

# AN INFLUENCE OF THE DOMAIN STRUCTURE ON PHENOMENA OF THE INTERNAL FRICTION IN THE PZT TYPE PIEZOCERAMICS

Justyna BLUSZCZ, Radosław ZACHARIASZ, Marek CZERWIEC, Jan ILCZUK

Silesian University, Faculty of Engineering, Department of Material Science,  
3 Żeromskiego St., 41-200 Sosnowiec, POLAND, tel. (032) 291-83-91  
e-mail: rzachari@us.edu.pl

## 1. INTRODUCTION

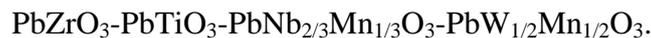
Ferroelectric ceramics of the PZT type because of its excellent dielectric piezoelectric and optical properties is widely used in electroacoustics with the medical equipment and in electronic industry. From the point of view of stability of its mechanical and electrical properties nowadays it is necessary to find out about phenomena taking place in the structure of this material [1]. Complexity of those phenomena is explained, first of all, by changes caused by interaction of defects of crystal lattice and domain structure [2-4].

The internal friction method, being a very important tool in the investigations of changes in the ferroelectric structure of ceramic materials, is one of the methods enabling to investigate those phenomena.

This work presents results of investigations of changes in the domain structure for a multicomponent ceramics of the PZT type using the above method.

## 2. MATERIAL TESTED

The solid solution of the PZT type with the following chemical composition was the material tested:



This material is characterized, among others, by low permittivity ( $\varepsilon_{33}^T / \varepsilon_0 \leq 260$ ) and high mechanical Q factor ( $Q_m > 2000$ ). For that reason it is used as high frequency electric transducers and pyroelectric detectors [5].

The specimens in question were obtained by hot pressing method ( $p = 2 \cdot 10^7$  Pa,  $t_s = 40$  minutes), and then they were polarized by low temperature method. In this methods specimens were heated to  $T_{\text{pol}} = (410 \div 430)\text{K}$  and polarized in the stable electric field  $E_{\text{pol}} = (3,5-$

$4,5) \cdot 10^6$  V/m for a period of time of  $t_{\text{pol}}=40$  minutes. The specimens used in the internal friction measurements had the following dimensions:  $(30 \times 10 \times 1)$  mm<sup>3</sup>. Measurements of the internal friction and resonance frequency of  $f_r$  specimen vibrations were conducted by a RAK-3 resonant mechanical spectrometer controlled by a computer [6]. Values of the dynamic Young modulus were determined on the basis of the relationship:

$$E = 94,68 \cdot \left(\frac{l_r}{h}\right)^3 \cdot \frac{m_d}{b} \cdot f_r^2 \quad [\text{Pa}], \quad (1)$$

where:  $l_r, h, b, m_d$  – length, thickness, width, mass of vibrating part of the specimen, respectively.

The internal friction method was selected for the tests on purpose, because as an investigation method is particularly sensitive to structural changes in solids. Since it is highly sensitive to changes in the concentration of defects responsible for the phenomena observed, it is possible to draw conclusions about the microstructure and its changes on the basis of macroscopic mechanical vibrations of the specimen. This methods enables to determine mechanical characteristics of materials in the resonance conditions [5].

### 3. RESULTS OF MEASUREMENTS

Figure 1 presents temperature relationships  $Q^{-1} = f(T)$  and  $E = f(T)$  obtained in three measurement cycles: for a non-polarized specimen in the initial state and polarized one.

For all determined relationships of the internal friction P1 and P2 peaks, of which temperature position changed visibly with successive cycles of the specimen heating process, were observed. At the temperature range from 550 to 600K a distinct P3 maximum was additionally observed. The temperature position of this maximum does not move in the successive measurement cycles, what proves that it does not have a relaxation character and is connected with the ferroelectric - paraelectric phase transformation.

In the case of the non-polarized specimen, in which due to the mechanical treatment there were a lot of defects and disordering of the crystal lattice occurred, there were P1 and P2 maxima with low height at  $T_1=377\text{K}$ ,  $T_2=467\text{K}$ , respectively.

