



Current leakage and transients in ferroelectric ceramics under high humidity conditions

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ABSTRACT

Lead zirconate titanate (PZT) is widely used in electromechanical devices such as ultrasonic transducers, sensors, ultrasonic motors, actuators and resonators. In harsh operating environments such as high humidity and high temperature the electrical resistance of the ceramic can decrease resulting in leakage currents. These can have a significant impact on device performance, particularly for low power applications. In this paper, the increase in leakage current is investigated and characterised under high humidity conditions. It is observed that the leakage current does not evolve smoothly over time, but is characterised by current transients. Detailed measurements of these current transients are reported, and their relationship to the conduction processes in the ceramic is investigated. A mechanism of conductive channel formation is proposed to account for the leakage current increase and current transients.

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1. Introduction

Lead zirconate titanate (PZT) ceramics are widely used in the manufacture of actuators in a range of devices including security locks, valves for fluid control, and diesel injector valves. The development of these applications in recent years has brought with it increased demands on performance and reliability under high electrical fields and in humid environments. Recent work [1] has shown that electromigration is the probable cause of leakage currents under d.c. bias in humid conditions, and a model mechanism based on the evolution of conducting pathways was proposed. Other work [2] has shown how the conduction mechanism may be related to the micro-structure and porosity of the ceramic.

Undoped PZT is known as a wide band gap semiconductor with a band gap of between 2.6 eV and 3.5 eV [3]. It also exhibits p-type conductivity due to the presence of low valence impurities substituting for higher valence Pb ions. This causes A-site (Pb ion) vacancies which act as electron acceptors leading to the production of holes [4]. This means that a Schottky barrier can be formed with the electrode material which can promote conduction in thin films [3,5]. Tunnelling can also be a mechanism for conduction in very thin films [5]. However, for films thicker than a few tens of nm, conduction is determined by the bulk of the material through Poole–Frenkel emission, where thermally excited electrons are assisted by the electric field into the conduction band [3]. The large piezoelectric coefficients in this class of “soft” PZT materials is

achieved by A-site vacancy substitution (e.g. La doping) which also confers high electrical resistivity [4]. Data from Ref. [4] would indicate a volume resistivity of $10^9 \Omega \text{ cm}$ at the temperature reported here. For the sample size and field strength used in this study we would expect a resistance of over $300 \text{ G}\Omega$ which, and a leakage current of less than 1 nA. The effects studied here are in the μA to mA range, so cannot be explained by these electronic conduction mechanisms.

There is evidence that the conduction mechanism in humid environments is ionic in character, including the observation of anodic breakdown features and sensitivity of the leakage current to the relative humidity and electrode material [1]. Measurements performed by the authors indicate that there is no significant (in this context <250 nA is considered insignificant) leakage current at the temperature of the measurements reported here (55 °C dry atmosphere). In fact a reduction of leakage current is usually observed on heating to this temperature from room temperature due to the expulsion of moisture.

Ionic silver migration from silver electrodes is commonly implicated in humidity-related conduction in otherwise insulating materials such as thick film insulators [6,7]. The proposed mechanism involves ionization of water at the silver cathode followed by release of silver ions which migrate under the applied electrical field. A similar mechanism has also been proposed previously for electrical changes in PZT with Ni electrodes [1,2]. Although an electromigration model explained the observed leakage currents, these studies did not present micro-structural details of the proposed conducting channels. The reason for this is that the amount of conducting material required to account for the observed conductivity is extremely small. The difficulty in detecting this means

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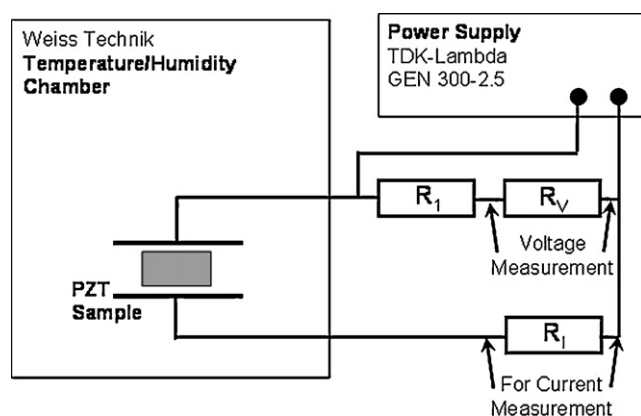


Fig. 1. Experimental arrangement.

indirect methods need to be used to obtain information on the conduction mechanism in PZT ceramics. Significant useful information can be obtained from the electrical characteristics of the leakage current. Lipscomb et al. [1] analysed the general trends in the evolution of the leakage current, but did not investigate the detailed short timescale features.

This study presents detailed time-resolved measurements of the evolution of humidity-induced leakage current in nickel electroded PZT devices. The measurements show significant fluctuations in the leakage current under a steady applied d.c. voltage. The detailed characteristics of the current transients are reported, and analysed statistically in time and frequency domains. It is shown that the current transients behave as a switched resistance-capacitance (RC) network, and a simple network model provides good agreement with experimental results. The evolution of the current transients with time and the switched RC characteristic support the proposal of an electromigration/breakdown mechanism for the leakage current in this type of device.

