A R C H I V E S O F M E T A L L U R G Y A N D M A T E R I A L S
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ELECTRICAL AND MECHANICAL EXAMINATION OF PLZT GRADED STRUCTURE FOR PHOTOVOLTAIC DRIVEN PIEZOELECTRIC TRANSFORMERS

ELEKTRYCZNE I MECHANICZNE CHARAKTERYSTYKI STRUKTURY PLZT DLA TRANSFORMATORÓW PIEZOELEKTRYCZNYCH ZE SPRZĘŻENIEM OPTYCZNYM

Lanthanum-modifed lead zirconate titanate (PLZT) ceramic materials have gained considerable attention due to their photostriction, which is the superposition of photovoltaic and piezoelectric effects. Idea of this photo effect implemented in construction of Piezoelectric Transformer (PT) can be used for direct converting photonic energy to electrical one by implementing photostrictive actuators with piezoelectric generator in one structure of piezoelectric transformer.
Possible application in electronic industry needs thoroughly electrical and mechanical characterisation of this new graded structure constructed from the PLZT/PZT material. We revealed by our measurements high electrical anisotropy of this graded structure and the Impedance Spectroscopy (IS) proved to be method capable present such inequality in form of well separated semicircles. Additionally for quality testing of the both materials integration we used nano-mechanical methods. Finally our dielectric measurement demonstrated complexity of the integration process because the summary characteristics of the graded structure is not a simply addition result of starting materials parameters but are deeply influenced by a predominant diffusion direction.
Keywords: piezoelectric transformer, functionally graded material, impedance spectroscopy

Roztwór stały cyrkonaniu ołowiu modyfikowany lantanem znajduje szereg zastosowań praktycznych wykorzystujących efekt fotostrykcji, który jest połączeniem efektu fotoelektrycznego z piezoelektrycznym. W przypadku transformatora piezo-elektrycznego nałożenie tych dwóch efektów może zostać wykorzystane do sterowania wielkością napięcia wyjściowego przy pomocy zmiany natężenia oświetlenia. Zjawisko fotostrykcji wpływa w takim przypadku na zmianę sumarycznych wielkości współczynników piezoelektrycznych wejściowej części tego transformatora. Ta część transformatora jest wykonana z materiału fotostrykcyjnego podczas gdy, druga generatorowa część, z typowej ceramiki piezoelektrycznej PZT. Te dwie ceramiki zostały połączone w jedną strukturę w procesie spiekania, na wskutek procesu dyfuzji, tworząc monolityczny Funkcjonalny Materiał Gradientowy (FGM).

Potencjalne aplikacje w zasilaczach z regulowaną przy pomocy światła wartością napięcia wyjściowego, wymagają charakterystyki właściwości elektrycznych i mechanicznych tej nowej struktury. W szczególności Spektroskopia Impedancyjna (IS) jest tu przydatną metodą badawczą ponieważ pozwala na separację i analizę wpływu poszczególnych składowych materiałów z osobna w postaci rozsuniętych półkoli. Zostały także przeprowadzone mechaniczne testy jakościowe gradientowej struktury łączącej oba materiały PLZT i PZT. W końcowym etapie przeprowadzono pomiary dielektryczne, które potwierdzają złożoność procesów integracyjnych takich struktur gradientowych ponieważ finalne charakterystyki nie są wypadkowym złożeniem przebiegów dla składowych materiałów.

1. Introduction

The Piezoelectric Transformer (PT) can be miniaturized to a greater degree than the electromagnetic construction since the energy stored by the elastic vibration is larger. PT boost up or down conversion is possible at efficiency levels not possible to achieve in traditional constructions. Resonant electromagnetic high-voltage transformers have an electrical Q_E factor of 20 to 30 whereas this equivalent for the piezoelectric transformer is its mechanical quality factor Q_M, which exceeds 2000. In particular, the piezoelectric system presents much higher power density, that makes it better suited to compact applications and additionally this environment friendly construction doesn't generate electromagnetic noise (no windings). It is worth to add that, piezoelectric

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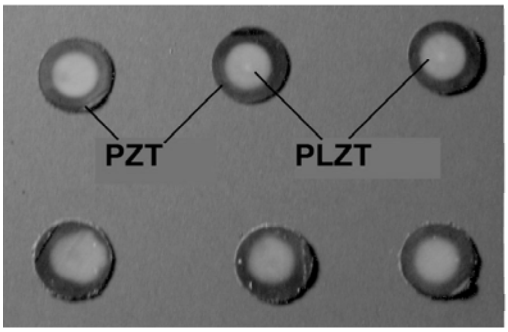
structure is non-flammable and safe in case of potential short-circuiting of the output terminal[1,2].

