition temperature for PBZT (a) and PLZT(b)

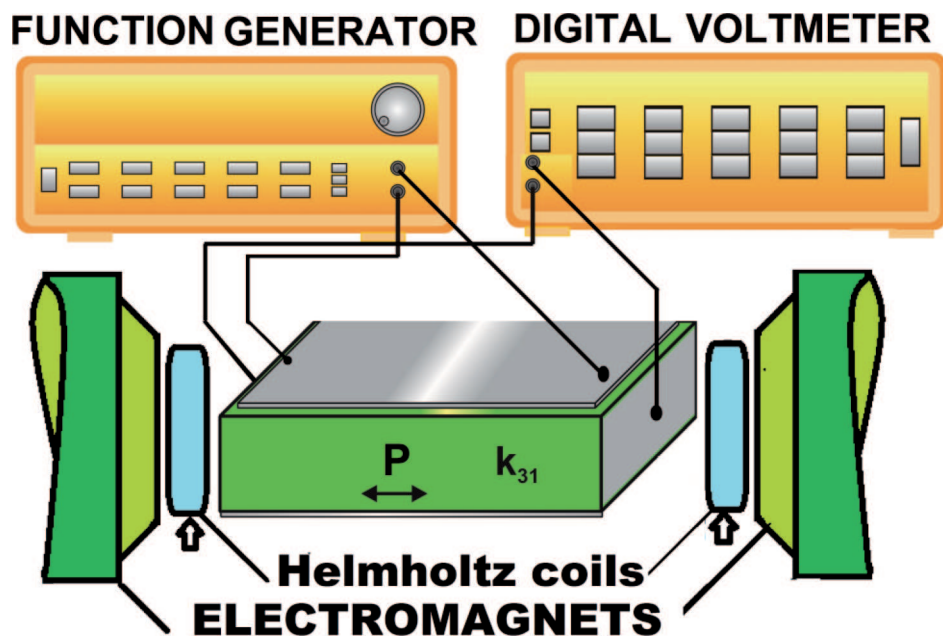


Fig. 4. PT gain measurements setup

dence of the intrinsic losses on the conductivity. This is probably due to the effect of the higher grain boundaries conductivity than internal grain one, and/or an interfacial donor electrons between the PBZT and PLZT upper and the bottom electrode.

3. Application parameters characterization and discussion

Magneto-piezoelectric Transformer (MPT) were made from our manufactured PBZT and PLZT ceramics and highly magnetostrictive ferromagnetic foil (0.2 mm) Terfenol D clamped as an upper surface (Fig. 4). The front and back surface of MPT is covered by solid silver electrode. The output voltage was measured by Agilent HP34401A multimeter, whereas the input signal was supplied by Hameg 8134 function generator.

3.1. Piezoelectric transformers gain in function of applied DC magnetic field

The novelty of proposed piezoelectric transformer lay in fact that excitation is caused by a current induced magnetic field in the Terfenol D upper layer, and the piezoelectric lower part detects the induced magnetostrictive extension. An AC current flow through a magnetostrictive layer generates a magnetic field transversal to the PT axis (Lorentz law). The current induced magnetic field vibrations are generated by magnetization vector rotation in a continuous way from the longitudinal axis or the transversal one. Following that, the magnetostrictive layer starts to vibrate under current flow accompanied field (H_{ac}). Because the layers of the bi-material are stress coupled, a vibrations is then transmitted to the piezoelectric plate. Consequently, an

output voltage is induced at the metallic electrodes of the piezoelectric plate due to direct piezoelectric effect. To summarize, this strain coupled response between an generated H and an induced E (or also vice versa) is known as the magnetoelectric effect (ME).

Figure 5 shows the ME induced gain, defined as the output to input voltage ratio, of PT working in the piezoelectric longitudinal resonance in the respect to magnetic field intensity. Presented in Fig. 5 concise gain response of the piezoelectric part transformer manufactured from PBZT and PLZT, exposed to external magnetic field, enable us to make the selection of the best performance material for this specific PT construction. The gain for PBZT based transformer is significant higher in comparison to very weak response of PLZT plate, so finally this material was proposed for further prototyping.

