J Electroceram (2012) 29:133–138
DOI 10.1007/s10832-012-9746-z

Application testing of Sr doping effect of PZT ceramics on the piezoelectric transformer gain and efficiency proposed for MEMS actuators driving

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Received: 26 November 2011 / Accepted: 6 June 2012 / Published online: 29 June 2012

Abstract Effect of Sr content in lead zirconate titanate PZT ceramic system on performance of piezoelectric transformer application were studied. Our experiments provide consistent evaluation of doping strontium amount on piezoelctric coefficients values and final voltage gain and efficiency of piezoelectric transformer. Extreme values of these parameters are preferred for MEMS device effective driving. Modification of ferroelectric materials with isovalent ions, however with the bigger radii than the original atoms, significantly affects their properties. Particularly the electromechanical coefficients of $(Pb_{1-x}Sr_x)(Zr_{0,70}Ti_{0,30})O_3$, for mole %'s as x=0.05, 0.10 and 0.15 ceramics exhibit marked increase, if the PSZT material stoichiometry is near the ferroelectric/relaxor phase boundary. To determine the piezoelectric coefficients, the resonance-antiresonance method was implemented. The results indicated that addition of Sr^{2+} ions in the amount of 5 mol% in the ceramic structure maximally increased the values of piezoelectric parameter to d_{31}=8.2 pC/N and mechanical quality factor Q_m=2902. Finally we demonstrated that in spite of the high piezoelectric coefficients for certain material only the practical examination must be applied in order to draw decisive application conclusions due to the complexity of the double energy conversion in such a specific device as the piezoelectric transformer.

Keywords MEMS · Piezoelectric Transformer · PSZT · Piezoelectric coefficients

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1 Introduction

Technical issues that accompany voltage supply of power electromechanical devices on the micro-scale MEMS [1] would be more complex without piezoelectric transformers (PT's) [2, 3], which could solve certain aspects of this problem. The available force which can be generated by the piezoelectric actuator is directly related to the magnitude of the applied voltage. Without PT in such micro power structures, for example in the micro-pumps, the practical implementation requires layered actuator construction with each layer being of the typical thickness in the range of several micrometers. The implementation of PT is proposed to simplify this technology and to gain higher driving voltage for actuator micro-technology (MEMS). Such a device must offer distinctive performance advantages, and moreover, it must be also compatible with various piezoelectric actuator system components and packaging.

Lead zirconate titanate $Pb(Zr_{1-x}Ti_x)O_3$ (PZT) perovskite material still has its leading position due to its highest piezoelectric and electromechanical coefficient values. This material constitutes one of the most important families of

Table 1 Evaluated values of PSZT sample density, grain size and unit cell dimensions

Parameter	Density [g/cm^3]	Grain size [nm]	a [10^{-10} m]	b [deg]
PSZT 5/70/30	7.6	2600	4.111±0.001	89.51±0.18
PSZT 10/70/30	7.4	3900	4.11±0.001	89.50±0.12
PSZT 15/70/30	7.3	4400	4.08±0.002	89.39±0.3

ferroelectric materials widely used for various devices, and especially for piezoelectric actuators. The optimization process of PZT-type material for certain applications is being scientifically explored due to the wide range of composi7tions and controlled ion substitutions. The role of La^{3+}, Nb^{5+} and Ba^{2+} ion substitution at the A and B site of perovskite structure was widely described in the literature [4–8]. La^{3+} is a well-known substitution on Pb^{2+} site, leading to the high-performance PLZT materials. Ba^{2+}, which has ionic radii of 1.43°A is also frequently used to substitute Pb^{2+} with an ionic radius of 1.32A°. Alternatively, certain properties of PZT were optimized by the addition of donor dopant ions, and Nb^{5+} is considered as a donor dopant for PZT materials, since it substitutes Zr^{4+}/Ti^{4+} ions.

