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Model Explanation of Electrical Characteristics Anomalies in PLZT Graded Structure for Piezoelectric Transformers

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Abstract

In this paper, the influence of one directional diffusion between the two components in functionally graded structure (FGS) on the impedance (dielectric) spectrum of the system was investigated. Lanthanum-modified lead zirconate titanate (PLZT) ceramic materials have gained considerable attention due to their photostriction which is the superposition of photovoltaic and piezoelectric effects. The idea of functionally graded materials (FGM) implemented in the construction of piezoelectric transformer (PT) can be used for direct converting photonic energy to electrical one by implementing photostrictive actuators with a piezoelectric generator in one graded structure of PT. The presented measurements revealed complexity of the integration process of functionally graded materials and high electrical anisotropy of this graded structure. Impedance spectroscopy (IS) proved to be the method capable to present such an inequality in the form of well separated semicircles. Additionally, it was demonstrated that the characteristics of the graded structure is not a simply addition result of starting materials parameters but is deeply influenced predominantly by a diffusion direction. Finally, the influence of the directional diffusion process on impedance spectra was explained by RC electrical model and confirmed experimentally by XPS method.
Keywords: Piezoelectric transformer, PLZT, PBZT, FGM, Diffusion

MODELOWE WYJAŚNIENIE ANOMALII CHARAKTERYSTYKI ELEKTRYCZNEJ W GRADIENTOWEJ STRUKTURZE PLZT PRZEZNACZONEJ NA TRANSFORMATORY PIEZOELEKTRYCZNE

W artykule zbadano wpływ jednokierunkowej dyfuzji pomiędzy dwoma składnikami struktury funkcjonalnie stopniowanej (FGS) na jej widmo impedancyjne (dielektryczne). Modyfikowana lantanem ceramika cyrkonianu tytanianu ołowiu (PLZT) zyskała znaczne zainteresowanie w związku z jej fotostrykcją, która jest superpozycją efektów fotowoltaicznego i piezoelektrycznego. Idea materiałów funkcjonalnie stopniowanego (FGM) zaimplementowana do transformatora piezoelektrycznego (PT) może być wykorzystana do bezpośredniej zamiany energii fotonicznej na elektryczną poprzez wykorzystanie fotostrykcyjnych oscylatorów wzbudzających z generatorem piezoelektrycznym w jednostopniowej strukturze PT. Przedstawione badania ujawniły złożoność procesu integracji materiałów funkcjonalnie stopniowanych i ich wysoką anizotropię elektryczną. Spektroskopia impedancyjna (IS) sprawdziła sie jako metoda zdolna do pokazania takiej nierówności w postaci dobrze rozdzielonych półokręgów. Dodatkowo zademonstrowano, że charakterystyka struktury stopniowanej nie jest wynikiem prostego sumowania właściwości materiałów wyjściowych, ale jest głównie mocno zależna od kierunku dyfuzji. Na koniec, wyjaśniono wpływ procesu dyfuzji kierunkowej na widma impedancyjne za pomocą elektrycznego modelu RC i potwierdzono doświadczalnie metodą XPS.
Słowa kluczowe: transformator piezoelektryczny, PLZT, PBZT, FGM, dyfuzja

1. Introduction

The electric properties of multi-component materials are sensitive to changes in the quantitative composition of particular components [1-4]. Compositionally graded systems show certain properties such as polarization offset which can be developed by asymmetrical leakage currents, originated by point defects and concentration-dependent band structure [5-7]. Diffusion (considered in this paper as only a migration of particles in a sample) between two layers can be seen as the origin of changes in the percentage composition of neighbouring layers which distinctly alter the final characteristics. In this paper the influence of predominant direction diffusion and its magnitude between two components of functionally graded material on the impedance spectrum of the system was investigated. A subtle inequality in the sintering temperature of one material in the FGM-type structure has a significant influence on the predominant direction of diffusion. Consequently, we do not have materials of resultant properties that we had them wished to be. Impedance spectroscopy (IS) appeared to be an efficient method for detecting such an inequality due to specific shape changes in a real and imaginary part of impedance spectra. For this reason, this easy to use tool can be recommended as a method that can determine the usefulness of final FGM material for a particular application.

