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
Volume 58 2013 Issue 4

DOI: 10.2478/amm-2013-0168

R. ZACHARIASZ*, J.A. BARTKOWSKA*, D. BOCHENEK*, P. NIEMIEC*

INTERNAL FRICTION IN THE FERROELECTRIC – FERROMAGNETIC COMPOSITES

TARCIE WEWNĘTRZNE W KOMPOZYTACH FERROELEKTRYCZNO – FERROMAGNETYCZNYCH

In the work, temperature $Q^{-1}(T)$ and amplitude $Q^{-1}(\varepsilon)$ dependences of internal friction for ferroelectric – ferromagnetic composites on the base of the PZT and ferrite were determined. The temperature dependences of internal friction Q^{-1} reveal the peak in the range of high temperatures. We have investigated the peak associated with the viscous-elastic mobility of ferroelectric domain walls. The internal friction due to the viscous-elastic mobility of ferroelectric domain walls was calculated and compared with the experimental data in the reference to the Wang's theory. Based on internal friction measurements and theoretical considerations, the peak was described. Additionally the amplitude (isothermal) $Q^{-1}(\varepsilon)$ dependences for the composites were made. This allowed for the interpretation of the maximum observed on the temperature dependences of the internal friction $Q^{-1}(T)$.

Keywords: internal friction, relaxation time, ferroelectric domain wall, Wang's model, composites

W pracy wyznaczano temperaturowe $Q^{-1}(T)$ i amplitudowe $Q^{-1}(\varepsilon)$ zależności tarcia wewnętrznego dla ferroelektryczno – ferromagnetycznych kompozytów powstałych na bazie PZT i ferrytu. Na wykresach temperaturowych zależności tarcia wewnętrznego, uzyskanych dla badanych materiałów, obserwowano występowanie maksimum w zakresie wysokich temperatur. Do opisu maksimum zastosowany został model Wanga, który uwzględnia oddziaływania związane z lepko-sprężystym ruchem ferroelektrycznych ścian domenowych. Model ten bardzo dobrze opisał wyniki doświadczalne zależności tarcia wewnętrznego Q^{-1} od temperatury. Dobra zgodność pomiędzy modelem, a danymi pomiarowymi sugeruje, że za powstanie maksimum odpowiedzialne są lepko-sprężyste ruchy ścian domenowych. Ponadto przeprowadzono pomiary amplitudowych (izotermicznych) zależności tarcia wewnętrznego $Q^{-1}(\varepsilon)$ dla badanych kompozytów. Pozwoliło to na interpretację maksimum obserwowanego wcześniej na temperaturowych zależnościach tarcia wewnętrznego.

1. Introduction

In order to widen a scope of applications of composite materials it is necessary to learn about physical, chemical and mechanical properties and relaxation properties of these materials. Rapid growth of modern investigation methods, mainly non-destructive ones, is observed in examinations of composite materials. Methods of mechanical spectroscopy are more often used; an internal friction method in particular. The internal friction (IF) is caused by irreversible energy losses occurring in solid bodies as a result of many processes in which defects of crystal lattice participate. It is characterized by inelastic behavior of bodies when external stresses act on them and it appears in a form of losses of some energy of mechanical vibrations (a part of the mechanical energy changes into thermal energy) [1-3]. This method is characterized by high sensitivity to changes in the concentration of point defects, interaction between defects of the crystal structure and to changes in the real structure of materials [4-6].

The temperature dependences of internal friction Q^{-1} reveal the peak in the range of high temperatures. We have investigated the peak associated with the viscous-elastic mobility of ferroelectric domain walls. The internal friction due to the viscous-elastic mobility of ferroelectric domain walls was calculated and compared with the experimental data in the reference to the Wang's theory. Based on internal friction measurements and theoretical considerations, the peak was described [7].

The method which enables to determine precisely mechanisms responsible for formation of the internal friction peaks on the $Q^{-1}(T)$ dependences is a measurement of the $Q^{-1} = f(\varepsilon_{am})$ amplitude relationships of the internal friction (AIF). Additionally the amplitude (isothermal) $Q^{-1}(\varepsilon)$ dependences for the composites were made. This allowed for the interpretation of the maximum observed on the temperature dependences of the internal friction $Q^{-1}(T)$ [8].

