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CORRELATIONS BETWEEN FATIGUE AND VARIATIONS IN MATERIAL CONSTANTS OF PLZT CERAMICS

KORELACIJE MED UTRUJANJEM IN SPREMEMBAMI MATERIALNIH KONSTANT PLZT-KERAMIKE

ADRIJAN LEVSTIK, V. BOBNAR, Z. KUTNJAK, M. KOSEC
Jožef Stefan Institute, P.O. Box 3000, 1001 Ljubljana, Slovenia

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Correlations between fatigue and material constants of bulk 8/65/35 PLZT ceramics were investigated by a simultaneous measurement of fatigue and piezoelectric resonance as a function of switching cycles. It was found that as remanent polarization, also the real and imaginary parts of the complex dielectric and piezoelectric constants decrease with increasing number of switching cycles. However, real and imaginary parts of the complex elastic compliance and the electrical, mechanical, and electromechanical quality factors remain unchanged during fatigue, which suggests that there is no correlation between the fatigue and the mechanical degradation of the sample.

Key words: PLZT, fatigue, piezoelectric resonance, electromechanical properties

Korelacije med utrujanjem in materialnimi konstantami 8/65/35 PLZT-keramike so bile raziskane s simultano meritvijo utrujanja in odvisnosti piezoelektrične resonance od števila preklopnih ciklov. Enako odvisnost od števila preklopnih ciklov kot remanentna polarizacija imajo tudi realni in imaginarni deli kompleksnih dielektrične in piezoelektrične konstante. Realni in imaginarni del kompleksne elastične konstante ter električni, mehanski in piezoelektrični faktor kvalitete pa se med utrujanjem ne spreminjajo, kar pomeni, da med utrujanjem ni prišlo do mehanske degradacije vzorca.

Ključne besede: PLZT, utrujanje, piezoelektrična resonanca, elektromehanske lastnosti

1 INTRODUCTION

Ferroelectric switching in thin films and bulk ferroelectric ceramics enables the application in memory elements and electrographic printing[1]. One of the material characteristics playing an important role in this application is fatigue - decreasing of the remanent polarization with increasing number of switching cycles. While the shape of the dielectric hysteresis loop and the value of the remanent polarization influence the quality of printing characteristics, the fatigue effect constrains the maximum number of rewriting cycles in PLZT material. Fatigue and its consequences for commercial applications of ferroelectrics have been recently extensively studied[2-5]. Electromechanical properties such as elastic, piezoelectric, and dielectric constants have been studied by the piezoelectric resonance method in various ceramic materials[6-8], however, their dependence on fatigue has not yet been reported.

In this work we present measurements of the fatigue and piezoelectric properties of 8/65/35 PLZT ceramics carried out simultaneously in order to assure equal conditions during cycling procedure for all measuring quantities. Such an experiment makes possible to identify existence of correlations between fatigue and variations in material constants.

2 EXPERIMENTAL METHODS

Bulk 8/65/35 PLZT ceramics used in these studies was prepared as described elsewhere[9]. The sample was cycled with an a.c. electric field at the frequency of 50 Hz and the amplitude of 15 kV/cm. During cycling, hysteresis loops were measured by Sawyer-Tower technique. After each measurement of the hysteresis loop, the sample was poled by the d.c. electric field of 15 kV/cm. Then, the piezoelectric resonance was studied by measurements of admittance $|Y|$ and phase angle θ in the frequency interval 1kHz - 1MHz.

From piezoelectric resonance data complex piezoelectric and dielectric constants and elastic compliance can be determined, since the admittance of the piezoelectric bar resonator is a function of the complex dielectric $\varepsilon_{33}^* = \varepsilon_{33}' - \varepsilon_{33}''$ and piezoelectric $d_{31}^* = d_{31}' - d_{31}''$ constants and complex elastic compliance $s_{11}^* = s_{11}' - s_{11}''$. The treatment of a piezoelectric bar resonator can be in the vicinity of the resonant frequency simplified by using the equivalent electric circuit, which was introduced by Damjanović[8]. Following his approach, the parameters of the equivalent circuit R, C, L, C_0, R_2, and x were determined by simultaneous fitting the experimental data to the expressions for the conductance $G = |Y|\cos\theta$ and susceptance $B = |Y|\sin\theta$ of the equivalent electrical circuit, which are expressed as

$$G = \frac{\omega^2 RC^2}{(1-\omega^2 LC)^2 + \omega^2 R^2 C^2} - \\ - x\frac{\omega C - \omega^3 LC^2}{(1-\omega^2 LC)^2 + \omega^2 R^2 C^2} + \frac{1}{R_2} \tag{1}$$