The electrical energy conversion in the piezoelectric transformer (PT) has potentially higher efficiency level than the traditional electromagnetic construction, since the energy stored by the elastic vibration is larger. In particular, the piezoelectric system presents a much higher value of power density coefficient, so that, it is better suited to compact applications. An additional merit of special importance is the fact that this environment friendly construction does not generate electromagnetic noise because it has no windings. It is worth adding finally that this electro-ceramic structure is non-flammable and safe in the case of potential short-circuiting of the output terminal [8].

The piezoelectric transformer efficiency increasing techniques are correlated with many strategies. At first position, there are theoretical calculations of resonator shape, geometry and resonant modes formulated the strategy of optimal controlling of the PT [9-11]. The second technological direction of improving PT performance is connected with development of advanced nano-powder technologies supported by various rare earth elements doping variations [12].

The last method is combined with domain engineering and design of multilayer or functionally graded structures (FGM). The optimal energy conversion of these constructions is achievable by the application of the highest performance materials into actuator and transducer parts of PT [13-15].

This successful construction is widely used for energy transformation. We additionally implemented a photo-actuator into the input part of piezoelectric transformer for photovoltaic energy harvesting. Such a photo-driven PT was constructed by introducing PLZT in functionally graded material configuration, because this particular ceramics exhibit large photostriction under illumination [16].

Subtle changes in the juncture part of FGM structure have a significant influence on conductivity and hence on the usefulness of material for the particular application. From many material testing methods we have proposed impedance spectroscopy as an efficient tool capable to estimate properly its electrical conductivity as well as grain to grain boundaries correlation and conclude on structural properties.

Additionally, impedance was found to increase due to fatigue and the initiation of microcracks and for this reason it can be also proposed for prior mechanical quality testing of this juncture part between two different materials in FGM structure [17].

2. Experimental

For clarification directional diffusion process influence on the impedance spectra we used the model proposed by Han and Choi [18, 19]. In order to reflect materials (systems) with different conductivity properties, only two values of R are assumed - 10^3 Ω (material A – a black circle) and 10^9 Ω (material B – a grey circle), whereas the value of capacitance is the same and equal C = InF. This very simple model is built with two parallel RC circuits connected in a series and this particular small difference changes the symmetry of Cole-Cole relaxation peaks. Thus, we can simulate three kinds of interaction: (i) material A – material A, (ii) material A – material B, and (iii) material B – material B. Each RC section has a relaxation time τ calculated as a product of resistance R and capacitance C. So, a particular RC relaxor has $\tau = 10^{-6}$ s and τ = 1 s in material A and material B, respectively. Each "molecule" consists of six such RC relaxors connected in one node. The simulation process was carried out on a system consisting of 246 such "molecules" using PSPICE software.

In the present work the gradation step of PLZT (3/52/48) and PBZT (16/54/46) (Fig. 1) the solid solution was manufactured using custom-designed steel die matrix to achieve 10 mm in diameters and 1 mm thick disk. The preparation method of graded structure is described in detail in [15].

The major obstacle to the efficient impedance measurement of functionally graded structures is caused by difficulties with the preparation of defect and crack-free joint part between the PLZT and PBZT materials. For testing the mechanical quality of this part we used nanoindentation technique for measurements of mechanical parameters along the radius of obtained disks. The nanoindentation tests were performed at constant room temperature with the TriboScope® nanoindenter (Hysitron Inc., Minneapolis, Minnesota). AC impedance spectroscopy for temperatures up to T = 900 K were carried out using Solartron 1260 system in the frequency range ν = 0.1 Hz - 2 MHz.

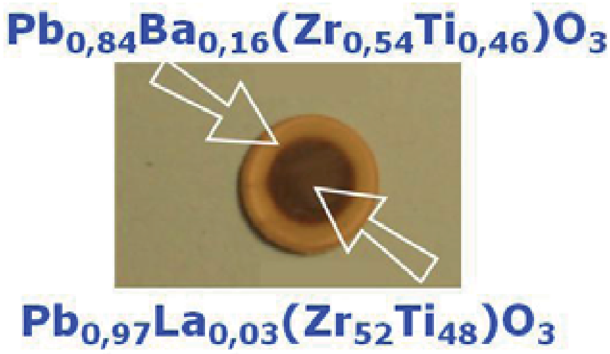

Fig. 1. Graded PLZT/PBZT ceramics.