Ions of the alkali-earth metal Sr^{2+} which has ionic radii of 1.27 °A can be used to substitute for Pb^{2+} (with an ionic radius of 1.32A°). The influence of Sr^{2+} doping on PZT properties especially attracted our attention due to its high impact on the value of electromechanical coefficients, having a high mechanical quality factor coupled with low electrical conductivity and high Curie point with excellent long term stability, which makes these ceramics prospective for PT's. The phase diagram of this system was presented by Ray N. Singh [9]. The carried out investigations concentrated mainly on the structural and piezoelectric properties of lead strontium zirconate titanate solid solution (PSZT) [10, 11]. Minor attention was paid to the investigations of the electric energy transformation. The main aim of this article is to report on the electrical properties of PSZT ceramics over the wide range of strontium content and on its influence on the piezoelectric transformer parameters. Our investigations focused on various Sr %mol doped PZT ceramics namely: $(Pb_{1-x}Sr_x)(Zr_{0,70}Ti_{0,30})O_3$, for mole %'s as x=0.05, 0.10 and 0.15 named as PSZT ceramics in the mentioned compositional range and indicated abbreviated as PSZT 5/70/30, PSZT 10/70/30 and PSZT 15/70/30.

The first question to be raised in this paper relates to the possible creation of the desirable PT performance sufficient for the integration with MEMS devices. The second enquiry, which is partly a consequence of the previous one, should consider how Sr addition influences the most important transformation parameters like gain and efficiency. And finally, the last question refers to which electromechanical parameters in practice are critical or progressively more significant for the energy transformation efficiency of the piezoelectric transformer.

2 Experimental

2.1 XRD characterization of the PSZT samples

The PSZT ceramics used in our experiment were prepared by the mixed oxide method (MOM). Assuming that the precursor stoichiometry persisted in the final ceramics, the cation ratio of Zr:Ti=70:30 was established as well as proposed Pb:Sr ratio using commercial PbO, $SrCO_3$, ZrO_2, TiO_2 reagents (Aldrich). Thermal synthesis of mixed and pressed powders was carried out at 925 °C for 2 h. Subsequently, the milled and sieved materials were pressed again into the cylindrical pellets and sintered at 1250 °C for 4 h. This procedure was repeated prior to the final sintering done at 1300 °C for 7 h. The Archimedes displacement method was applied to evaluate the sample's density and the obtained values are given in Table 1. Grain

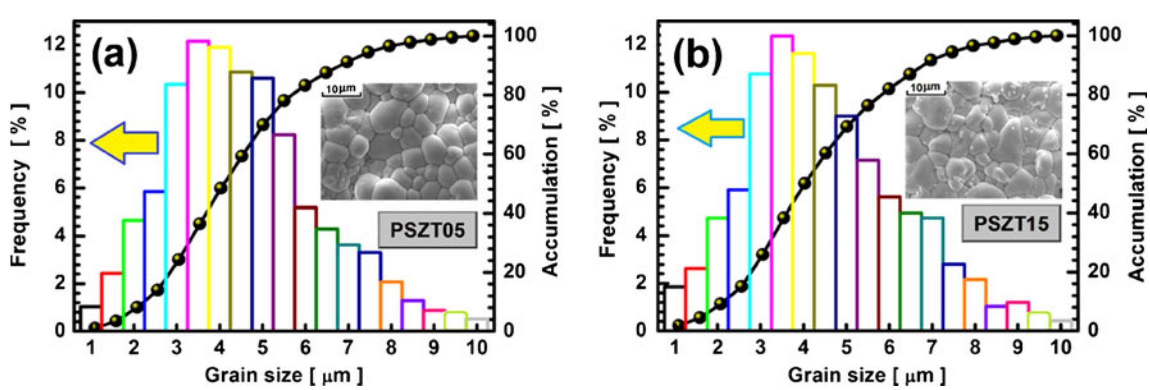

Fig. 1 Histogram plots showing the grain size distribution of PSZT 5/70/30 (**a**) and 15/70/30 (**b**) ceramics and adequate SEM images of the fractured surfaces