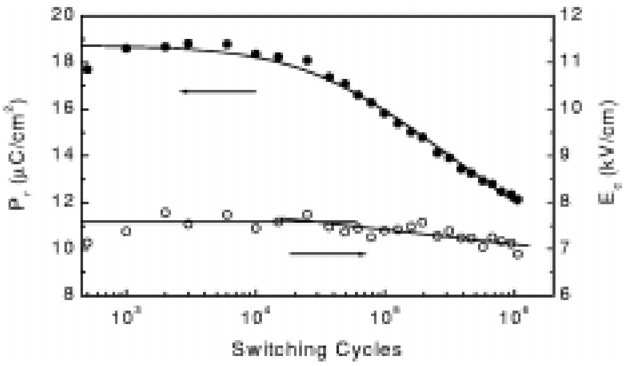

Figure 1: The remanent polarization P_r and the coercive field E_c versus switching cycles. Solid lines are guides to the eye

Slika 1: Remanentna polarizacija P_r in koercitivno polje E_c kot funkciji preklopnih ciklov. Polne krivulje so vodilo očem

$$B = \frac{\omega C - \omega^3 LC^2}{(1-\omega^2 LC)^2 + \omega^2 R^2 C^2} + $$
$$+ x\frac{\omega^2 RC^2}{(1-\omega^2 LC)^2 + \omega^2 R^2 C^2} + \omega C_0 \qquad (2)$$

The set of complex material constants was then calculated from the parameters of the equivalent circuit.

3 RESULTS

The ferroelectric fatigue is shown in **Figure 1**. The remanent polarization P_r decreases steeply above 2 x 10^4 switching cycles, above which also the coercive field E_c shows a weak change in the slope.

Figure 2 shows experimentally determined conductance G and susceptance B in a complex plane and the results of the simultaneous fitting to eqs. (1) and (2) at four different numbers of switching cycles. The radius of the circles starts to decrease rapidly after 10^4 switching cycles, indicating suppression of the piezoelectric resonance amplitude which occurs together with the decreasing of the remanent polarization.

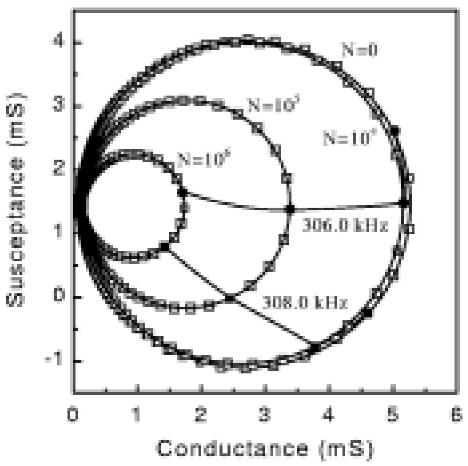

Figure 2: Susceptance B versus conductance G obtained at four different numbers of switching cycles. Dots represent experimental points. Solid curves represent fits to eqs (1) and (2)

Slika 2: Susceptanca B kot funkcija prevodnosti G pri štirih različnih vrednostih preklopnih ciklov. Točke predstavljajo eksperimentalne rezultate. Polne krivulje so rezultat prilagajanja enačb (1) in (2)

The real and imaginary parts of the complex material constants as a function of switching cycles are shown in **Figure 3**. At the same number of switching cycles as in the case of the remanent polarization dependence, also real and imaginary parts of the complex dielectric and piezoelectric constants start to decrease. Contrary to this, the elastic compliance is nearly independent on switching cycles. A similar behavior of the piezoelectric constant d_{33} was recently observed in PZT thin films[5]. As can be seen in **Figure 3**, the electrical $Q_e = \varepsilon_{33}'/\varepsilon_{33}''$, mechanical $Q_m = s_{11}'/s_{11}''$, and electromechanical $Q_{me} = d_{31}'/d_{31}''$ quality factors are all independent on switching cycles. Namely, the real part of the particular constant has the same dependence on the increasing number of switching cycles as its imaginary part. This suggests that there is no correlation between the fatigue and the mechanical degradation of the sample, like for instance the