3. Results and discussion

3.1. Mechanical quality testing

The main problem in the technology of FGM materials in which the structural gradient changes in a step way is connected with the preparation of defect and crack-free joint part between the two different materials. Consistency test of this fundamental part were conducted by nanoindentation technique, which allowed us to inspect the values of hardness and elastic modulus along the radius of obtained disks. The final quality proof was carried out by the mapping of hardness and elastic modulus distribution on the sample surface. From the uniform shape of hardness and elastic modulus distribution line in the juncture part of graded structure (Fig. 1), we can indicate the lack of cracks and accumulation of point defects in this interpart area. The values of mechanical parameters as a function of position along the radius of the PLZT/PBZT disk are presented in Fig. 2. The hardness value changes from H = 8 GPa to H = 8.2 GPa along the radius (2 %) and the Young Modulus values increase from the E = 117 GPa to E = 119 GPa (1 %). The results of these measurements

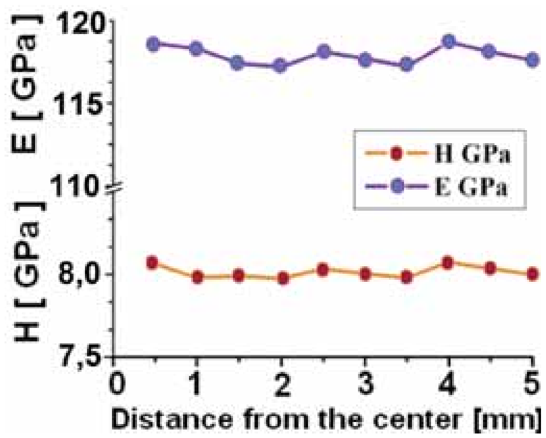

Fig. 2. Hardness and elastic modulus as a function of position along the radius of the graded structure PLZT/PBZT.

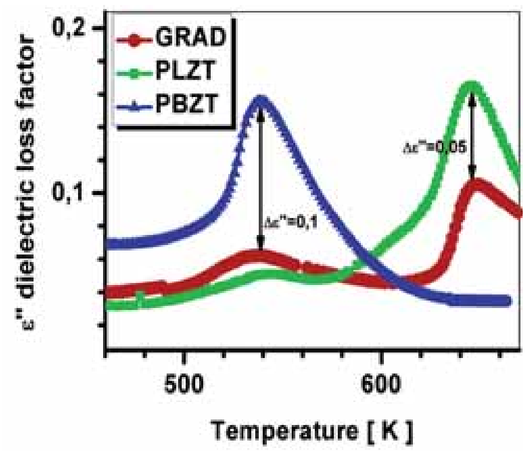

Fig. 3. The temperature dependency of dielectric loss factor (tanδ) at frequency f = 1 kHz for the PBZT, PLZT and PLZT/PBZT ceramics.

distinctly prove good mechanical quality of our final functionally graded structure.

3.2. Dielectric measurements

The graded structure PLZT/PBZT, and separate PBZT and PLZT samples were used to measure the dielectric loss factor (ε'') as a function of temperature. The determined temperature dependence of the dielectric loss factor (ε''), with an f = 1 kHz frequency of the measured signal, it is presented in Fig. 3. From the dielectric charts of the graded structure it is visible that the concise characteristic is not a direct sum of starting materials parameters. We have found that in FGM structures ε''(T) curves reveal anomalies in the vicinity of temperature corresponding to the tetragonal-cubic (FT-PC) phase transition. We have found that the maximal value of loss factor in graded PLZT/PZT material did not appear at the same temperatures as in the separated PLZT and PBZT materials. The values of Curie temperature were slightly shifted to the lower level in comparison to the separated materials. These shifts were equal ΔT = 8 K in the PBZT material and ΔT = 7 K in PLZT. The maximal loss factor value in graded PLZT/PZT material decreased in comparison to the separated PLZT and PBZT materials. The values were shifted to the lower level, and these shifts were equal Δε'' = 0.1 in PBZT material and Δε'' = 0.05 in PLZT. In our opinion, the lower values of low frequency dielectric loss factors is the result of a decreasing number of ion and electron mobility paths in the diffused juncture part between the two materials.

