Proceedings of the Estonian Academy of Sciences,
2017, **66**, 4, 462–466
https://doi.org/10.3176/proc.2017.4.07
Available online at www.eap.ee/proceedings

FUNCTIONAL MATERIALS

Heat losses in ferroelectric ceramics due to switching processes

Olga Malyshkina*, Anton Eliseev, and Rostislav Grechishkin

Tver State University, Sadovij per. 35, 170002 Tver, Russia

Received 22 April 2017, revised 23 August 2017, accepted 23 August 2017, available online 30 November 2017

Abstract. In the present work, the heat losses of ferroelectric ceramics due to the switching processes were studied in high-amplitude AC electric fields in a wide frequency range of 50 to 1500 Hz. We showed the existence of a correlation between the time dependences of switchable polarization and self-heating temperature. Based on the approximation of the experimental data, an analytical expression was obtained for describing the decrease in the switched polarization with increasing electric field frequency corresponding to the exponential law. The method of estimating volumetric heat capacity coefficient by using the heat dissipation during the switching process was proposed.

Key words: ferroelectric ceramics, switching processes, heat losses.

1. INTRODUCTION

Due to favourable combination of piezoelectric properties and coefficients of electromechanical coupling, the ferroelectric ceramics of lead zirconate titanate family (PZT) is the main functional material for the production of microelectromechanical systems (MEMS) [1–3]. Among the further problems waiting for their solution are those of minimizing different kind of losses in piezoceramic elements working at high power regimes under high electric field strengths. Dictated by the nowadays increase of industrial applications of piezoelectric actuators a great attention is paid to fundamental studies of the problems of their reliability [4,5].

The main drawbacks of the piezoelectric material performance are connected to the existence of different kinds of losses. The latter are generally classified into three main types: dielectric, mechanical and electromechanical ones [4,6]. In ferroelectrics the dielectric losses are associated with the hysteresis in the spontaneous polarization switching in alternating electric fields (dielectric hysteresis loop), i.e. with the domain wall moving. Mechanical losses are characteristic of such applications of piezoceramics as ultrasound motors [7–9]. They are the higher, the lower is the mechanical figure of merit Q_m. In the resonance mode followed by the increase of vibration [4, 10–12] these losses lead to the rise of excess heat build-up related with field-induced mechanical stresses. Electromechanical losses as described by [4,12] appear due to transformation of the electric energy (electric displacement D, polarization P) into mechanical deformation (stresses) owing to piezoelectric effect. Thereby a considerable amount of heat is released, which is classified as heat losses (a self-heating) [9,13–16].

The authors of [4] mark out the following four causes of dielectric and electromechanical losses: (1) related with domain wall motion; (2) point crystal lattice defects; (3) microstructural losses at the grain boundaries because of material crystallinity; and (4) ohmic losses observed mainly in materials with large electric conductivity. In piezoceramic materials the losses of the first kind

* Corresponding author, Olga.Malyshkina@mail.ru

dominate over other ones. They are related with the motion of domain walls including dielectric, elastic and electromechanical hysteresis losses [13]. As a result of the losses significant undesirable self-heating comes into existence [9,14,15].

The current theoretical understanding and experimental characterization of the complex self-heating processes in piezoelectric ceramics are far from being complete. In the present work, we focus our attention on the study of heat losses and switching processes in ferroelectric ceramics due to the switching processes in large AC electric fields of different frequency.

2. MATERIALS AND METHODS

A study is made of the polarization processes in samples of PZT ferroelectric ceramics ($(Pb_{0.95}Sr_{0.05}(Zr_{0.53}Ti_{0.47})O_3)$) with a perovskite structure in AC sine electric fields with an amplitude of 500–2100 V/mm in a frequency range of 50 to 1500 Hz. The experimental results given below were obtained for samples with a thickness of the size of 1 mm × 5 mm × 5 mm. Sides 5 mm × 5 mm were supplied by silver electrodes.