There are two ranges of output voltage (gain) dependence in Figure 5. The first area of proportional dependence is present in the low magnetic field range from 0 to 200 Oe. There is apparent linear relationship of the ME induced output voltage to the intensity of magnetic

field. The measured, in low amplitude of magnetic field, absolute value of output voltages reach the ratio of 0.1 in respect to the input voltage (gain).

In second region of investigations, in the higher values of magnetic fields (above 200 Oe), the dependence go into saturation and further increase in magnetic field is not reflected in gain rise, and even the decrease in the piezoelectric response is visible.

To determine piezoelectric coefficients of investigated materials, the resonance – antiresonance method was applied and the results are presented bin Table 2. The calculated values indicated that addition of Ba²⁺ ions in 16% mol in the ceramic structure increased the value of piezoelectric parameters to $g_{31} = 4.1$ pC/N and $Q_m = 454$. Finally, we demonstrate that in spite of high piezoelectric coefficients for PLZT material, only practical examination can give as a result decisive application conclusions, especially in complex applications of Piezoelectric Transformer, with double energy conversions and influence of external field.

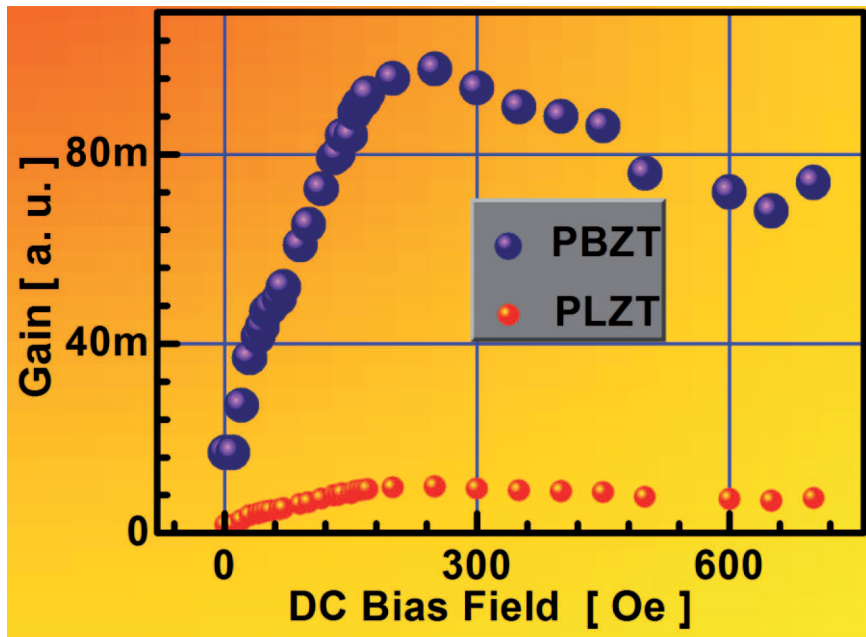


Fig. 5. PT gain dependence in the respect to magnetic field intensity

TABLE 2

Parameter	k_p	$k_{31} \times 10^{-11} [C/N]$	$d_{31} \times 10^{-12} [C/N]$	$g_{31} \times 10^{-12} [C/N]$	Q_m
PBZT	0.48	0.26	155	4.1	454
PLZT	0.27	0.15	870	0.65	650

4. Conclusions

In this work we investigated PBZT and PLZT ceramics, as a prospective materials with the best piezoelectric coefficients for specific application of piezoelectric transformer (PT) with direct magnetic feedback.

Presented magnetic field influence on transformer parameters was fundamental basis for the practical design of energy converters, in which the piezoelectric part are made from particularly efficient $(\text{Pb}_{0.84}\text{Ba}_{0.16})(\text{Zr}_{0.54}\text{Ti}_{0.46})\text{O}_3$ ceramics.