2. Experiment

For the purpose of obtaining ferroelectric-ferromagnetic composites, PZT type powder was used with ferroelectric properties as well as nickel-zinc ferrite pow-

---

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

der $Ni_{0.64}Zn_{0.36}Fe_2O_4$ with ferromagnetic properties. The ferroelectric powder comprised two PZT type compositions: $Pb_{0.90}Ba_{0.10}(Zr_{0.53}Ti_{0.47})O_3$+2%at.$Nb_2O_5$ (PBZTN) and $Pb_{0.94}Sr_{0.06}(Zr_{0.46}Ti_{0.54})O_3$+0.25%at. Cr_2O_3 (PSZTC) according to work [9, 10]. The main component of the composite – ceramic PZT type powder – was synthesized using sintering of a mixture of simple oxides in solid phase (compaction by free sintering) under the following conditions: $T_{synth} = 850°C$, $t_{synth} = 2$ h. The second component of the composite with ferromagnetic properties-ferrite powder $Ni_{0.64}Zn_{0.36}Fe_2O_4$ (described in the work [11]) was synthesized using calcination under conditions of 1100°C/4 h. The synthesized ceramic powder constituted 90%, while the ferrite powder - 10%, of the PZT-NiZn composite. After proportionally weighing and mixing components, synthesizing was carried out using sintering of the mixture of simple oxides in a solid phase (compaction by a free sintering method) under the following conditions: $T_{synth} = 1050°C$ and $t_{synth} = 4$ h. Compaction of the synthesized composite powder (sintering) was carried out using free sintering of compacts under the following conditions: $T_s = 1250°C/t_s = 2$ h. Two ferroelectric-ferromagnetic composites were obtained, marked in the following way: PBZTN-NiZn, PSZTC-NiZn. Silver electrodes were applied to surfaces of composite samples for the purpose of carrying out electric tests.

The measurements of the temperature and amplitude dependences of internal friction Q^{-1} and temperature dependences of Young's modulus $Y(T)$ were performed by automatic resonance mechanical spectrometer RAK-3 type controlled by computer.

3. Results and discussion

In the figure 1 the temperature dependences of the internal friction $Q^{-1}(T)$ and Young's modulus $Y(T)$ at the frequency $f = 610$ Hz are presented. For both composites, on the dependences $Q^{-1}(T)$ at the temperature range from 250°C to 420°C, clear increase of the internal friction values is observed (characteristic maximum).

Simultaneously, on the temperature dependences of the Young's modulus $Y(T)$, the characteristic local minimum (at the temperature about 340°C for the PBZTN-NiZn composite and about 355°C for the PSZTC-NiZn composite) and next its intense growth is observed. Such behavior confirms occurrence in this temperature area the phase transitions. Considerable diffuseness of the internal friction on the dependences $Q^{-1}(T)$ is connected with overlapping two phase transitions, occurring in the PZT-NiZn composites: the phase transition of the electric subsystem and magnetic subsystem. For both composites, below 250°C, on the $Q^{-1}(T)$ as well as $Y(T)$ dependences anomalies showing about occurrence another phase transitions was not observed. The PBZTN-NiZn composite shows lower values of the internal friction (mechanical losses) Q^{-1} as well as higher value of the Young's modulus Y in comparison with the PSZTC-NiZn composite.

Figures 2 and 3 demonstrate the temperature dependences of the Young's modulus $Y(T)$ and the internal friction $Q^{-1}(T)$ for the tested composite samples for two different resonance frequencies, which values at the room temperature are respectively: $f_1 = 610$ Hz, $f_2 = 636$ Hz (for PBZTN-NiZn) and $f_1 = 610$ Hz, $f_2 = 643$ Hz (for PSZTC-NiZn). It is visible clearly, that together with increasing the values of the frequencies, the values of the Young's modulus are increasing. It is compatible with dependence used in calculations:

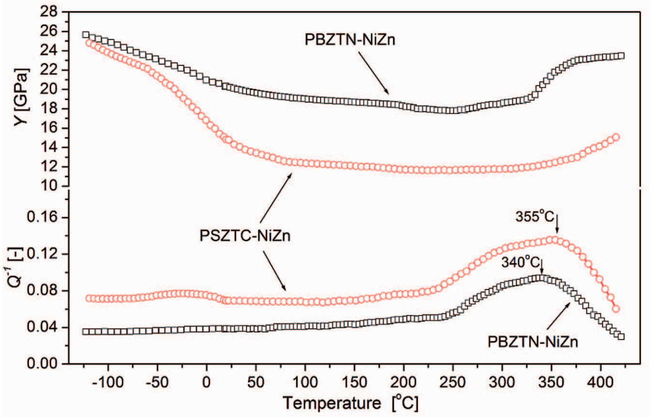

Fig. 1. Temperature dependences $Q^{-1}(T)$ and $Y(T)$ for PBZTN-NiZn and PSZTC-NiZn composites at the frequency $f = 610$ Hz

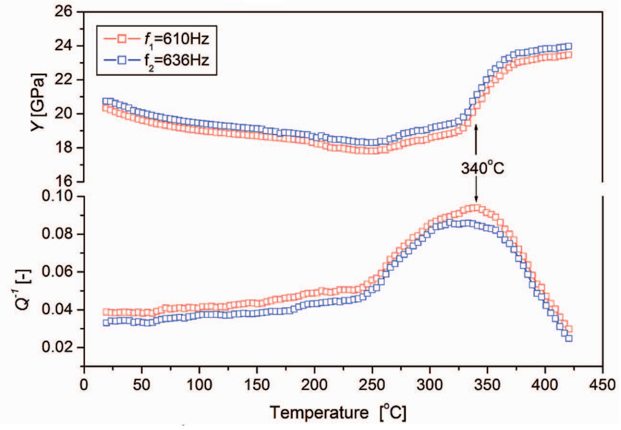

Fig. 2. Temperature dependences $Q^{-1}(T)$ and $Y(T)$ for PBZTN-NiZn composite at different frequencies: $f_1 = 610$ Hz, $f_2 = 636$ Hz

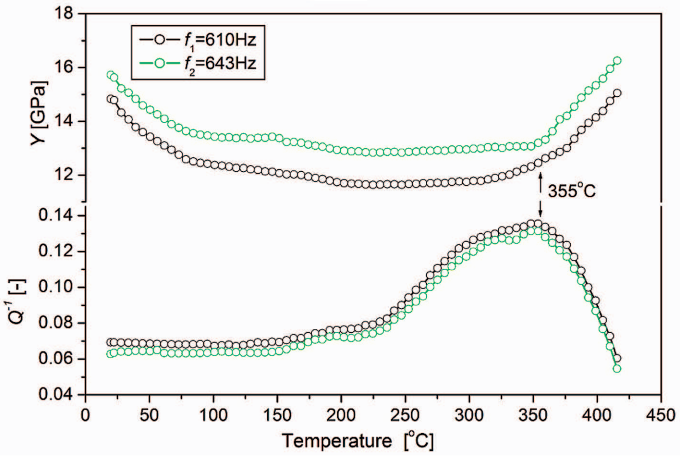

Fig. 3. Temperature dependences $Q^{-1}(T)$ and $Y(T)$ for PSZTC-NiZn composite at different frequencies: $f_1 = 610$ Hz, $f_2 = 643$ Hz

$$Y = 94.68 \times \left(\frac{lr}{h}\right)^3 \times \frac{m_d}{b} \times f_r^2, \qquad (1)$$

where: l_r, h, b i m_d – respectively: length, thickness, width and mass of vibratible part of the sample.