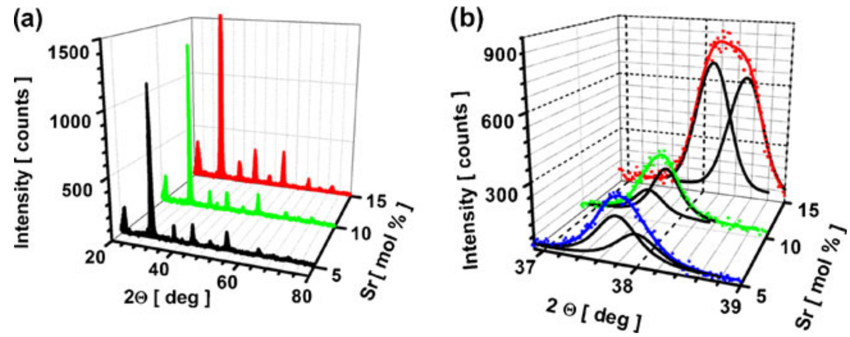

Fig. 2 XRD pattern of the sintered PSZT ceramics (**a**). The shape of chosen (111) reflections for PSZT x/70/30 ceramics for $x=5$, $x=10$ and $x=15$ mol% (**b**)

morphology and elements' distribution were examined by scanning electron microscope (SEM), JSM-5410 with energy dispersion X-ray spectrometer (EDS). The grain size measurement was performed on the ceramics fracture surface. The samples were broken in the ambient atmosphere, then covered with sputtered gold and placed in the vacuum (10^{-5} Torr) chamber of the electron microscope.

Figure 1 shows histogram plots of the grain size distribution for PSZT ceramics with (a) 5 mol% and (b) 15 mol% of strontium content and with the adequate SEM micrographs. In each case over 100 grains were analyzed with the image analysis software. The grain size was measured as the equivalent spherical projected diameter d. And as one can see in Fig. 1, the grain size spread slightly deviate from a normal distribution, and in both cases the most frequent grain size was of 3.5 μm. One can notice that PSZT ceramics prepared at the same conditions had similar grain size distributions. It is worth mentioning that the mixed oxide method (MOM) generated a wide grain size distribution, from 1 to 10 μm. Based on the experimental data in Fig. 1, the arithmetic mean grain size, d_m, was calculated using the following formula:

$$d_m = \left(\frac{1}{N}\right) \sum_{N}^{i=1} y_i d_i \qquad (1)$$

where d_i is the midpoint size for each interval, y_i the frequency of occurrence in the size interval, and N is the total number of occurrences (N is the sum of y_i over all size intervals). The arithmetic mean particle sizes, d_m, were 5.21 and 5.26 μm for PSZT ceramics with (a) 5 mol% and (b) 15 mol% of strontium content, respectively.

XRD measurements were carried out on all the samples using Huber diffractometer (Seemann–Bohlin geometry) with monochromatic CuKα_1 radiation (30 kV, 30 mA). The angular scale of the received diffraction diagrams was scaled to 2θ (Bragg–Brentano geometry) by Au standard (JCPDS number 12-0403). The X-ray diffraction patterns (XRD) of ceramics with 5, 10 and 15 mol% of strontium content obtained at room temperature are shown in Fig. 2.

The obtained diffraction diagrams are patterns for perovskite-type structure (all indexes connected with the perovskite structure were assigned). Non-perovskite reflections were not observed, which means that sintering conditions were chosen properly (Fig. 2(a)).

The observed diffraction lines are not symmetrical in varying degree and therefore they were interpreted as multiplets with unseparated components; however, the 100-th line was omitted since it shows broadening and asymmetry for apparatus reasons. The number of the components as well as split of pseudoregular reflection 111 into two components (Fig. 2(b)) and lack of the 200 reflection split both indicate that unit cell has not perpendicular and equal edges. The crystallographic structure of samples is described as regular with slight distortion. According to the results presented by Raj N. Singh [9] the unit cell is of rhombohedral symmetry. Based on separated multiplets the unit cell parameters were obtained for all ceramics with varying content of strontium (Table 1).

