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JECS-10989; No. of Pages 9
ARTICLE IN PRESS
Journal of the European Ceramic Society xxx (2016) xxx–xxx
Contents lists available at www.sciencedirect.com
Journal of the European Ceramic Society
journal homepage: www.elsevier.com/locate/jeurceramsoc

Electrical fatigue behavior of lead zirconate titanate ceramic under elevated temperatures

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ARTICLE INFO

Article history:
Received 29 September 2016
Received in revised form
19 December 2016
Accepted 21 December 2016
Available online xxx

Keywords:
Electrical fatigue
Temperature effect
PZT

ABSTRACT

The electrical fatigue behavior of lead zirconate titanate (PZT) ceramics is investigated under different temperatures. A bipolar triangular electric field with the amplitude of ±1.5 kV/mm and the frequency of 50 Hz is applied to samples up to 1×10^6 cycles. The fatigue rate is found to be temperature dependent, and the fatigue degradation is represented by the loss of remnant polarization, dielectric constant, and piezoelectric constant increased with loading cycle numbers. The degradation, involving surface damage and crack propagation, is more pronounced in samples cycled at lower temperatures, and increases with increasing number of cycles. The temperature effect on fatigue degradation of the properties is described based on the field shielding effect caused by surface damage and fatigue-induced cracks. The effect is more dominant in case of higher cycling numbers and lower temperature fatigue due to higher strain mismatch between switchable and non-switchable domains. Moreover, Raman spectroscopy is used to determine the influence of fatigue on the ferroelectric domains in different areas of the specimens.

1. Introduction

Lead zirconate titanate (PZT) ceramics are widely used for a number of applications such as actuators, sensors, and transducers [1–3]. Their performances depend on the ability of ferroelectric domains to switch under an applied electric field. For long term service, a decay of ferroelectric polarization and strain (i.e. strain induced by ferroelectric domain switching) during the application of cyclic loading usually occurs; this phenomenon is called ferroelectric fatigue [4–7]. Ferroelectric fatigue is caused by a decrease in domain switchability, which can originate from (a) field shielding due to mechanically damaged near-electrode surfaces [8,9] and (b) the pinning effect due to the accumulation of charged defects, i.e. oxygen vacancies, captured at domain walls [10–12], which is widely known to be one of the prominent effects causing ferroelectric fatigue [10,13,14]. Many researchers have reported the temperature dependence of ferroelectric properties [15–18]. They found that polarization and strain induced by an external applied field are sensitive to temperature; in the case of applied bipolar fields, both polarization and strain decrease with increasing temperature, whereas the opposite effect occurs under unipolar fields. Although the temperature dependence of the ferroelectric properties was widely studied [12,19–22], the effect of temperature on the fatigue behavior of bulk ferroelectric ceramics – especially concerning the relationship with the ferroelectric domain texture – is not completely understood. Therefore, in this study, the effect of temperature on the bipolar fatigue behavior of PZT ceramics is investigated. Moreover, Raman spectroscopy is used to determine the ferroelectric domain orientation distribution in order to link domain pinning with fatigue.

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http://dx.doi.org/10.1016/j.jeurceramsoc.2016.12.037

Please cite this article in press as: M. Promsawat, et al., Electrical fatigue behavior of lead zirconate titanate ceramic under elevated temperatures, J Eur Ceram Soc (2016), http://dx.doi.org/10.1016/j.jeurceramsoc.2016.12.037

2. Experimental procedure

2.1. Electrical fatigue

Cylinder-shaped commercial soft lead zirconate titanate (PZT) samples (Thales Underwater Systems, Australia) with a diameter of 6 mm and 20 mm length and an average grain size of 5 μm were used in this study. The samples were cut into disc shape and then a small portion of the curved edge of the discs was cut in order to obtain a flat surface. This flat edge surface was prepared to study field shielding effects using SEM and Raman scattering. The disc and flat edge surfaces were polished to the final dimension of 6 mm diameter and 1 mm thickness by using 800, 1000, 1500 and 4000 grit SiC papers, sequentially. The sketch of sample preparation is shown in Fig. 1. The well-polished samples were annealed above the Curie temperature (T_C = 350 °C) at 500 °C for 5 h to remove the residual stresses arising from the polishing process. The electrode was deposited on the parallel surfaces of the samples with a colloidal silver paste. Samples were poled under an applied electric field of 1.7 kV/mm at 120 °C for 10 min. The piezoelectric constant (d_{33}) was measured on the poled samples 24 h after the poling process by a d_{33} meter (ZJ-6B). The relative permittivity (ε_r) was measured by a LCR meter (GW, Instek). Previous neutron experiments have confirmed the tetragonal structure of samples from this batch [23]. To investigate polarization fatigue behavior, samples were immersed in silicon oil and a bipolar sinusoidal electrical cyclic load up to ±1.5 kV/mm and at the frequency of 10 Hz was applied across the electrode surfaces up to 10^6 cycles using a high voltage amplifier (20/20 Trek). Fatigue tests were operated at the temperatures of 25, 50, 100, 150 and 200 °C. For each fatigue temperature, three samples were used and the results obtained from all samples were averaged. During fatigue tests, polarization hysteresis loops were measured using a Sawyer-Tower circuit. To determine a development of fatigue surface damages and cracks, the microstructure of the cross-sectional surfaces of fatigued samples were observed by scanning electron microscope (JEOL, JSM-6010LV).

