Journal of Optoelectronics and Advanced Materials Vol. 6, No. 1, March 2004, p. 341 - 344

FEATURES OF DIELECTRIC POLARISATION IN THE PSN-PT FERROELECTRIC CERAMICS

K. Bormanis*, A. I. Burkhanov[a], A. V. Shil′nikov[a], A. Sternberg, S. A. Satarov[a], A. Kalvane

Institute of Solid State Physics, University of Latvia, Riga, LV 1063, Latvia
[a]Volgograd State Architectural and Engineering Academy, 1 Academicheskaya st., Volgograd, 400074, Russia

Lead titanate solid solutions exhibit extreme values of certain physical parameters. We have investigated ferroelectric relaxor ceramics of (1-x)[$Pb(Sc_{1/2}Nb_{1/2})O_3$] - x[$PbTiO_3$] (PSN-PT) composition near the morphotropic phase boundary. Results are discussed taking account into the relaxor properties related to defects and disordering of the PSN-PT system.

(Received October 13, 2003; accepted after revision February 2, 2004)

Keywords: Ceramics, Ferroelectrics, Dielectric properties

1. Introduction

The (1-x)[$Pb(Sc_{1/2}Nb_{1/2})O_3$]-x[$PbTiO_3$] (PSN-PT) solid solution is interesting for fundamental research as well as practical applications. The PSN-PT system has a morphotropic phase boundary (MPB) between the rhombohedric and tetragonal phases at lead titanate concentration around 0.35~0.43 [1]. Possible existence of a monoclinic phase between the rhombohedric and tetragonal phases has been reported [2]. Appearance of such an intermediate phase in perovskite systems of $PbTiO_3$ has been considered theoretically [3]. The dielectric and piezoelectric parameters of $Pb(Mg_{1/3}Nb_{2/3})O_3$ - $PbTiO_3$ (PMN-PT) and PSN-PT ferroelectric solid solutions (FESS) are known to be extremely high near the MPB [4]. Besides, admixtures to the FESS allow to obtain optimum characteristics of the meterials. E.g., modifying of the PSN-PT ceramics with 0.3Fe_2O_3 increases mechanical quality (Q_m=297) by a factor of 4.4 [5], which allows to use the PSN-PT ceramics in powerful transducers and ultrasound drivers requiring high values of electromechanical coupling and piezoelectric coefficients.

Since PSN - one of the components of the FESS - is a disordered ferroelectric (relaxor), the low frequency and infra-low frequency studies [6] of a prospective material like PSN-PT seem to be of a high priority. The features dielectric response of the PSN-PT system to low and infra-low frequency signals studied in a wide interval of temperatures at different intensities of the measuring field are presently reported.

2. Samples and methods

The ferroelectric solid solution ceramics of 0.58PSN-0.42PT were prepared by conventional technologies at the Institute of Solid State Physics of the University of Latvia. The composition had a morphotropic phase boundary [7]. The samples were furnished with fired silver paste electrodes of the size 2 × 3 × 0.8 mm. The real ε'(T) and imaginary ε"(T) parts of the dielectric permeability at frequencies of 1, 10, 100, and 1000 Hz and field intensities that did not exceed 1 V/cm were measured on a bridge. At first the ε'(T) and ε"(T) curves were obtained on a one-year aged sample at heating

* Corresponding author: bormanis@cfi.lu.lv

from -170 °C up to 310 °C at the rate of one degree per minute. Effective dielectric permeability $\varepsilon'_{eff}=P/\varepsilon_o E_o$ and dielectric loss $\varepsilon''_{eff}=S/\varepsilon_o \pi E_0^2$ (here P is polarisation, E_o - the field amplitude at measuring polarisation loops, ε_o - dielectric constant, S - the area of the loop) were measured by a modified Sawyer-Tower circuit at different E_o and frequencies of 0.1, 1 and 10 Hz at heating from the temperature –190 °C (liquid nitrogen) up to +150 °C.