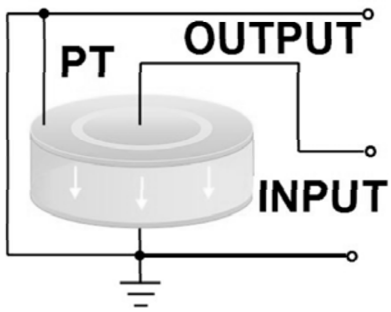
There are many strategies of increasing energy transformation efficiency in piezoelectric transformers. The main part of investigations is directed towards theoretical calculation of resonator shape, geometry and resonant modes [3,4]. The second direction of improving PT performance is connected with material science development and particularly with nano-powder technologies improved by various rare earth elements doping or solid solutions variation[5].

The last method is connected with advanced design of graded or multilayer structures. The optimal energy conversion of these constructions is possible by application the highest performance materials both in actuator and transducer part of PT [6-8].

However, this device is mainly used for energy transformation we proposed additional application of it for photovoltaic energy harvesting and conditioning in the same device. Our new construction is assembled from two parts. The input part is made from photostrictive material, exhibiting light induced strain (photo-actuator). Good candidate for such material for input part of PT is be (Pb, La)(Zr, Ti)O_3 (PLZT) ceramics, which exhibit large photostriction under illumination (bright part of the disk presented in Fig. 1. a) [9,10]. The output part of our device is made from typical PZT hard ceramics that is usually used in piezoelectric transformer constructions (dark part of the disk presented in Fig. 1. a). We consolidated the both part together during the sintering process creating gradual juncture part between mentioned above two materials.



a)



b)

Fig. 1. PLZT/PZT-type FGM ceramics disks prepared for Piezoelectric Transformer fabrication (a) and schematic idea of PT (b)

Potential application in electronic industry needs thoroughly electrical and mechanical characterisation of this new FGM structure constructed from the PLZT/PZT material. We had expected high electrical anisotropy of this graded structure and we used the Impedance Spectroscopy (IS) method, that is capable present such inequality in form of separated impedance semicircles. Additionally we implemented dielectric measurements for further characterization of this interesting graded structure. Finally we used nano-mechanical methods for integration quality testing and measurements of mechanical parameters PLZT/PZT Functionally Graded Material for photovoltaic driven Piezoelectric Transformers.

Generally Impedance Spectroscopy (IS) proves to be an efficient method capable to detect contribution of grains and the grain boundaries resistance to the complex impedance of the PZT compound, properly estimate its electrical conductivity as well as corresponding activation energies and conclude on structural properties [11-13]. Subtle changes in piezoelectric materials structure have significant influence on conductivity and implementation for particular applications. IS is shown also to be a method for detecting the defects distribution. In case of PBZT solid solution optimal amount of isovalent dopant can be estimated due to change in electrical properties. In this material calcium dopants are located in the Pb vacancies, due to ionic radii similarity, decreasing the amount of point defects. We can find optimal amount of this dopant by process of monitoring the values of conductivity [14].

IS can be also effectively implemented for studying the aging process in ceramics, since the above mentioned structural changes influence the impedance spectra. Value of impedance was found also to increase due to fatigue and initiation of microcracks in electroceramics [15-17]. Taking into account mentioned above structural defects we proposed using mechanical quality testing of ceramics sample prior to impedance measurements.

1.1. Experimental

In the present work the gradation step of PLZT (3/52/48) and PZT (54/46) solid solution was manufactured using custom-designed steel die matrix to achieve 10mm in diameters and 1 mm thick disk. The preparation method of graded structure is described in detail in [8]. 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 PZT materials. For testing the mechanical quality of this part we used nanoindentation technique for measurements 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=900K were carried out using Solartron 1260 system in the frequency range =0.1 Hz - 2 MHz.

2. Results and discussion

2.1. Mechanical quality testing

The main problem in technology of graded structures is connected with the preparation of defect and crack-free joint part between the both materials. For consistency test of this part we used nanoindentation technique for measurements values of micro-mechanical parameters along the radius of obtained disks. The final quality proof was carried out by the mapping of hardness and elastic modulus distribution along the sample radius. Uniform slope shape of tip displacements during the material penetration, (Fig. 1.b) distinctly indicates lack of the cracks and accumulation of point defects in this interpart area. The values of the hardness and elastic modulus as a function of position along the radius of the PZT disk is presented in Fig. 1a. The hardness value is changing from the H=9 GPa to H=9,4 GPa along the radius (4%) and the Young Modulus values increasing from the E=127 GPa to E=129 GPa (2%), so that the juncture part of this graded structure has good mechanical quality.