No conspicuous changes of the lattice constants were observed with the increasing strontium content, which is in good agreement with the previous reports [8]. It is commonly known that lead (Pb^{2+}) ions are replaced by strontium (Sr^{2+}) ions in the perovskite structure without vacancy

Table 2 Parameters value of the proposed transformers

PT	PSZT 5/70/30	PSZT 10/70/30	PSZT 15/70/30
C_0 [pF]	28	44	210
$\varepsilon_{33}^T \times 10^{-12}$	214	336	1604
f_{r1} [Hz]	213350	221406	224893
f_{r2} [Hz]	568196	589560	581966
f_{Ar1}[Hz]	216575	225718	227593
f_{Ar2} [Hz]	572451	596973	594819
V_r [m/s]	2062	2140	2096

C_0 static capacitance of PT
ε_{33}^T relative permittivity along the poling direction
f_{r1} first resonance frequency of PT
f_{r2} second anti-resonance frequency of PT
f_{Ar1} first anti-resonance frequency of PT
f_{Ar2} second anti-resonance frequency of PT
V_r the acoustic wave propagation speed of the material

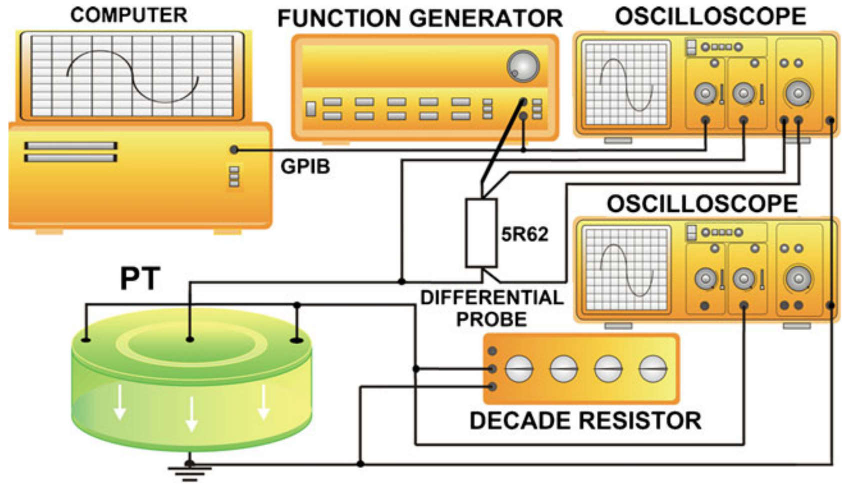

Fig. 3 Measurements setup for gain and efficiency evaluation of PSZT based PT

creation [9]. Taking into consideration the similarity of both kinds of ions no considerable changes of the lattice constants should be expected, although strontium is dissolved in the perovskite structure.

2.2 Application parameters—characterization and discussion

The measurement setup is presented in Fig. 3. PT was driven by sinusoidal voltage with 1 V amplitude generated by the function generator Hewlett-Packard HP3325A. The function generator output power is too low (mW range) and it is not sufficient in the vicinity of resonance. The power amplifier must be used in order to keep the supplied power characteristics sufficiently high for the measurements. The input and output voltage amplitudes were measured by two-channel oscilloscopes Agilent DSO3102A and DSO3202A. The load in the output part (represented by pure resistor decade) varied from 10 Ω to 100 kΩ. The current in input circuit was measured by differential probe Agilent N2772A connected to oscilloscope as a voltage drop across the small resistor (5.62Ω). The samples were finally polarized at 2 kV/mm DC electric field (E_p) for 10 min, at the temperature T_p=373 K. The characteristic parameters of manufactured piezoelectric transformers are presented in Table 2.