Dielectric hysteresis loops were measured by the modified Sawyer–Tower method simultaneously with distant temperature control with the aid of thermal vision camera Testo–875-1. High voltage amplifier TREK 677B was exploited as a source of high AC voltage. Measurement were performed under ordinary laboratory environment without sample thermostating. At the start time the AC field was applied to the sample. The time dependence of the sample temperature and the dielectric hysteresis loops were measured simultaneously.

3. RESULTS

On applying an AC electric field of fixed amplitude (E_a), the samples initially demonstrate non-saturated minor hysteresis loops that change their shape during further exposure. This reshaping is accompanied by a self-heating of the sample. As distinct from the hysteresis loops appearing initially at the start time of field application (Fig. 1a), the shape of saturated hysteresis loops (Fig. 1b) depend on AC field frequency. The time of their formation also depends on frequency. This fact becomes evident by the demonstration of the time dependence of switched (reversible) polarization (P_{rev}) (Fig. 2a). Maximal increase of the switching polarization is observed when the sample self-heating temperature approached 80–90 ^{o}C. It is necessary to mention that the changes in the spontaneous polarization time dependence correspond with that of the sample temperature (Fig. 2b). Thus, the performed measurements have shown that the self-heating maximal temperature depends not only on the applied field amplitude [16,17], but also on its frequency.

After forming of the loop when the switched polarization approaches saturation the sample temperature does the same. Inasmuch as the sharp increase of the switched polarization (Fig. 2a) was accompanied by a decrease of the coercive force (Fig. 3a), it is natural to assume that the decrease of the coercive force with the increase of the temperature is responsible for the loop forming in time. It is interesting to note that the temperature course of the coercive field during forming the loop (Fig. 3b) is similar to that observed earlier in PZT samples under ordinarily conditions of heating in thermostat camera.

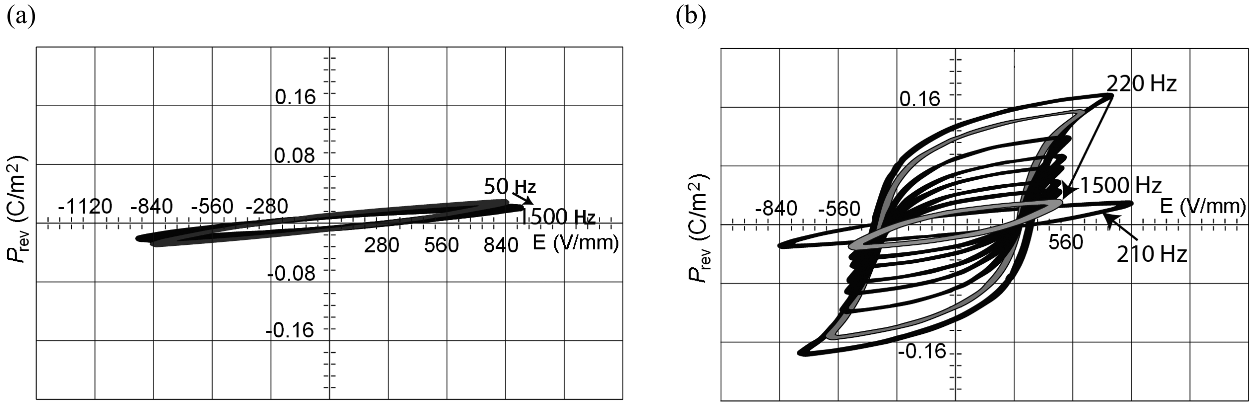

Fig. 1. Dielectric hysteresis loop evolution of PZT sample. (a) at the start time of field application of different frequencies; (b) maximal value. E_a = 850 V/mm. Vertical (280 V mm^{-1}/div) and horizontal (0.08 C·m^{-2}/div) scaling are the same for both (a) and (b) graphs.