3.3. Impedance spectroscopy

From the results of AC impedance measurement we indicated distinctly resolved two semicircles for the graded structure PLZT/PBZT in the *Z'* and *Z''* plots (Fig. 4c), whereas in the separated materials these plots were almost coherent (Figs. 4a and 4b). The presence of two semicircular arcs is resulting from cascading effect of the parallel combination of resistive and capacitive elements (RC), arising from the grain impedance of PBZT and PLZT material. The high frequency semicircles were associated with PBZT participation while the second semicircle, at low frequency, indicates the electric contribution of PLZT ceramics to the low frequency semicircle. Impedance spectra were analyzed by means of

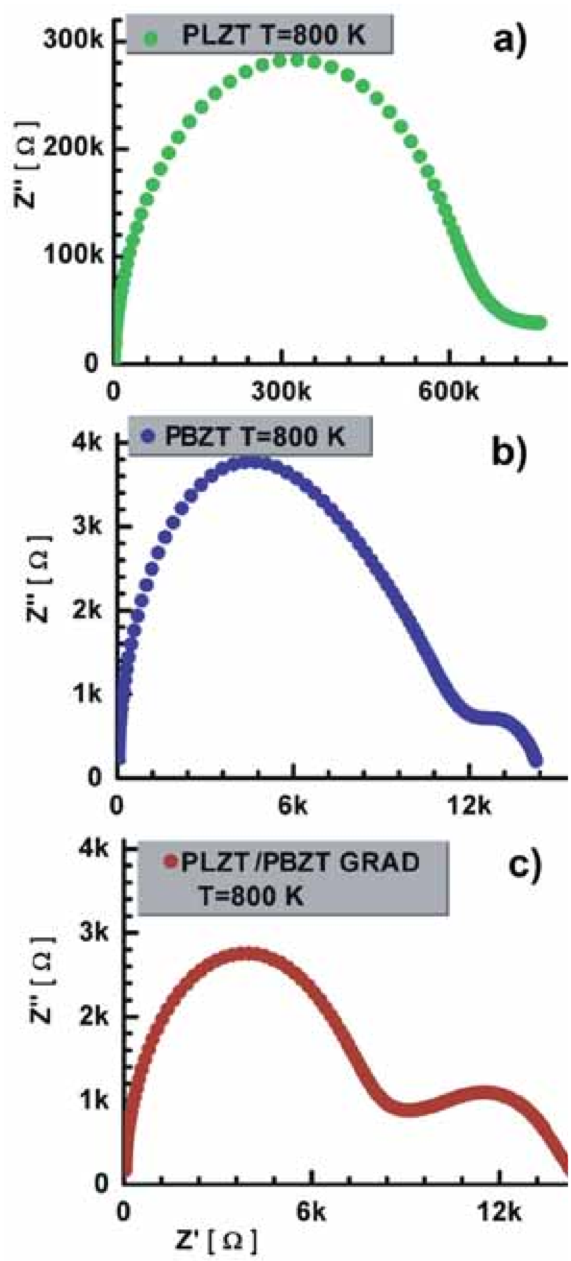

Fig. 4. Complex impedance plot (Nyquist plot) at temperature T = 800 K: a) PLZT, b) PBZT, and c) functionally graded PLZT/PBZT ceramics.

non-linear least-squares fitting of equivalent circuits (CNLS-analysis). Our analysis of these plots was conducted to evaluate the value of grain conductivity for the discussed samples. The variation of grain resistance for PBZT at temperature T = 800 K changes from R_g = 6.2 kΩ to R_g = 3.7 kΩ, whereas for PLZT from R_g = 610 kΩ to R_g = 2.1 kΩ. We observed that the summary characteristics of the graded structure is not a simply addition result of starting materials performance, especially for low frequency range. We proposed a hypothesis that this fact is deeply influenced by a predominantly diffusion direction. We assumed that during the high temperature sintering process ion diffusion from one material predominated the second ion movement resulting in a resistance domination of one particular material in the final graded structure. To explain this phenomenon, we developed an electrical model consisting of over 2500 electrical analogs of separated PLZT and PBZT grains impedance.

3.4. Computer simulations

The influence of diffusion in a two-component system with the use of computer simulations was analyzed. The diffusion was regarded as mixing of neighbouring phases without paying attention to a detailed description of mechanism of this process. We considered an arrangement in which a layer of material A (20 % of whole system) was sandwiched between two layers of material B (Fig. 5a). In the starting position there is no diffusion and only one relaxation process is observed which can be seen as one symmetric semicircle in the impedance plot. In the above mentioned situation only one absorption peak is visible although there are two materials with different relaxation times. Probably it is due to the fact that for the sample in the starting position each layer can be treated as a separate homogeneous system with Debye's response. Considering the middle layer, dispersion in the virtual sample can be observed. However, this model has one vital limitation, namely in the case of a homogeneous dielectric sample (all resistors and capacitors have the same value) neither the absorption peak nor dispersion can be observed though they are characteristic of the materials with relaxation processes [20]. Owing to this model in the case of a homogeneous virtual sample the materials which only possess a steady *dc* conductance due to ion-motions can be reflected [20].