The variation of the characteristic parameters of fabricated piezoelectric transformers with Sr concentration at room temperature is referred in Table 2. The static capacitance as well as the relative permittivity rose distinctly together with increasing Sr content for the investigated ceramics, whereas such tendency is not observed in the speed of acoustic wave, which is the highest at 10 mol% of Sr dopant. High values of permittivity are typical for the ferroelectric soft ceramics, therefore we can predicted lower values of the electromechanical coefficients k_p for PSZT 15/70/30 ceramics. Finally, we recorded the smallest differences of resonant and anti-resonant frequencies for 10 mol% of Sr in PSZT ceramics, which can suggest the highest piezoelectric coefficient for this particular ceramics.

The effect of material properties on the PT voltage gain and efficiency was obtained for 1 kΩ load. The recorded curves exhibit maxima at the fundamental resonance frequency, which is the characteristic property of piezoelectric transformer as a resonant device. It is visible in Fig. 4, that the frequency of these maxima is strongly dependent on Sr mol% content and it shifts to lower values with increasing dopant amount.

As it is clearly seen, the highest gain value from the measured PT's is for PSZT 5/70/30 material device and is equal 1.5 (Fig. 4(a)), whereas gain of PSZT 10/70/30 and 15/70/30 PT's are at the level of 1. In the light of this

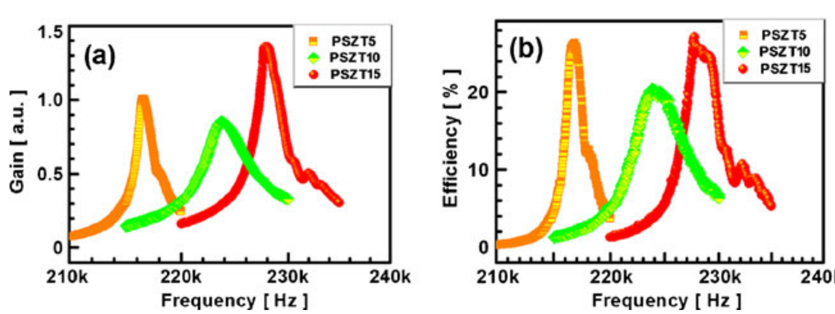

Fig. 4 Sr doping influence on PT gain (**a**) and efficiency (**b**) made from PSZT ceramics

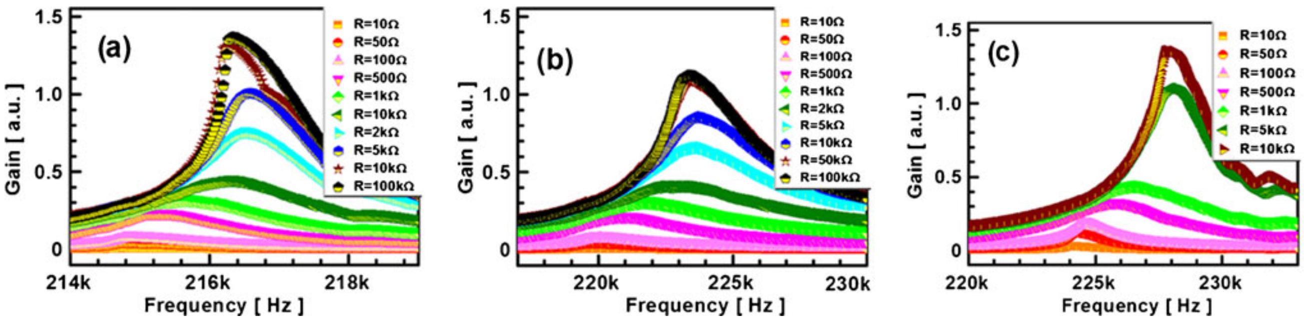

Fig. 5 Gain characteristics of increasing Sr mol% content in PSZT x/70/30 ceramics with different load value for $x=5$ (**a**), $x=10$ (**b**) and $x=15$ (**c**)

measurement we emphasize the fact that Sr doping can significantly change the PT's output voltage.