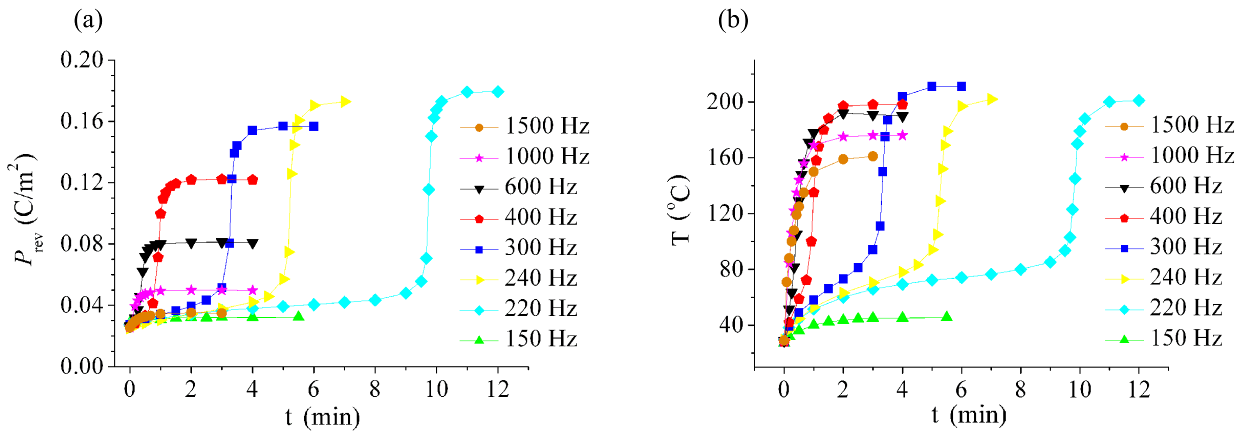

Fig. 2. Dependence of switched polarization (a) and heating temperature of PZT sample (b) on time during exposure electric fields of different frequencies. $E_a = 850$ V/mm.

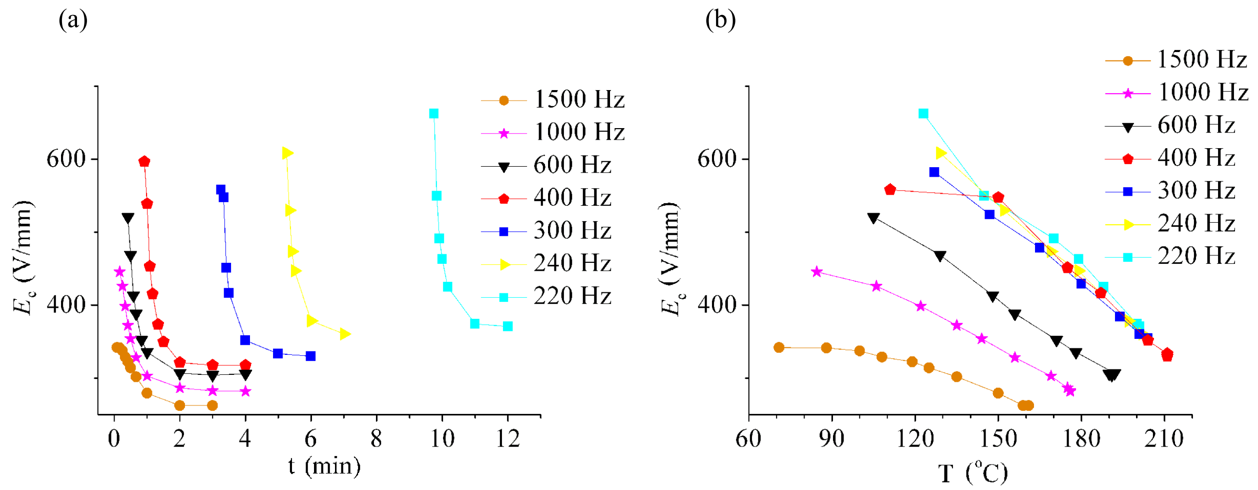

Fig. 3. Coercive field time (a) and temperature dependence of PZT sample; (b) during self-heating in electric field of different frequencies. $E_a = 850$ V/mm.