2.2. Raman characterization

To reveal the effect of temperature and electric field cycling on the local domain texture nearby the surfaces, a high spatial resolution polarized Raman spectrometer (Horiba Jobin Yvon LabRAM HR-800) with nominal 42 mW Ar-laser source and 514 nm wavelength was employed. For each fatigue temperature, one sample was employed to characterize Raman spectra. The laser beam spot size is 1 μm on the sample's surface. The backscattered Raman spectra of poled non-fatigued samples, fatigued samples at room temperature and at 150 °C were collected at different areas of each sample, namely close to one electrode and far away from the electrode (i.e. half-way through sample thickness), as illustrated in Fig. 1. Both the incident light on the sample and the backscattered Raman light were polarized using linear polarization filters choosing a parallel polarized configuration. Raw Raman spectra were initially normalized with reference to the integrated intensity of the whole spectrum, and then fitted with commercial software (Labspec 4.02, Horiba Jobin Yvon) using a combination of Gaussian-Lorentzian peak functions. The concept of angular dependence between the domain orientation and Raman intensity was applied to determine the domain orientation on the lateral surface (i.e. the surface parallel to the applied electric field). For this experiment, the normalized intensity of the A_1 mode around 225 cm^{-1} peak at rotation angles of 0° to 180° was used to investigate the effect of fatigue and temperature on the domain orientation of the PZT samples. Details of the measurement concept and setup can be found in previous contributions by Deluca et al. [24,25] and Pojprapai et al. [26]. In order to reconstruct the local ferroelectric domain orientation distribution, a Reverse Monte Carlo (RMC) procedure has been used to best fit the experimental Raman data according to the procedure described in Röhrig et al. [27].

3. Results and discussion

3.1. Influence of fatigue and temperature on ferroelectric, dielectric and piezoelectric properties

Polarization-electric field (*P*–*E*) curves measured at different electrical loading cycles of the samples fatigued at temperatures of 25, 50, 100, 150 and 200 °C are shown in Fig. 2(a)–(e), respectively. The figures show saturated, highly temperature dependent *P*–*E* hysteresis loops, in accord with our previous works [28,29]. It can be seen from the result that *P*–*E* hysteresis loops are dependent on both temperature and number of cycles. At 1 × 10^3 cycles, remnant polarization (P_r) and coercive field (E_c) decrease with increasing fatigue temperature, i.e. the P_r and E_c measured at 25 °C are about 38 μC/cm^2 and 11 kV/cm, respectively, while those measured at 200 °C are about 20 μC/cm^2 and 6 kV/cm, respectively. For the samples fatigued at ≤ 100 °C, the *P*–*E* loop shapes undergo a large shape change with the cycle number. In particular, after fatigue testing (at 10^6 cycles) there is a strong decrease in P_r and an increase in

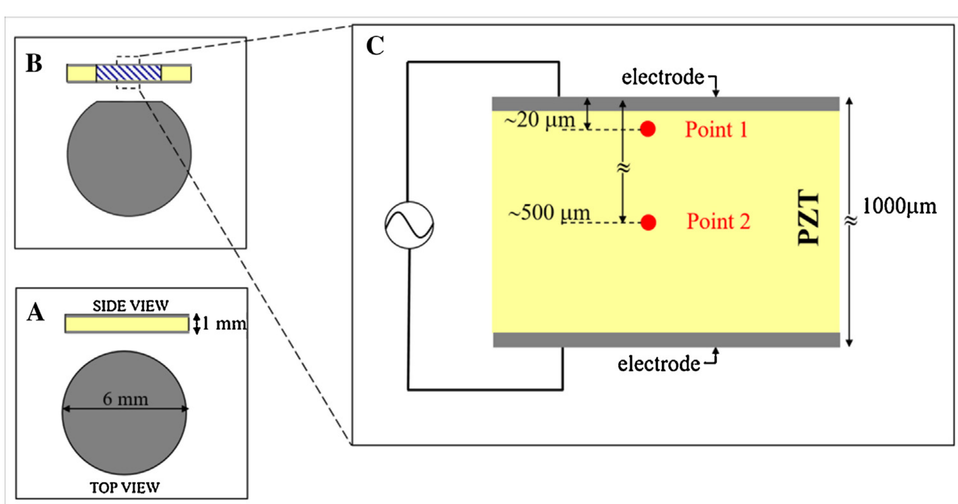

Fig. 1. The sketches of sample preparation (a) before and (b) after edge cut. (c) the location of Raman analysis (on the flat edge).