3. Results and discussion

The ε'(T) curves of 0.58PSN-0.42PT at frequencies of 1, 10, 100 and 1000 Hz are shown in Fig. 1a. No shift of the temperature T_m of the ε'(T) maximum is observed between 100 Hz and 1kHz (at both frequencies T_m≈290 °C), i.e., relaxor behaviour of the phase transition is absent. It is consistent with other data [5] showing absence of the dependence of T_m on frequency. An abrupt increase of ε'(T) seen in Fig. 1a at T≈160 °C at frequencies of 1 Hz and 10 Hz has been also observed by other authors at the same temperature at frequencies of 100 Hz and 1 kHz in 0.58PSN-0.42PT ceramics modified with Fe_2O_3 [5]. Thus, comparing with other data [5] allows to conclude that admixture of Fe_2O_3 to the 0.58PSN-0.42PT system does not lead to appearance of high-temperature relaxation of polarisation (HRP) merely displaying it at a lower temperature. In Fig. 1b the thermal behaviour of tgδ at different frequencies is shown in a logarithmic scale. An abrupt increase of tgδ(T) is seen to occur at T>20 °C, particularly pronounced at frequencies of 1Hz and 10 Hz. The high-temperature maximums of tgδ(T) at T≈270 °C, i.e., near T_m, are related to the transition from ferroelectric to paraelectric phase.

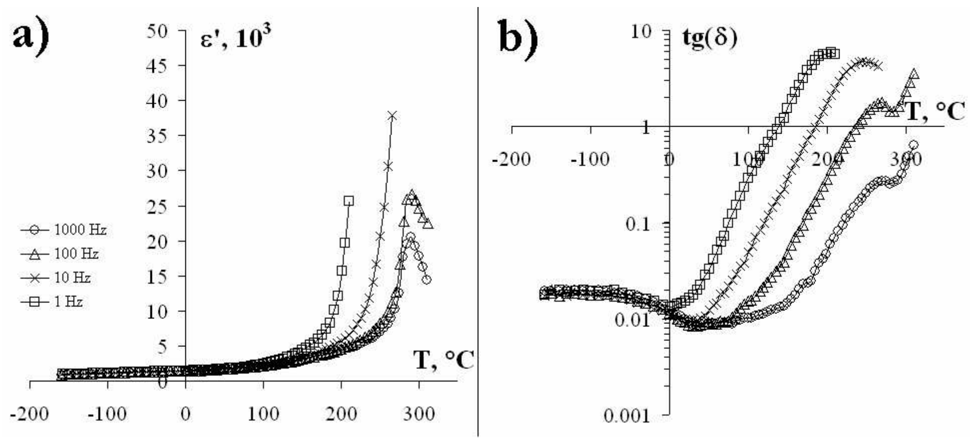

Fig. 1. Thermal behaviour of dielectric permeability ε'(T) - (a) and tgδ - (b) at frequencies 1, 10, 100 and 1000 Hz.

The high-temperature maximums on the tgδ(T) curve at frequency 10 Hz and 1Hz are observed at T≈245 °C and T≈205 °C, respectively. The activation energy calculated from the well known expression (e.g., [8]):

$$U_a = \kappa \frac{T_2 \cdot T_1}{T_2 - T_1} \ln(\frac{\nu_2}{\nu_1}) \quad (1)$$

where T_1, T_2 are temperatures of tgδ maximums at frequencies ν_1 and ν_2, respectively, is found to be U_a≈1.24 eV. The value is consistent with the conclusion [5] that oxygen vacancies provide a considerable contribution to properties of PSN-PT. Thus, the mentioned HRP of the material (Fig. 1a), as in case of the PZT ceramics [9], is due to presence of oxygen or lead vacancies. As seen in Fig. 1b, a very broad maximum of tgδ at T≈-110 °C is discerned at all frequencies in the low temperature interval between -150 °C and -20 °C where dependence of tgδ on the frequency in ultra-weak fields is negligible. The low-temperature relaxation of polarisation in PSN-PT is well pronounced in the behaviour of polarisation characteristics under fields of medium and strong intensity (Fig. 2).

Dependence of the remanent to maximum polarisation ratio P_r/P_{max} on temperature at frequency 0.1 Hz is shown in Fig. 2a. In this case the P_r/P_{max} ratio represents the normalised polarisation corresponding to every value of the amplitude E_o at the given temperature. As seen from Fig. 2a, the relaxation of the P_r/P_{max} ratio depending on the amplitude of the measuring signal is considerable within the interval from -110 °C to -20 °C. This relaxation most likely is related to the domain wall dynamics within the region of the morphotropic transition as, for example, observed earlier in the PMN-PT-PZ system at PT=0.35 [10].