2. Experimental arrangement

Commercially available PZT samples are investigated. The samples are $2.5\text{ cm} \times 0.5\text{ cm} \times 0.015\text{ cm}$ and have nickel electrodes on both sides of the larger surface area. The experimental arrangement is shown in Fig. 1, which illustrates how samples are connected in the electrical circuit.

A 240 V d.c. potential difference is supplied by the power supply. Working voltages up to a maximum of 2 kV/mm are typical for bulk and multilayer piezoelectric actuators in applications such as valves and piezoelectric diesel injectors [7]. This is rather lower than the field strengths that can be sustained in thin film devices due to the scaling up of breakdown field strength with decreasing film thickness. A value of 250 V (1.7 kV mm^{-1}) was chosen as a typical voltage for many applications. The effects of varying the applied field on leakage current and lifetime have been reported previously [1]. The current through the sample and the voltage across the sam-

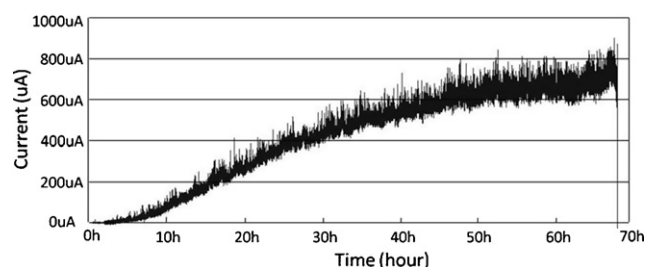


Fig. 3. Leakage current of PZT sample under 55 °C, 90 RH 240V d.c.

ple are calculated by measuring the voltage across R_V and R_1 . The resistor values are: $R_1 = 10\text{ M}\Omega$, $R_V = 20\text{ k}\Omega$ and $R_1 = 20\text{ k}\Omega$ (to a 2% error value).

The system noise on the current measurement under ambient conditions has been evaluated by using test resistor samples instead of PZT samples. Three different test resistor samples of $0.5\text{ M}\Omega$, $1\text{ M}\Omega$ and $2\text{ M}\Omega$ were used and the current behaviour measured with the 240 V d.c. applied. These current measurements were taken over a 10 s period with a sampling rate of 1 ms. Plots of the probability density function of particular current values for the three test resistor samples are given in Fig. 2.

It is found that the measurement resolution for the $0.5\text{ M}\Omega$ test resistor is $4\text{ }\mu\text{A}$, for the $1\text{ M}\Omega$ test resistor is $2\text{ }\mu\text{A}$ and for the $2\text{ M}\Omega$ test resistor is $1\text{ }\mu\text{A}$. It can be seen that, if the resistance of the samples is from $0.5\text{ M}\Omega$ to $2\text{ M}\Omega$, the system noise is within $\pm 2\text{ }\mu\text{A}$.

Fig. 1 also shows the PZT ceramic in the temperature/humidity chamber. The PZT was stressed by subjecting it to a harsh environment of 55 °C, 90% relative humidity (RH) and a 240 V d.c. bias. Voltage and current measurements were made over a 68 h period. Measurements were taken every 1 ms for the long-term current leakage profile investigation and every $1\text{ }\mu\text{s}$ for detailed data investigation. Voltage recordings (not presented here) confirmed that there were no significant fluctuations in the applied voltage.

3. Results

A typical result of the leakage current is presented in Fig. 3, where measurements are taken every 1 ms for 68 h.

The leakage current is observed to increase from the noise level of $2\text{ }\mu\text{A}$ to approximately $700\text{ }\mu\text{A}$. It can be seen in Fig. 3 that there is an initial period of up to 3 h when the leakage current remains at approximately the initial level. After this period the leakage current accelerated increases to $\sim 400\text{ }\mu\text{A}$ within the first 30 h. Thereafter, the leakage current approaches towards the final value of $\sim 700\text{ }\mu\text{A}$. It can be seen that over the 68 h period the leakage current becomes highly variable with the appearance of the current fluctuations. These fluctuations are significantly larger than the noise level reported in Fig. 2 and are therefore attributed to real current transients through the sample.

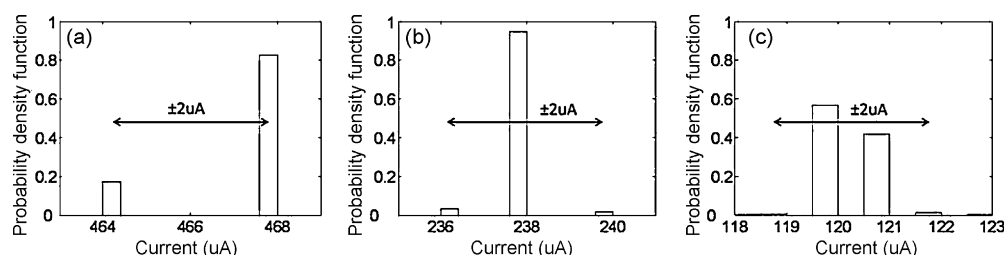


Fig. 2. System noise evaluations with: (a) $0.5\text{ M}\Omega$ resistor sample; (b) $1\text{ M}\Omega$ resistor sample; (c) $2\text{ M}\Omega$ resistor sample.

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