In Figs. 5d and 5e, the 5 and 10 steps of one-way diffusion between both layers are shown. The penetration of first phase enables us to observe a distinct asymmetric extension of a semicircle in the Cole-Cole diagram (Fig. 6a). So on the side of a low-frequency range another relaxation process with lower amplitude appears but the main process coming from the whole virtual sample is considerably dominated.

In Figs. 5b and 5c, the 5 and 10 steps of two-way diffusion between neighbouring layers towards electrodes are presented. An increase in the magnitude of diffusion (Fig. 6b) results in the strong domination of a new process over the main one.

In Fig. 6c, a comparison between the 10th step diffusion in two directions and the 5th step in one direction is presented. It is visible how the penetration of layers influences the appearance of a second semicircle. Not only the direction, but also the magnitude of the infiltration process is associated

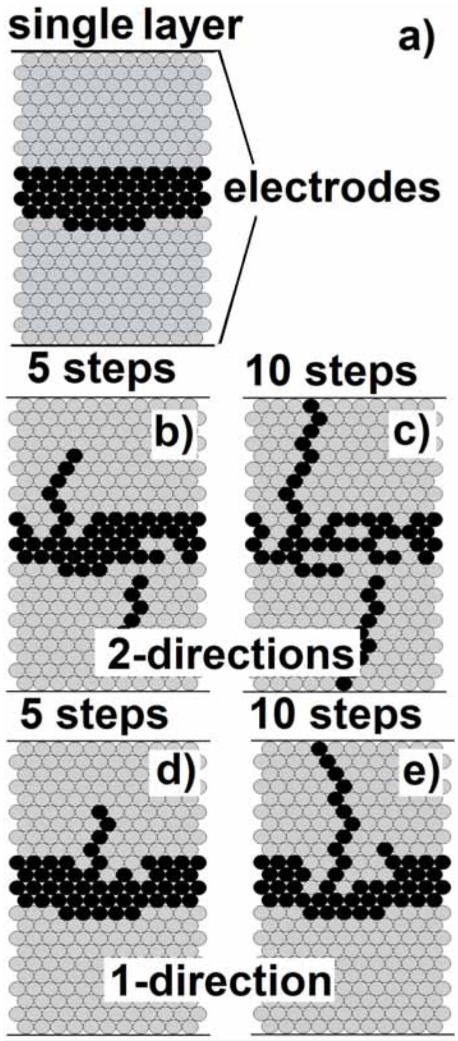

Fig. 5. Schematic diagram of diffusion process model: a) the starting position for further simulations , b) and c) 5 and 10 steps of diffusion in two directions,respectively, d) and e) 5 and 10 steps of diffusion in one direction, respectively.

with the separation of semicircles. The direction of diffusion seems to be responsible mainly for the radius of a particular semicircle, especially the one way penetration gives an effective separation of the second semicircle and the deeper the penetration, the more separated the semicircles are.

3.5. XPS measurements

To determine how the chemical c omposition varies as a function of position, the line of barium Ba3d5 was chosen as a marker. The basic arguments supporting such a choice was as follows: barium is a component of only one ceramics (PBZT), therefore the intensity of the spectral line corresponding to Ba3d5 enabled us to determine the distance of diffusion of PBZT into PLZT area and to find out the type of the gradient (continuous or stepwise). The results of measurements carried out for Ba3d5 spectral line are given in Fig. 7. One can see in Fig. 7 that maximum value of Ba3d5 line intensity recorded at the measuring points situated at the distance of 0.25 mm from each other, decreases from 52×10^3 counts/s (point 1; Fig. 7b), via 41×10^3 counts/s (point 2) to 37.8×10^3 counts/s (point 3) (Fig. 7a). The 2 mm distance of the observed change in barium line intensity can be described