Additionally, stability of the temperature position of the characteristic maxima with the change in the measurement frequency shows that a phase composition taking place in the composite materials is responsible for formation maxima of internal friction. Changes in height of the maxima also show their relationship with phase changes. According to the model developed by J. F. Delorme and P. F. Gobin, describing the $Q^{-1}(T)$ dependences in the area of the phase change of the internal friction is described by a formula in the form [12]:

$$Q^{-1} = \frac{KG}{\omega} \cdot \frac{\partial m}{\partial T} \cdot \frac{\partial T}{\partial t}, \tag{2}$$

where: K – material constant, G – modulus of rigidity, ω – frequency of the specimen vibrations; $\omega = 2\pi f$(f – frequency), $\frac{\partial m}{\partial T}$ – quantity of the material undergoing the change at the unit temperature change, $\frac{\partial T}{\partial t}$ – rate of the temperature changes.

Therefore, the increase height of the maxima with a decrease in the measurement frequency is fully confirmed by the Delorme-Gobin model.

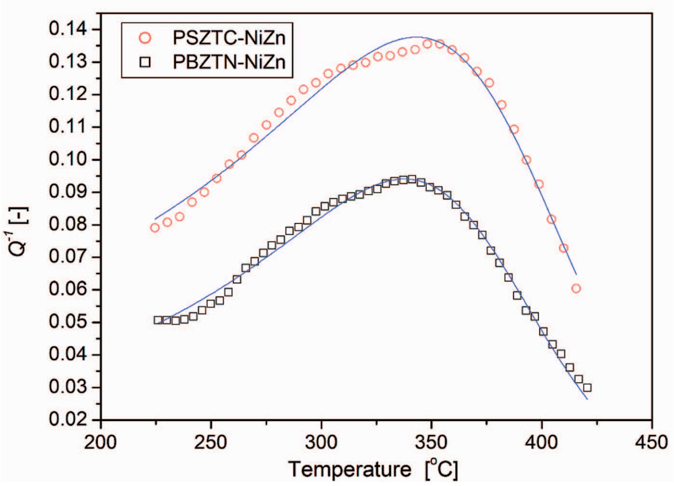

Fig. 4. The peak of internal friction as function of temperature for PBZTN-NiZn and PSZTC-NiZn composites. Solid lines present theoretical curves

In this paper we also attempted the theoretical description of the maxima on the dependences $Q^{-1}(T)$ in the temperature range from 250°C to 420°C, based on the Wang's model.

The existence of ferroelectric domains in crystals composites can decrease the elastic energy of the crystals but it increases the interface energy, so there is a definite number of ferroelectric domain walls in the crystals at each temperature. When $T \rightarrow T_C$, the interface energy $\rightarrow 0$ for order parameter $\rightarrow 0$ so the density of the ferroelectric domain walls $N \rightarrow \infty$, Wang *et al.* [13] have detailed the relation between N and the temperature as:

$$N = N_0/(T_C - T), \tag{3}$$

where N_0 is a constant independent of temperature.

When $T \rightarrow T_C$, the order parameter $\rightarrow 0$, the viscous coefficient Γ of the mobility of ferroelectric domain walls in the crystal $\rightarrow 0$. Combs and Yip got the expression for the viscous coefficient by computer simulation as:

$$\Gamma = A\exp\left(-B/(T_C - T)\right), \tag{4}$$

According to Wang's [13] theory the internal friction due to the viscous-spring movement of the ferroelectric domain walls is:

$$Q^{-1} = h_3 \frac{\omega\tau}{1 + \omega^2\tau^2}, \tag{5}$$

where: $\tau = \tau_0 = A_0 \exp\left(-B/(T_C - T)\right)$,$h_3 = 2N_0\varepsilon_0^2 C'/J'k$, and $A, B, N_0, k_0, \varepsilon_0$ are constants independent of temperature, $\omega = 2\pi f$, f – frequency of the maxima and $A_0 = A/k_0$.

The calculated results and experimental data of the internal friction are shown on Figure 4. There is a good agreement between experimental data and results of calculation. Our results are coincident with experimental data, where B [K] = 8.98; 10.91 and A_0 [s] $=1.19\times10^{-3}$; 0.61×10^{-3} for the PSZTC-NiZn and PBZTN-NiZn composites respectively. The values of the h_3 parameter are equal 2.75×10^{-4} and 1.89×10^{-4} respectively for both composites (PSZTC and PBZTN).