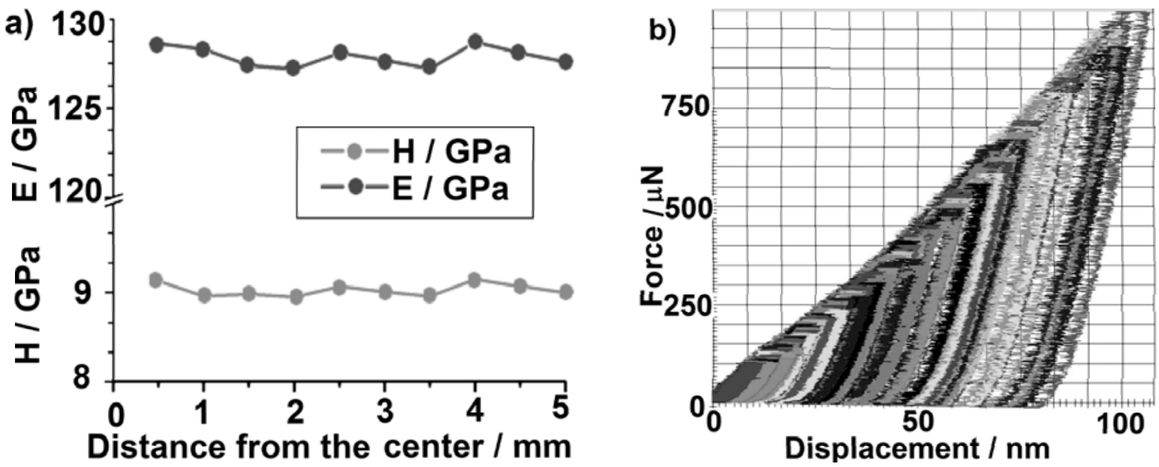


Fig. 1. The values of the hardness and elastic modulus as a function of position along the radius of the graded structure PLZT/PZT (a) and load-displacement curves (b)

2.2. Dielectric measurements

The graded structure PLZT/PZT and separate PZT, PLZT samples were used for measurements of the dielectric constant (ε') and the dielectric loss factor (ε") as a function of temperature. The determined temperature dependence of dielectric permittivity (ε') is presented in Fig. 2.a and dielectric loss factor (ε") in Fig. 2.b with a signal frequency f=1kHz. We have found that in FGM structures, both ε'(T) and ε"(T) curves reveal anomalies in the vicinity of temperature corresponding to the tetragonal-cubic (F_T-P_C) phase transition. We discovered that maximal permittivity in graded PLZT/PZT material not appeared at the same temperatures as in separated PLZT and PZT ceramics and is shifted to the lower values in comparison to separated materials. These temperature shift values are equal ΔT=8 degree in case of PZT and ΔT=24 degree in PLZT. We can observe similar situation in case of dielectric loss factor characteristics. We detected again the shifted maxima temperatures and the corresponding shifts are equal ΔT=6 degree in PZT and ΔT=25 degree in PLZT (Fig.2.b). We have noticed that, the dielectric characteristics of the PLZT/PZT graded structure is not a direct sum result of separate PLZT and PZT dielectric diagrams.