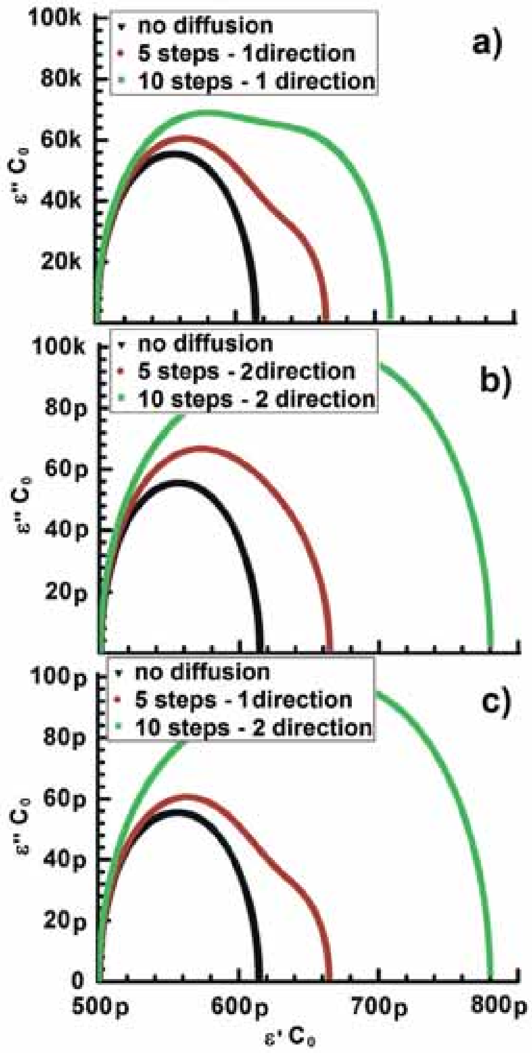

Fig. 6. Results of simulations data on symmetry of relaxation peaks for one and two way diffusion direction.

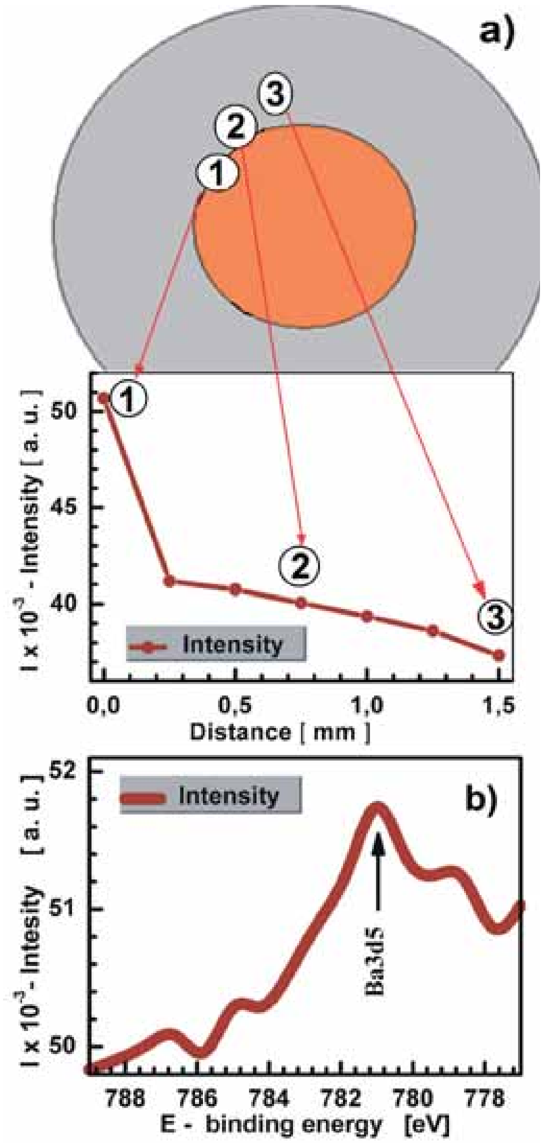

Fig. 7. The measured by XPS method chemical composition variation as a function of position. The line of barium Ba3d5 intensity measured along the distance from the middle of juncture part between the PLZT and PBZT materials (a) among the chosen as a marker points (1;2;3). Barium energy spectrum of XPS measurement at the point 1 (b).

as a continuous gradient type which was created during FGM manufacturing due to one directional diffusion process.

4. Conclusions

We revealed by our measurements that the high electrical anisotropy of PLZT/PBZT functionally graded structure is related to the diffusion direction. A simple electrical model was successfully used to simulate the influence of diffusion on the FGM sample properties. It was confirmed that the impedance spectroscopy is an easy to use method, capable to reveal the level of such an inequality in the form of well separated semicircles.

Additionally, we explained using electric measurement and values of grain conductivity, the complexity of the integration process of functionally graded structures. Finally, XPS method proved that the juncture part between two materials exhibited a continuous position-dependent chemical composition gradient.

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