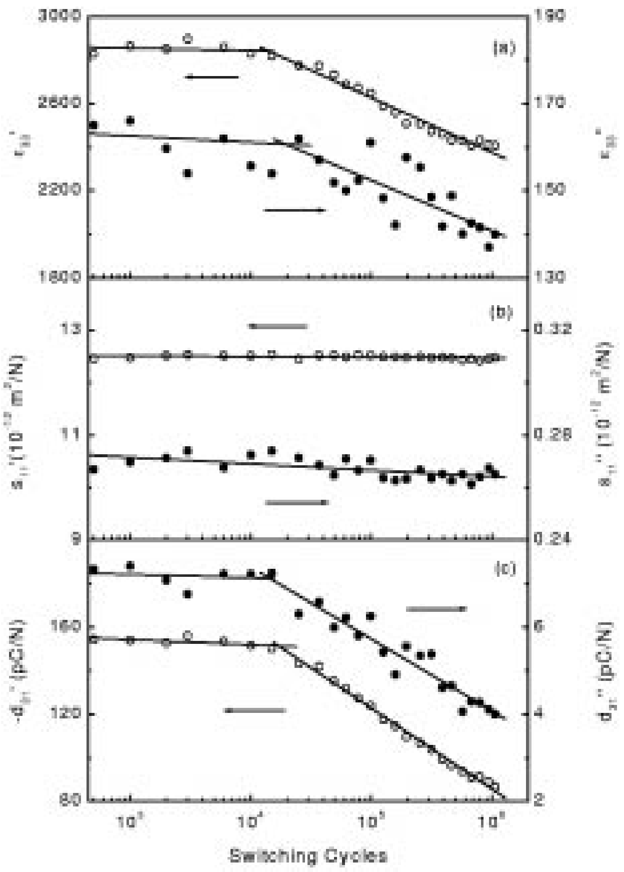

Figure 3: Various material properties as a function of switching cycles: (a) the real ε_{33}' and imaginary ε_{33}'' parts of the complex dielectric constant, (b) elastic compliances s_{11}' and s_{11}'' and (c) piezoelectric constants d_{31}' and d_{31}''. Solid lines are guides to the eye

Slika 3: Materialne konstante v odvisnosti od števila preklopnih ciklov: (a) realni ε_{33}' in imaginarni ε_{33}'' del kompleksne dielektrične konstante, (b) elastični konstanti s_{11}' and s_{11}'' in (c) piezoelectrični konstanti d_{31}' in d_{31}''. Polne krivulje so vodilo očem appearance of microcracks. It is interesting to note that the thermal annealing of the fatigued sample at 590°C nearly completely restored the remanent polarization to its initial value, pointing out that the pinning of domain walls due to entrapment of space charges at the electrode or domain interface is most likely responsible for the fatigue[4].

The simultaneous measurement of piezoelectric properties and the fatigue has an important advantage in comparison to the case where measurements are performed in separate runs on different samples. Separate measurements would introduce an additional data scattering due to the known problems with cleaning procedure and electrode deposition, but the most important problem is onshelf aging - variations in dielectric properties with time in zero-field conditions. This is particularly important in PLZT ceramics where decrease in the dielectric constant for more than 20% can be observed within few hours after annealing. This phenomenon makes almost impossible to bring two different (or one annealed) samples in exactly the same state since their histories will never be exactly the same after thermal annealing.

4 CONCLUSIONS

The fatigue in bulk 8/65/35 PLZT ceramics reflects itself only in dielectric and piezoelectric properties, but not in elastic compliances. On the basis that the electrical, electromechanical and mechanical quality factors are independent on switching cycles, it was concluded that there is no correlation between the ferroelectric fatigue and the mechanical degradation of the sample.

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