4. DISCUSSION

The experiments show that for saturated dielectric hysteresis loops the increase of the electric field frequency results in a decrease of reversible polarization values, which obeys the exponential rule (Fig. 4, curve 1). Approximation of experimental data making use of exponential regression with the aid of MathCad14 (Fig. 4, curve 2) resulted in an analytical expression for the frequency dependence of the polarization value P:

$$P = P_s \times \exp(-f \times \tau) + P_{irr}. \qquad (1)$$

Here P_s is the maximum possible value of reversible polarization in C/m^2, P_{irr} is the irreversible component polarization, τ is the time constant in s, characterizing

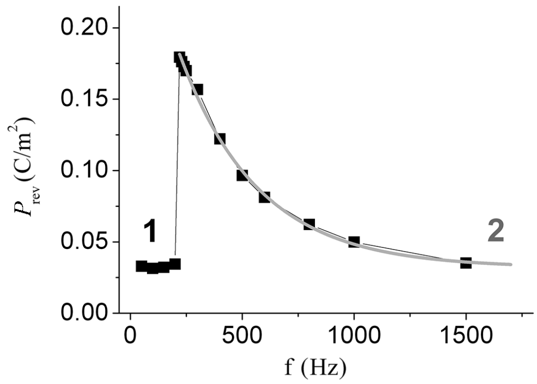

Fig. 4. Dependence of the switchable polarization value on frequency (curve 1 – experiment, curve 2 – calculation). $E_a = 840$ V/mm.

the exponential decay of polarization, and f is the frequency in Hz. The corresponding numerical data are $P_s = 0.28 \pm 0.01$ C/m^2; $\tau = 2.83 \pm 0.01$ s; $P_{irr} = 0.03 \pm 0.005$ C/m^2. The maximal experimentally observed P_{rev} has a value of $0.64 \times P_s$.

The temperature variation of the PZT ceramics sample due to electrocaloric effect for external electric field change of 1kV/mm is not more then 3 K [18]. So it may be concluded that the observed temperature changes of the sample during switching from 20 to 160 K are related to both dielectric and heat losses. Their mechanisms may be described in the following way:

$$EP = \frac{UQ}{dS} = \frac{UIt}{V} = \frac{Wt}{V} = \frac{W}{Vf} = \frac{Q_v}{f}, \quad (2)$$

where E is the intensity of applied field, P is the polarization calculated by dielectric hysteresis loop data, U is the generator voltage, d is the sample thickness, Q is the charge, S is the sample area, I is the switching current passing during the cycle time t, f is the measurement frequency of the AC field, and V is the sample volume. The quantity $Q_v = EPf$ is the dissipation power of a sample volume unit. In essence, this energy is responsible for heating the sample.

Figure 5 shows the dependence of heating rate (dT/dt) at the start time on Q_v. As can be seen, they have a linear dependence, regardless of whether measurements were taken at the same value of the AC field, but for different frequency (Fig. 5a), or at one frequency in different AC fields (Fig. 5b).

Based on this, we suggested the method of estimated volumetric heat capacity coefficient ($c_v = \rho \cdot c$) by the following equation [16]:

$$\frac{dT}{dt} = \frac{1}{\rho c} \cdot \frac{v}{V} \cdot Q_v. \quad (3)$$

Here V is the sample volume, ρ – density, c – specific heat. Coefficient $\frac{1}{c_v} \cdot \frac{v}{V}$ can be interpreted as the slope for linear function $dT/dt(Q_v)$, v/V is the ratio of the switchable sample volume (v) to the total volume (V). It can be expressed as the ratio of the reversal polarization (P_{rev}) to the maximum allowed (P_s). As shown earlier for switching of PZT ceramic we had $P_{rev}/P_s = v/V = 0.64$. Coefficient $\frac{1}{c_v} \cdot \frac{v}{V}$ was determined from the graphs (Fig. 5). As result, we obtain the value of the volumetric heat capacity of PZT ceramics for AC field with different frequencies (Fig. 5a): $c_v = (2.6 \pm 0.2) \cdot 10^6$ J/m^{3}K; with different amplitudes (Fig. 5b): $c_v = (2.4 \pm 0.2) \cdot 10^6$ J/m^{3}K. In the general case the volumetric heat capacity depends on temperature and, in accordance with the available data [19], c_v value for PZT are $2.25 \cdot 10^6$ (T = 27 °C) and $2.45 \cdot 10^6$ (T = 127 °C) J/m^{3}K. Since the sample temperature during the loop forming was raised to 200 °C (Fig. 2b) and depended on the frequency, it is obvious that spread of the c_v values can be related with the sample temperature fluctuations. Thus, the specific heat is less for the case of smaller AC field amplitude, which corresponds to a lower sample heating temperature.