The latter assumption is supported by the behaviour of $tg\delta_{eff}$ shown in Fig. 2b at different frequencies and E_o=11 kV/cm. At different frequencies the maximums of $tg\delta_{eff}(T)$ appear at different temperatures within the interval from -90 °C to -40 °C. The activation energy of switching polarisation estimated from (1) is $U_a \approx 0.78$ eV. which is much lower than activation energy of the high-temperature relaxation of polarisation mentioned above. The value of U_a is consistent with data of model ferroelectrics (e.g., TGS [11]) where it may be ascribed to mobility of the 180-degree domain boundaries.

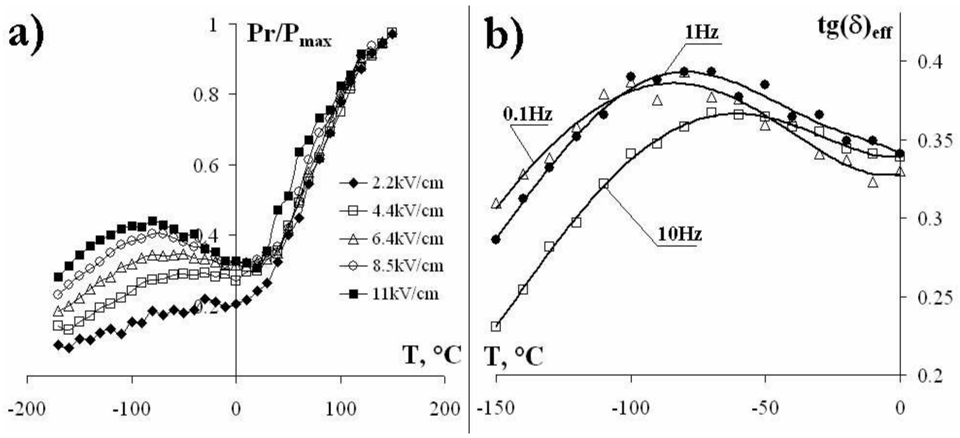

Fig. 2. Thermal behaviour of P_r/P_{max} ratio at frequency 0.1 Hz - (a) and $tg\delta_{eff}$ at frequencies 0.1, 1 and 10 Hz and field amplitude of 11 kV/cm - (b).

4. Conclusion

In piezoceramics $0.58Pb(Sc_{1/2}Nb_{1/2})O_3$-$0.42PbTiO_3$ was observed two types of low frequency relaxation of polarisation - high-temperature relaxation provided by oxygen vacancies, and low-temperature relaxation related to the domain wall dynamics in the region of the morphotropic transition.

Acknowledgements

The study has been supported by European Commission, "EXCELLENCE CENTRE OF ADVANCED MATERIAL RESEARCH AND TECHNOLOGY", Institute of Solid State Physics, University of Latvia; by the RFFI (grant #02-02-16232), the Ministry of Education of the Russian Federation (grant #E02-3.4-424), and by grant #202.03.02.04 of "Prioritary Science and Technology Research of Higher Education".

References

[1] Y.-H. Chen, K. Uchino, M. Shen, D. Viehland, J. Appl. Phys. **90**, 3 (2001).
[2] J-M. Kiat, Y. Uesu, B. Dkhil, M. X. Matsuda, C. Malibert, G. Calvarin, Cond-mat. **1**, 12 (2001).
[3] V. Ju. Topolov, A. V. Turik, Solid state physics (FTT) **44**, 7, 1295 (in Russian) (2002).
[4] M. Lejeume, J. P. Boilot, Mater. Res. Bull. **20**, 493 (1985).
[5] J. S. Kim, Y. H. Chen, K. Uchino // J. Kor. Phys. Soc. **32,** S1248 (1998).
[6] A. V. Shil'nikov, Proc. AS USSR, Ser. phys. **51**, 10, 1726 (in Russian) (1987).
[7] V. J. Tennery, K. W. Hang, R. E. Novak, J. Am. Ceram. Soc. **51**, 671 (1968).
[8] Ju. M. Poplavko, Physics of dielectrics, Vischa schkola, Kiiv (1980).
[9] V. I. Dimza, A. E. Krumin, Proceedings of University of Latvia **250**, 67 (1976).
[10] A. I. Burkhanov, A. V. Shil'nikov, S. A. Satarov, K. Bormanis, A. Sternberg, A. Kalvane, Proc. conf. "Intermatic - 2003", MIREA, Moscow, 2003. p. 96-99.
[11] A. V. Shil'nikov, L. G. Bulatova, E. S. Popov, L. A. Vasiljeva, L. I. Doncova, N. A. Tichomirova, L. I. Tserkasova, Proc. "Physics of Dielectrics and semiconductors" 41, Volgograd, (1978).