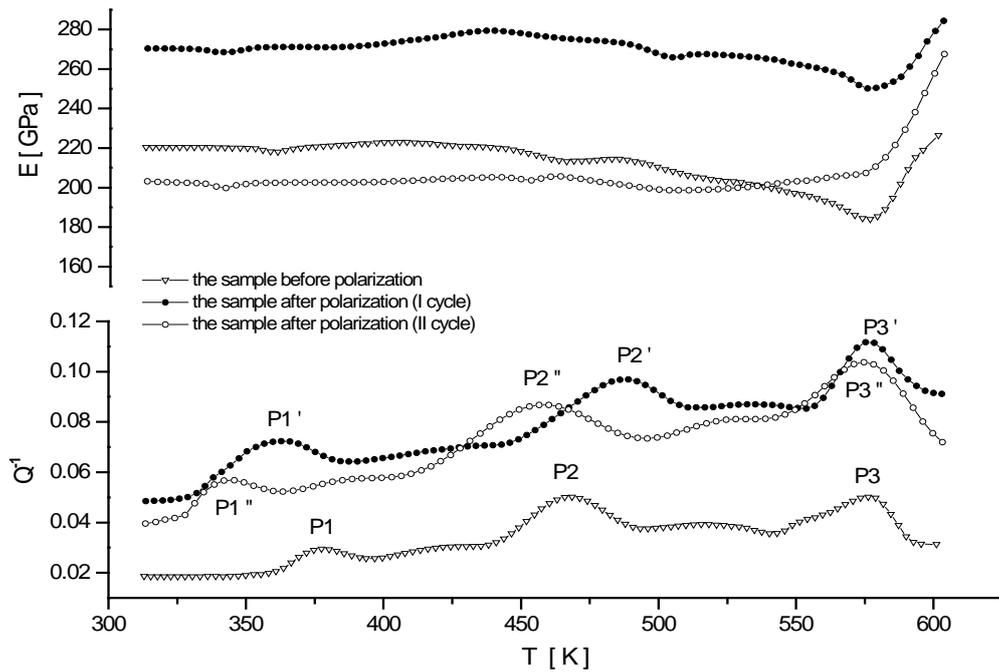


Fig.1. Temperature relationships of the internal friction  $Q^{-1}=f(T)$  and dynamic Young modulus  $E=f(T)$  for a non-polarized and polarized specimen

After performing 2 measurement cycles for a polarized specimen temperature displacement of P1 and P2 peaks, changes in the background values of the internal friction and values of the dynamic Young modulus E were observed. Process connected with changes taking place in the structure of the tested specimens is a mechanism responsible for such behaviour of the P1 and P2 maxima. As a result of polarization ordering of the domain structure and disappearance of some defects took place and it caused greater mobility of domain walls, resulting in an increase in the background value of the internal friction. The second measurement process conducted for that specimen revealed a displacement of the P1 and P2 peaks towards lower temperatures and a decrease in the internal friction value. A disorder of the domain structure, caused partly by heating in the 1 measurement cycle and loss of some oxygen vacancies, is a reason for it.

During all measurement cycles changes in the  $E=f(T)$  relationship, which were particularly visible in the area of the P3 peak presence, connected with the phase transformation, were also observed.

In the area of the P1 and P2 presence for a non-polarized and polarized specimen activation energy and value of a preexponential factor in the Arrhenius law  $\tau_0$  were determined [7]. Values are presented in table I.

The origin of the P1 and P1' is mainly connected with an influence of point defects, to a great extent in a form of oxygen vacancies. The activation energy values determined, which are near energy connected with migration of oxygen vacancies in the ferroelectric materials of the PZT type, confirm it. Energy is in a range about 0,6-0,8 eV [8].

The observed increase in energy values in the case of P2 and P2' peaks is connected with a complex process taking place in the ceramic structure, originating mainly from presence point defects and inelastic movement of the domain walls.

Table I. Characteristic parameters in the area of presence of the P1 and P2 maxima for a non-polarized and polarized specimen

Type of parameter	Non-polarized specimen		Polarized specimen	
	P1	P2	P1'	P2'
T <sub>p</sub> [K]	377	467	363	489
H [eV]	0,89±0,03	1,13±0,02	0,79±0,02	1,20±0,02
τ <sub>o</sub> [s]	(2,42±0,06)·10 <sup>-16</sup>	(1,35±0,08)·10 <sup>-16</sup>	(1,69±0,06)·10 <sup>-15</sup>	(8,22±0,06)·10 <sup>-17</sup>

In order to specify mechanisms responsible for presence of P1 and P2 peaks amplitude relationships of the internal friction were determined in the area of presence of those peaks and in the paraelectric phase (T=593K). The ε values were determined on the basis of an appropriate relation:

$$\varepsilon_{\max} = \frac{4d}{\pi ml^2} \frac{\alpha_n^2}{f_r^2} \frac{C_e V_p \nu_1}{g \delta} \times 10^{-5}, \quad (2)$$

where:

d-specimen thickness [cm],

α<sub>n</sub>-constant (0,879),

C<sub>e</sub>-capacitance formed between a fixed electrode and the grounded reed [pF],

V<sub>p</sub>-voltage on the electrodes [V],

l<sup>2</sup>- surface of the vibrating part of the specimen [cm<sup>2</sup>],

ν<sub>1</sub>-the amplitude of the ac drive voltage [V],

m-mass of the vacilaiting part sample [g],

f<sub>r</sub>- frequency of specimen vibrations [Hz],

g-the gap distance between the sample and electrode [cm],

δ -damping decrement,

and a graphic representation of the  $Q^{-1}=f(\varepsilon)$  relations obtained is presented in fig.2.