5. CONCLUSIONS

Our experiments showed that the linear dependence of the self-heating temperature of the PZT ceramic samples (and, hence, the power of heat losses) on the electric field frequency is observed only at low-frequency region.

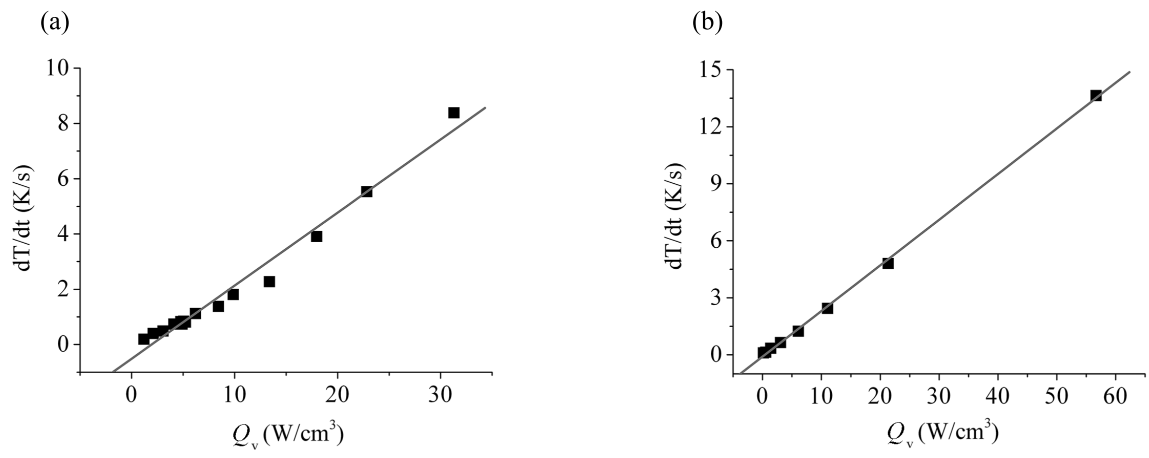

Fig. 5. Function dT/dt (Q_v) at start time of the AC field. Curve (a) for different frequencies (E_a = 840V/mm); (b) – for different amplitudes AC (f = 300 Hz).

This means that the application of formula (2), according to which the dissipated thermal power must linearly depend on the frequency, is limited. These restrictions are due to the motion of domain walls. This is confirmed by the fact that the maximum sample temperature change corresponds to a sharp change in the shape of dielectric hysteresis loop. Thus, the reason for the heat losses is the switching of the domain structure.

ACKNOWLEDGEMENT

The presented work was performed under the State Assignment №3.8032.2017/BP of the Ministry of Science and Education of Russia. The publication costs of this article were covered by the Estonian Academy of Sciences and the University of Tartu.