The efficiency versus frequency characteristics are shown in Fig. 4(b). The recorded efficiency maxima do not differ significantly for PT made from PSZT 5/70/30 and PSZT 15/70/30 ceramics but have 30 % higher value in comparison with PSZT 10/70/30 (Fig. 4(b)). These results make a case for the experimental examining significance of piezoelectric transformers due to the fact that gain characteristics reach the peak value not for the same Sr dopant content as efficiency. Consequently, the optimal doping strategy must reach compromise between what gain value we would like to achieve and how high efficiency level should be.

2.3 Piezoelectric PSZT transformer gain and efficiency versus load phenomena

In order to confirm the above mentioned phenomenon in detail we carried out the measurements of gain and efficiency as a function of load. The obtained results are presented in Figs. 5 and 6.

The obtained gain results are presented in Fig. 5 (a, b and c) in the reference to Sr content ranging from 5 to 15 mol% in PSZT ceramics, respectively. The significant changes in gain appeared in the discussed dependences, in the wide load range. Therefore, it allows us to conclude that increasing gain value follows the increase of load in the vicinity of the mentioned frequency.

Our investigation lends support to the view that efficiency as a function of the resistive load reaches the top values for PSZT samples at different values to the gain (Fig. 6 (a, b and c)). The optimum resistance value could be properly resolved using efficiency measurement or modeled analytically. The maximum efficiency loads are 2 kΩ for 5 mol%; 1 kΩ for 10 mol% and 0.5 kΩ for 15 mol% of Sr content, respectively.

2.4 Piezoelectric parameters evaluation for PSZT transformers

To complete the results obtained above, i.e. the energy transfer phenomena and correlation of Sr content to the optimum PT performance, the evaluation of electromechanical parameters was carried out. To determine piezoelectric coefficients, the resonance- antiresonance method was implemented using impedance and phase measurement PSZT ceramics [12]. The impedance frequency spectrum was measured on Agilent 4294A impedance analyzer in the frequency range from 100 kHz to 1 MHz.

Figure 7 (a, b and c) show the complex impedance spectra of PSZT ceramics measured in the fundamental resonance frequency range. The change in Sr content gives

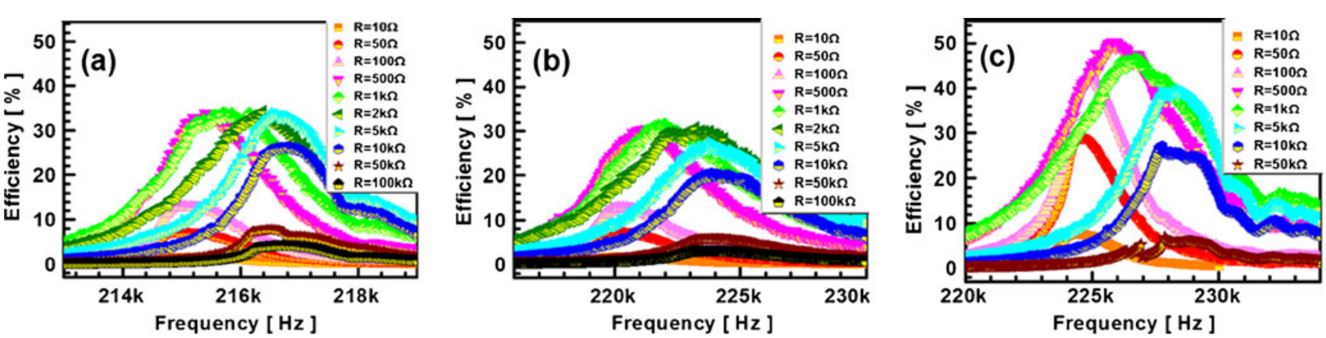

Fig. 6 Efficiency characteristics of increasing Sr mol% content in PSZT x/70/30 ceramics influence with different load value for $x=5$ (**a**), $x=10$ (**b**) and $x=15$ (**c**)