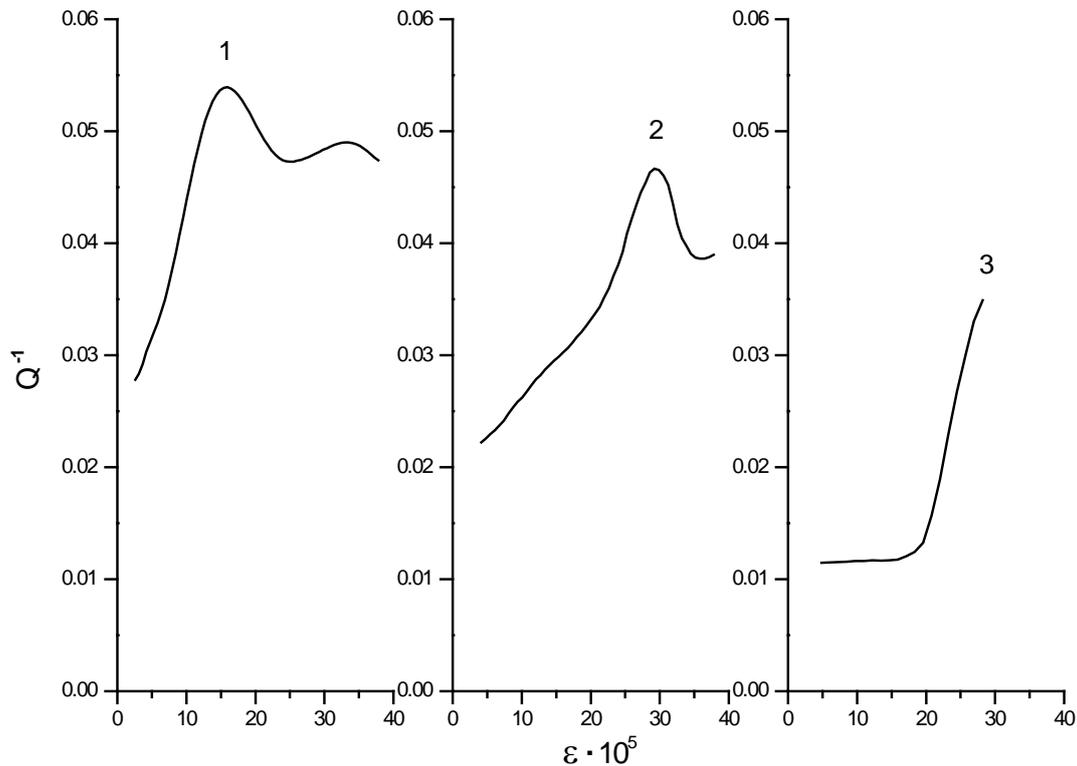


Fig. 2. The relationships of the internal friction  $Q^{-1}=f(\varepsilon)$  for non-polarized specimen: T=340 K (curve 1), T=454 K (curve 2), T=593 K (curve 3)

Curve 1 determined at temperature about 340 K is of a complex character. Its first part, consistent with the Granato - Lucke's model, confirms an interaction of point defects and domain walls. Two characteristic maxima are a proof of presence of microdeformations connected with the specimen deformation during vibrations and movement of the domain walls [8]. Changes connected with the amplitude relation of the internal friction in the area of the P2 peak (T=454 K) are presented in curve 2. With an increase in deformation mobility of the domain walls increases, because there is a decrease in the concentration of oxygen vacancies. The maximum visible in the  $Q^{-1}=f(\varepsilon)$  diagram confirms greater mobility of the domain walls. The phase transformation of piezoceramics into paraelectric phase is connected with disappearance of domain walls, what is proved by the course of curve 3 in the diagram of the amplitude relationship of the internal friction. The straight line character of that curve is caused by interaction of only point defects and dislocations.

#### 4. CONCLUSIONS

1. Changes observed in position of the P1 and P2 peaks on the  $Q^{-1}=f(T)$  temperature relations confirm changes taking place in the domain structure of the PZT type piezoceramics,
2. The determined parameters of the Arrheniu's law prove that the P1 and P2 maxima originated from the interaction of point defects and domain walls and are of relaxation character,
3. The performed investigations of the  $Q^{-1}=f(\varepsilon)$  amplitude relation showed that presence of point and linear defects was responsible for the internal friction in the paraelectric phase in the multicomponent compounds of the PZT type.

#### REFERENCES

1. R. Zachariasz, J. Ilczuk, *Wpływ defektów strukturalnych na zjawiska tarcia wewnętrznego w piezoceramice typu PZT*, Prace Naukowe Instytutu Telekomunikacji i Akustyki Politechniki Wrocławskiej nr 83, Wrocław 2001, s. 453-458
2. Wang Zhi Yong, Chen Ting Guo, *Acta Physica Sinica*, 7, 764-772, 1998
3. R. C. Buchanan, T. R. Armstrong, R. D. Roseman, *Ferroelectrics*, 135, 343-369, 1992
4. L. Sagalowicz, F. Chu, P.D. Martin, D. Damjanovic, *Journal of Applied Physics*, 88, 2000
5. J. Ilczuk, J. Dudek, Z. Surowiak, *Molecular and Quantum Acoustics*, 18, 101-110 (1997)
6. R. Zachariasz, J. Ilczuk, A. Chrobak, *Ceramics*, 66, 630-636 (2001)
7. A. Puskar, *Internal Friction of Materials*, Cambridge International Science Publishing, Cambridge 2001
8. K. Carl, K. H. Hardtl, *Ferroelectrics*, 17, 473 (1978).