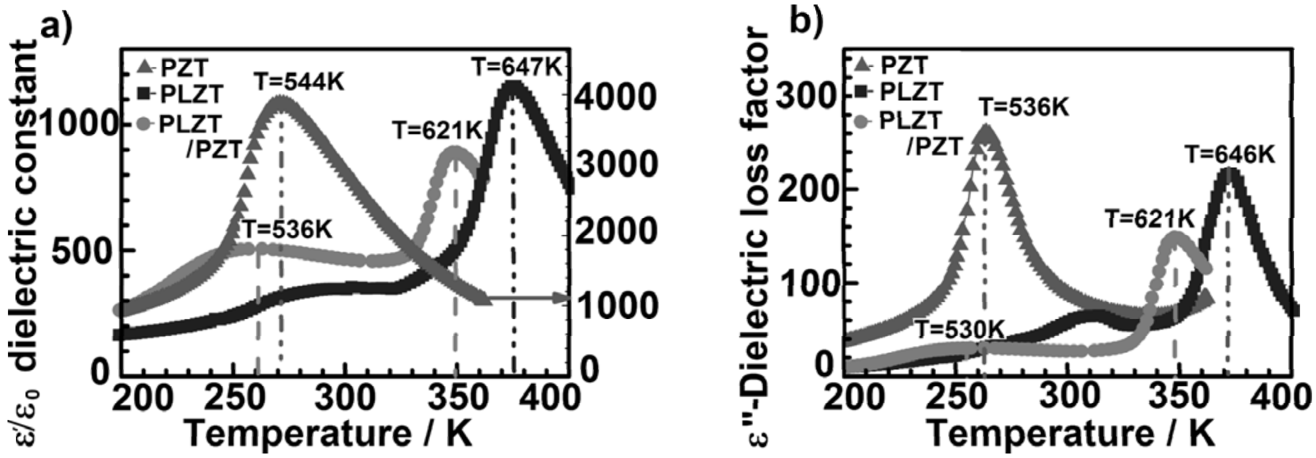


Fig. 2. The temperature dependence of dielectric permittivity (ε') (Fig. 2.a) and dielectric loss factor (tanδ) (Fig. 2.b) at frequency f=1kHz for the PZT, PLZT and PLZT/PZT ceramics

2.3. Impedance spectroscopy

From the results of AC impedance measurement, we indicated distinctly resolved two semicircles for the graded structure PLZT/PZT in the Z‘ and Z“ plots (Fig. 3.c), whereas in the separated materials these plots were almost coherent (Fig. 3.a and b). The presence of two semicircular arcs is resulting from cascading effect of parallel combination of resistive and capacitive elements (Fig. 3.d) arising from impedance PZT and PLZT material. The high frequency semicircles can be associated with PZT 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 non-linear least-squares fitting of equivalent circuits (CNLS-analysis). We analysed these plots to evaluate the value of grain conductivity for the discussed samples and to obtain the conductivity activation energies from Arrhenius plot. The value of grain resistance for PZT at temperature T=900 K changes from Rg=1,1 kΩ to Rg=0,72 kΩ whereas for PLZT from Rg=38 kΩ to Rg=1 kΩ.

To summarize, complex characteristics of the graded structure appeared to be a not a simply addition result of starting materials parameters. In authors opinion, predominantly diffusion direction deeply influenced final structure properties. We assumed that during the high temperature sintering process ion diffusion from one material overweight the second ion movement resulting in resistance domination of one particular material in final graded structure. For explanation of this phenomenon we developed electrical model consisted from over 1000 electrical analogs of grain and grain boundary impedance, that is particularly sensitive to diffusion direction and the results will be published.