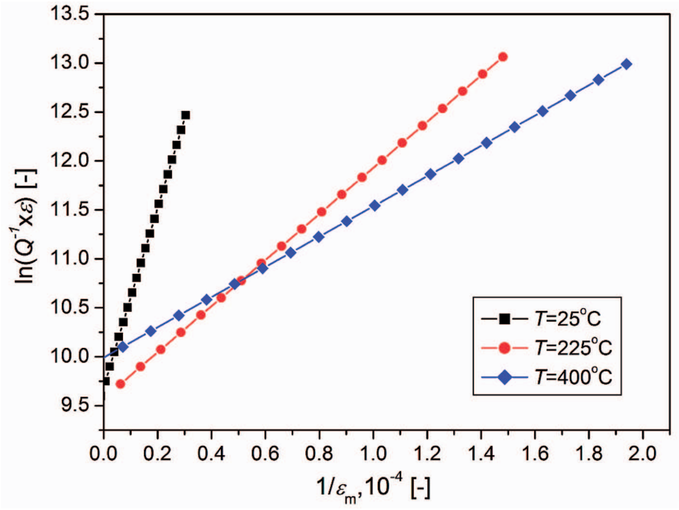

Fig. 5. The dependences of $\ln(Q^{-1} \times \varepsilon_m)$ = f($1/\varepsilon_m$) for the tested PBZTN-NiZn composite

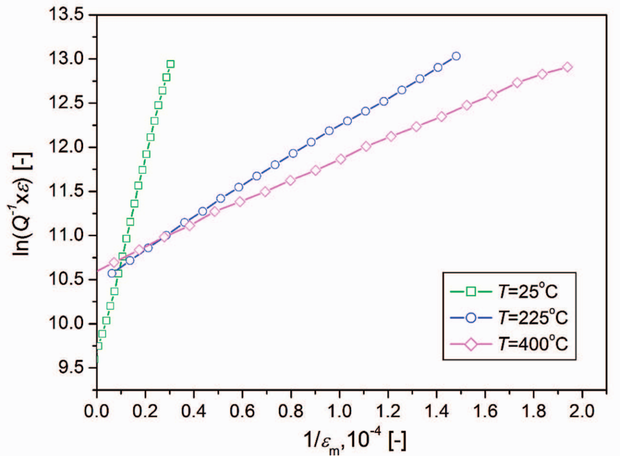

Fig. 6. The dependences of $\ln(Q^{-1} \times \varepsilon_m)$ = f($1/\varepsilon_m$) for the tested PSZTC-NiZn composite

To analyze in details the changes occurring in the above investigated composites and also below the temperature of phase transition, the measurements of amplitude dependences of internal friction in the function of deformation of the sample $Q^{-1}(1/\varepsilon_m)$ were conducted. On the figures 5 and 6 the amplitude dependences of internal friction in half-logarithmic coordinate $\ln(Q^{-1} \times \varepsilon_m) = f(1/\varepsilon_m)$ for tested composites are presented. The amplitude dependence of the internal friction Q^{-1} is described by the following formula:

$$Q^{-1} = \frac{C_1}{\varepsilon} \exp\left(-\frac{C_2}{\varepsilon}\right), \tag{6}$$

where $C_2 = (K \times a \times \delta)/L_C$, ($K$, a, δ - material constant).

The analysis of the dependences obtained was carried out on the basis of the A. Granato and K. Lücke model. The model assumes that dislocations are fastened by point defects. There is, therefore, certain characteristic length of dislocation lines, which is marked as L_C and is inversely proportional to C_2 coefficient [14]. In the figures 5 and 6 it is visible that rectilinear dependences with different inclination depending temperature were obtained. Such rectilinear dependences prove that ferroelectric domain wall can be treated as a set dislocation lines. After analyzing every rectilinear dependence, the values of the C_2 and next of the L_C coefficients are calculated (Table 1). The fact of increase in the L_C values at subsequent temperatures, especially at the temperature 400°C in paraelectric phase, show more and more sizes of ferroelectric domain walls. It confirms the changes occurring as a result of phase transition in investigated composites.