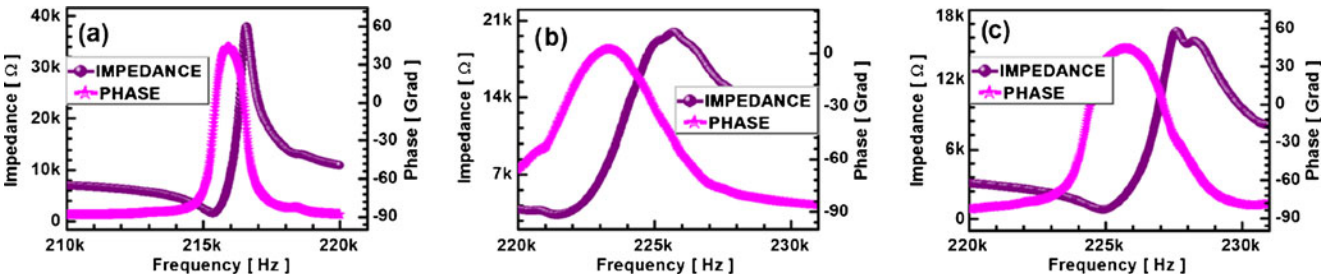

Fig. 7 Fundamental resonant characteristics of increasing Sr mol % content in PSZT x/70/30 ceramics influence for $x=5$ (**a**), $x=10$ (**b**) and $x=15$ (**c**)

a significant effect on the impedance spectrum. At higher Sr content impedance curve consists of wide maxima and shows a small change in the phase value, whereas the 5 mol% Sr doped PSZT ceramics exhibit sharp, narrow amplitude and phase peak.

The calculated piezoelectric parameter values are presented in Table 3. The other calculated parameters do not exhibit the same tendency. Mechanical quality factor Q_m drops with increasing Sr content contrary to the piezoelectric coefficient d_{31} with steady increase at the same time. However the electromechanical coupling factors k_p and k_{31} fluctuate regardless of Sr content.

3 Conclusions

The presented measurements reveal complex picture of the energy conversion phenomena in the PSZT ceramics used for piezoelectric transformer construction:

1. As it is seen from the gain characteristics we can slightly tune PT's resonant frequency by Sr dopant content. This fact makes the application of piezoelectric transformers into MEMS or PCB circuits more feasible, where the matching of the resonance frequency by the element's dimension is very restricted or even impossible.
2. Contrary to the previous expectations we have to admit that although the mechanical quality factor ensures the best gain value for 5 mol% of Sr content, the best transformation efficiency is not met at the same time. Particularly, the PSZT ceramics with 15 mol% of Sr content appear to have the best transformation efficiency. Therefore, piezoelectric coefficient d_{31} seems to be the key factor here.
3. As a final remark drawn from our investigations we demonstrate that PT efficiency material dependence is more complex. Only the practical examination of PT parameters can be effectively used for the application conclusions.

Table 3 The evaluated piezoelectric coefficient values of PSZT ceramics

Parameter	k_p [-]	k_{31} [-]	d_{31} [10^{-12}C/N]	g_{31} [VmN^{-1}]	Q_m [-]
PSZT 5/70/30	0.12	0.07	8.2	0.038	2901
PSZT 10/70/30	0.22	0.13	16.4	0.029	252
PSZT 15/70/30	0.17	0.10	56.3	0.021	344

Acknowledgement The work presented was performed within the grant NN507 352635 supported by Polish Ministry of Science and Higher Education. One of the authors (J.E.) would like to acknowledge support granted by the Grant Agency of the Czech Republic under the project No. GAČR P102/10/1139.

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