REFERENCES

1. Jha, A. R. *MEMS and Nanotechnology-Based Sensors and Devices for Communications, Medical and Aerospace Applications*. CRC Press., Baco Raton, 2008.
2. Jalili, N. and Afshari, M. *Piezoelectric-Based Vibration Control: From Macro to Micro, in: Nano Scale Systems*. Springer, New York, 2010.
3. Visvanathan, K. and Gianchandani, Y. B. Microheaters based on ultrasonic actuation of piezoceramic elements. *J. Micromech. Microeng.*, 2011, **21**, 085030 (10pp).
4. Uchino, K. and Giniewicz, J. R. *Micromechatronics*. Marcel Dekker, New York, 2003.
5. Tong, Yi-Z., Minghao, Z., and Pin, T. Fracture of piezoelectric ceramics. *Adv. Appl. Mech.*, 2002, **38**, 147–289.
6. Liu, G., Zhang, S., Jiang, W., and Cao, W. Losses in ferroelectric materials. *Mater. Sci. Eng. R Rep.*, 2015, **89**, 1–48.
7. Ueha, S., Tomikawa, Y., Kurosawa, M., and Nakamura, N. *Ultrasonic Motors: Theory and Applications*. Clarendon Press, Oxford, U.K., 1993.
8. Shigematsu, T., Kurosawa, M. K., and Asai, K. Nanometer stepping drives of surface acoustic wave motor. *IEEE Trans. Ultrason. Ferroelect. Freq. Control*, 2003, **50**, 376–385.
9. Senousy, M. S., Li, F. X., Mumford, D., Gadala, M., and Rajapakse, R. K. N. D. Thermo-electro-mechanical performance of piezoelectric stack actuators for fuel injector applications. *J. Intell. Mater. Syst. Struct.*, 2009, **20**, 387–399.
10. Harlow, J. H. *Electric Power Transformer Engineering*. CRC Press, Baco Raton, 2004.
11. Crowell, B. *Vibrations and Waves, in: Light and Matter online text series*. Fullerton, California, 2006.
12. Tooley, M. *Electronic Circuits: Fundamentals and Applications*. Elsevier, 2006.
13. Ikeda, T. *Fundamentals of Piezoelectricity*. Oxford University Press, 1996.
14. Zhang, Q. M., Wang, H., and Zhao, J. Effect of driving field and temperature on the response behavior of ferroelectric actuator and sensor materials. *J. Intell. Mater. Syst. Struct.*, 1995, **6**, 84–93.
15. Sakai, T. and Kawamoto, H. Durability properties of piezoelectric stack actuator. *Japan. J. Appl. Phys.*, 1998, **37**, 5338–5341.
16. Zheng, J., Takahashi, S., Yoshikawa, Sh., and Uchino, K. Heat generation in multilayer piezoelectric actuator. *J. Amer. Ceram. Soc.*, 1996, **79**, 3193–3198.
17. Malyshkina, O. V. and Eliseev, A. Yu. Power dissipation during dielectric loop evolution in PZT ceramics. *Ferroelectrics*, 2015, **480:1**, 10–15.
18. Shebanovs, L., Borman, K., Lawless, W. N., and Kalvane, A. Electrocaloric effect in some perovskite ferroelectric ceramics and multilayer capacitors. *Ferroelectrics*, 2002, **273:1**, 137–142.
19. Kallaev, S. N., Omarov, Z. M., Bilalov, A. R., Rabadanov, M. Kh., Sadykov, S. A., and Bormanis, K. Specific features of the thermal physical properties of relaxor ceramics based on lead zirconate titanate. *Phys. Solid State*, 2009, **51**, 1524–1526.

Lülitusprotsesside poolt ferroelektrilistes keraamikates tekitatud soojuskaod

Olga Malyshkina, Anton Eliseev ja Rostislav Grechishkin

On uuritud lülitusprotsesside poolt ferroelektrilistes keraamikates tekitatud soojuskadusid kõrge amplituudiga vahelduvas elektriväljas ja suures sageduste vahemikus 50–1500 Hz. On näidatud korrelatsiooni olemasolu ümberpolariseerumise ajaliste sõltuvuste ja isekuumenemise temperatuuri vahel. Toetudes eksperimentaalandmete lähendusele, on saadud analüütiline avaldis, mis kirjeldab vahelduva polarisatsiooni eksponentsiaalset kahanemist elektrivälja sageduse suurenemisel. On välja pakutud meetod, mis lubab hajunud soojuse põhjal hinnata volumeetrilise soojusmahtuvuse koefitsienti lülitusprotsessis. On näidatud, et isekuumenemise temperatuuri lineaarne sõltuvus elektrivälja sagedusest on täheldatav ainult madalate sageduste piirkonnas. See piirang ja samuti soojuskadude põhjus seostatakse elektrivälja vaheldumisel toimuva domeenide seinte liikumisega.