TABLE 1

Specification of values of the C_2 coefficient and L_C/L_{C0} length for the tested composites

	PBZTN-NiZn	PSZTC-NiZn
C_2 coefficient at T = 25°C	9.78	9.54
L_C at T = 25°C	1.00	1.00
C_2 coefficient at T = 225°C	2.56	2.82
L_C at T = 225°C	3.80	3.40
C_2 coefficient at T = 400°C	1.52	1.32
L_C at T = 400°C	6.43	7.22

4. Conclusions

Results of investigations of two ferroelectric – magnetic composites are presented in the work. It was found that the changes in the temperature dependences of the Young's modulus $Y(T)$ and internal friction $Q^{-1}(T)$ correspond to the temperature range of the phase transition determined from electric measurements of the $\varepsilon(T)$ dependences. Investigations of the amplitude dependences of the internal friction and their analysis with use of the Granato-Lücke model enable to obtain a lot of important information on processes taking place in the ceramic materials. Based on dielectric permittivity measurements and theoretical considerations, the values of the magnetoelectric coupling coefficient were specified.

REFERENCES

[1] R. Zachariasz, A. Zarycka, J. Ilczuk, Determination of the lead titanate zirconate phase diagram by the measurements of the internal friction and Young's modulus, Material Science – Poland **25**, 781-789 (2007).

[2] A. Puskar, Internal Friction of Materials, Cambridge International Science Publishing, Cambridge (2001).

[3] C. Pawlaczyk, M. Olszowy, E. Markiewicz, E. Nogas-Ćwikiel, J. Kułek, Dielectric behaviour and pyroelectricity In SBN70-PVC composites, Phase Transitions **80**, 177-183 (2007).

[4] B.L. Cheng, M. Gabbay, M. Maglione, G. Fantozzi, Relaxation motion and possible memory of domain structures in barium titanate ceramics studied by mechanical and dielectric losses, J. Electroceramics **10**, 5-18 (2003).

[5] R. Zachariasz, D. Bochenek, Low frequency elastic and anelastic properties of $Pb(Fe_{1-x}Nb_x)O_3$ ferroelectric ceramics, The European Physical Journal Special Topics **154**, 253-256 (2008).

[6] D. Bochenek, R. Zachariasz, PFN ceramics synthesized by a two-stage method, Archives of Metallurgy and Materials **54** (3), 903-910 (2009).

[7] J.A. Bartkowska, J. Bluszcz, R. Zachariasz, J. Ilczuk, B. Brus, Domain wall motion effect In piezoelectric ceramics, J. Phys. IV France **137**, 19-21 (2006).

[8] R. Zachariasz, B. Bruś, A. Zarycka, M. Czerwiec, J. Ilczuk, Application of measurements of internal friction amplitude dependences for tests of ceramic materials, Physica Status Solidi A **205**(4), 1120-1125 (2008).

[9] R. Zachariasz, D. Bochenek, Parameters of ceramics obtained on the base PZT used to build electroacoustic converters, Journal de Physique IV **137**, 189-192 (2006).

[10] D. Bochenek, P. Niemiec, R. Zachariasz, A. Chrobak, G. Ziółkowski, Ferroelectric – ferromagnetic composites based on PZT type powder and ferrite powder, Archives of Metallurgy and Materials **58** (3) (2013).

[11] D. Bochenek, P. Niemiec, P. Wawrzała, A. Chrobak, Multiferroic Ceramic Composites Based on PZT Type Ceramic and NiZnFe, Ferroelectrics **448**, 96-105 (2013).

[12] R. Zachariasz, J. Ilczuk, D. Bochenek, Influence of the technology conditions on the mechanical and dielectric properties of the PZT-base piezoceramic transdcers, Solid State Phenomena **89**, 303-308 (2003).

[13] Y.N. Huang, Y.N. Wang, H.M. Shen, Internal friction and dielectric loss related to domain walls, Phys. Rev. B **46** (5), 3290-3295 (1992).

[14] A. Granato, K. Lücke, The vibrating string model of dislocation damping, Physical Acoustics **4A**, 225-276 (1966).

Received: 20 September 2013.