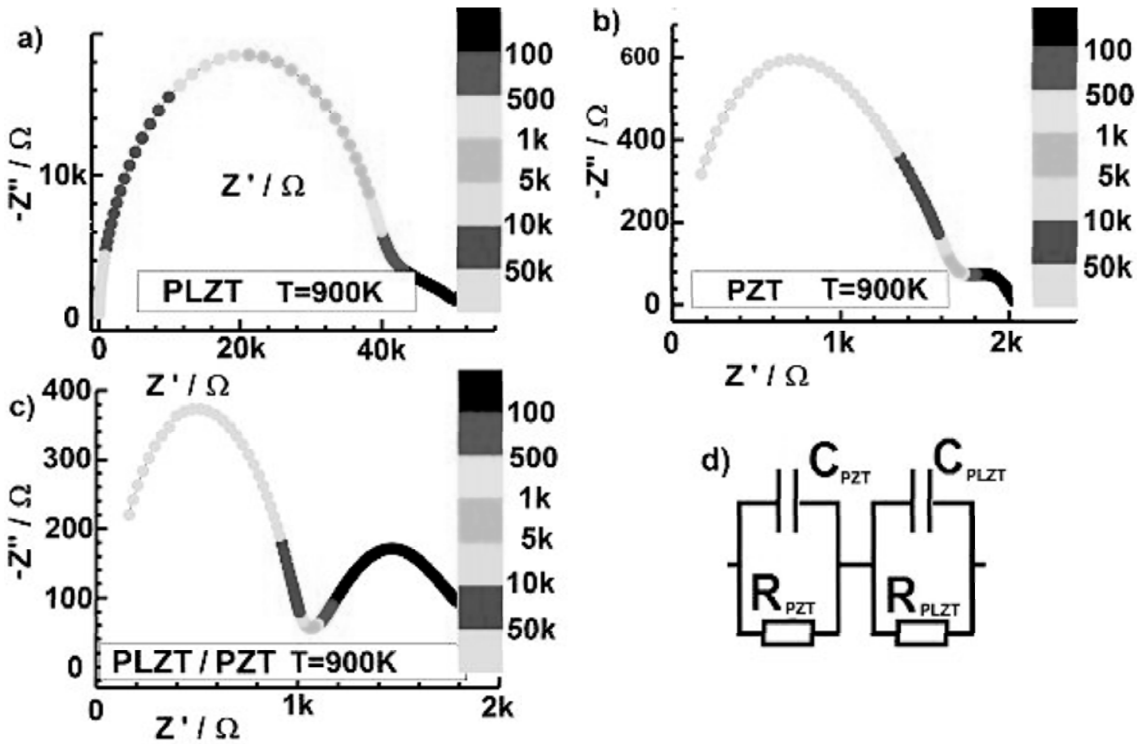


Fig. 3. Complex impedance plot (Nyquist plot) for the sample of PLZT (a), PZT (b) Functionally Graded Structure PLZT/PZT (c) ceramics at temperature T=900 K and electrical analog of investigated structure (d)

Additional information about total conductivity and the level of activation energies can be evaluated from the DC measurement (temperature range to T=900 K). Arrhenius plots of total conductivity derived from DC resistance of graded PLZT/PZT sample is shown in figure 4.b. One can see 3 regions of different activation energy, which can be associated with the stability regions between phase transitions of separated PZT and PLZT ceramics. The conductivity activation energy in the first region, in which temperature increase from room to PZT phase transition (T_C=536 K), is equal E_a=0,79 eV (Fig. 4.a), the next range between the Curie temperature of phase transitions of PZT and PLZT is equal E_a=1eV (TC=636 K) and the last E_a=0,87eV, respectively. The difference in activation energy values of total conductivity proves to be indicator of phase transition temperatures.

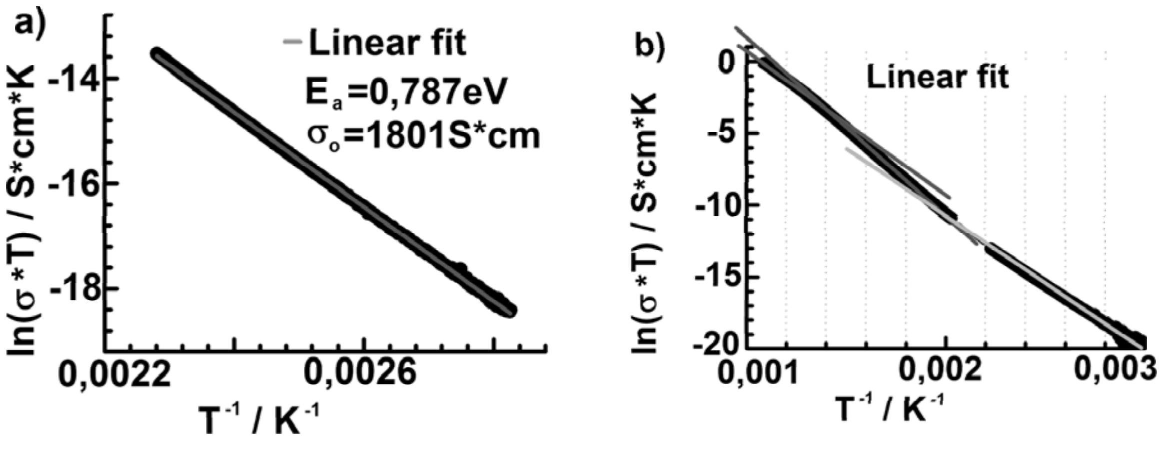


Fig. 4. The temperature dependence of DC total conductivity at temperature range 293-513K (a) and at temperature range 293-900K (b) of graded PLZT/PZT ceramics

3. Conclusions

The measurements revealed high electrical anisotropy of the PLZT/PZT graded structure and the Impedance Spectroscopy proved to be method capable reveal the level of such inequality in form of well separated semicircles. Additionally, dielectric measurement and values of activation energy explained the complexity of the integration process of Functionally Graded Structures. The juncture part mechanical quality results show, that final PLZT/PZT graded structure is good candidate for implementation in light driven piezoelectric transformers.

Acknowledgements

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