The property of linear voltage gain as a function of magnetic field to the value of 200 Oe can be effectively used in magnetic field transducers in wide frequency range from 70 kHz to 1MHz.

Acknowledgements

This work was supported by the Polish Ministry of Science and Higher Education (grant no. N N507 352635).

REFERENCES

- [1] J. Brink, D.I. Khomskii, *J. Phys. Condens. Matter* **20**, 43 (2008).
- [2] W. Eerenstein, N.D. Mathur, J.F. Scott, *Multiferroic and magnetoelectric materials*. *Nature* **442**, 759 (2006).
- [3] D.V. Efremov, J. van den Brink, D.I. Khomskii, *Nature Mater.* **3**, 853 (2004).
- [4] A.M.L. Lopes, J.P. Araujo, V.S. Amaral, J.G. Correia, Y. Tomioka, Y. Tokura, *Phys. Rev. Lett.* **100**, 155702 (2008).
- [5] Ch. Jooss, L. Wu, T. Beetz, R.F. Klie, M. Beleggia, M.A. Schofield, S. Schramm, J. Hoffmann, Y. Zhu, *Proc. Natl. Acad. Sci. U.S.A.* **104**, 13597 (2007).
- [6] N. Ikeda, H. Ohsumi, K. Ohwada, K. Ishii, T. Inami, K. Kakurai, Y. Murakami, K. Yoshii, S. Mori, Y. Horibe, H. Ki, *Nature London* **436**, 1136 (2005).
- [7] C.H. Li, F. Wang, Y. Liu, X.Q. Zhang, Z. Cheng, Y. Sun, *Phys. Rev.* **79**, 172412 (2009).
- [8] Nicola A. Hill, *Why Are There so Few Magnetic Ferroelectrics?* *J. Phys. Chem. B*, **104**, 6694 (2000).
- [9] R.A. Islam, H. Kim, S. Priya, H. Stephanou, *Piezoelectric transformer based ultrahigh sensitivity magnetic field sensor*, *App. Phys. Lett.* **89**, 152908 (2006).
- [10] S. Priya, R. Islam, S. Dong, V. Viehland, *Recent advancement in magnetoelectric particulate and laminate composites*, *J. Electroceramics* **19**, 147 (2007).
- [11] L. Kozielski, M. Adamczyk, K. Rusek, M. Pionka, *Light influence on nanomechanical characterisation of PLZT ceramics / Wplyw swiatla na charakterystyki nanomechaniczne ceramiki PLZT* *Archives of Metallurgy and Materials* **54**, 717 (2009).
- [12] C. Miclea, C. Tanasoiu, L. Amarande, C.F. Miclea, *Effect of Lead Titanate-Zirconate Additions into Barium Titanate Ceramics on the Dielectric and Piezoelectric Properties of Mixed Compounds*, *Ferroelectrics*, **319**, 57 (2005).
- [13] P. Poosanaas, K. Uchino, *Photostrictive effect in lanthanum-modified lead zirconate titanate ceramics near the morphotropic phase boundary*, *Mater. Chem. Phys.* **61**, 36 (1999).
- [14] A.K. Shukla, V.K. Agrawal, I.M.L. Das, J. Singh, D.P. Singh, K.N. Sood, *Structural, dielectric and electromechanical properties of Cr-doped PLZT close to the morphotropic phase boundary region*, *Phase Transitions* **79**, 875 (2006).
- [15] F. Xu, S. Trolier-McKinstry, W. Ren, Baomin Xu, *Domain wall motion and its contribution to the dielectric and piezoelectric properties of lead zirconate titanate films*, *J. Appl. Phys.* **89**, 1336 (2001).
- [16] T. Jungk, E. Soergel, *Contrast Mechanism for Visualization of Ferroelectric Domains with Scanning Force Microscopy*, *Ferroelectrics* **334**, 29 (2006).