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Micro-computer tomography—An aid in the investigation of structural changes in lead zirconate titanate ceramics after temperature-humidity bias testing

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Abstract Lead zirconate titanate ceramic actuators are extensively used in electronic and electro-mechanical devices. Under humid conditions with a d.c. bias leakage currents increase power consumption In some materials the increasing leakage current is accompanied by the evolution of features on the anode surface, but little is known of any changes to the internal structure of the material. This research applies an advanced imaging technique (micro-computer tomography) to non-destructively investigate these features. This allows the evolution of internal features over time to be studied. The findings reveal that the microstructure of the ceramic was significantly altered during environmental testing.

Keywords Lead–titanate–zirconate · PZT · Micro computer tomography · Voids · Breakdown · Temperature humidity bias testing

1 Introduction

The many advantages of piezo-ceramic actuators have led to their widespread use in a variety of applications including valves, diesel injectors or door locks. A key advantage of the use of piezo-electric technology in these applications is the extremely low power consumption that can be achieved compared to electromagnetic technology. However humid environments can cause leakage currents in lead zirconate titanates (PZT) actuators [1, 2], possibly related to the internal structure of the ceramic. It is also well documented that repeated mechanical and electrical cycling of PZT ceramics results in irreversible structural change and performance loss [3, 4]. Conventional techniques for investigating such structural changes such as scanning electron microscopy require internal features to be sectioned prior to examination. This destroys the features being studied, making it impossible to study the evolution of structural changes over time. Here we report for the first time the application of micro computer tomography (μCT) to non-destructively observe internal void and crack formation due to temperature–humidity bias testing within PZT ceramics.

CT [5, 6] is a non-invasive, non-destructive visualization method which has been applied in a wide range of biomedical [6] and industrial [7] applications. In CT scanning X or γ rays are emitted from a source and pass through the sample to a detector. The intensity of the radiation signal on the detector is dependent on the linear attenuation characteristics of the sample material and the distance from the source to the detector. This variation in attenuation through a sample is defined as its profile and this can be reconstructed to form a single 'slice' through the sample material. In CT many such slices are taken at different orientations around the subject. This large amount of data can then be manipulated by the application of image reconstruction algorithms until a 3 dimensional image of the sample is produced [8].

Although this technique is highly useful in the assessment of fault and crack formation within structures [9] there has been no attempt to apply μCT to visualize piezo- ceramics. This is due to the high lead content within such materials,

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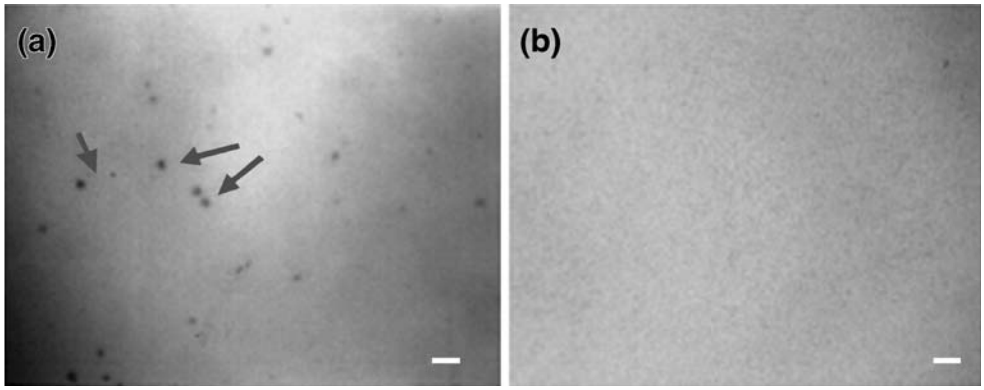

Fig. 1 Photomacrographs of a ceramic sample after environmental testing and etching. The figures show that after testing at 350 V (**a**) there are a large number of discolorations (*arrows*) apparent compared to a control sample (**b**) where no such discolorations are present. *Bar*=100 μm

which limits the penetration of the X- rays to a few millimetres. However, for the small parts that are the subject of this paper (typically 0.15 mm thick), this limitation was less of a problem, and parts could be tested that were representative of dimensions used in production devices.

The research presented here shows how such a technique can show internal structures and changes in these structures during environmental testing. The technique is applied here to a study of the formation of features on the electrodes and within PZT samples subjected to dc electrical fields in conditions of high humidity.

2 Methods

The samples tested are a range of commercially available piezo-electric, soft PZT ceramic. These are 5 mm×24 mm×160 μm thick slices with nickel electrodes on both sides of the ceramic. Samples were placed into an environmental chamber (Weiss-Gallenkamp 350 SB) and a 250 V direct current bias applied via gold sprung contacts. The temperature is fixed at 55 °C and the relative humidity set at 85%. In order to prevent failure by condensation the samples are allowed to acclimatize for 30 min before the d.c. bias is applied. Samples were removed and examined after the resistance of the ceramic had dropped to below 1 MΩ. the progression of the changes in electrical properties is described in more detail elsewhere [10].

Samples were examined for surface and internal defects by the application of brightfield macroscopy (BFM), environmental scanning electron microscopy (Philips XL30 ESEM), and μCT (Xtek, Benchtop CT 160Xi).

The μCT procedure applied using a tungsten filament, with the current and accelerating voltage parameters set at 105 μA and 160 kV respectively. The μCT took approximately 1000 slices of each sample (n=6) and reconstruction and analysis was performed on a separate computer. The whole process took approximately 3 h. The μCT images were 3D reconstructed however for greater clarity of features the subsequent figures show reconstructed 2D slices through the ceramic.

Etching to remove the nickel electrode was necessary for BFM and ESEM to allow the visualization of discoloured regions. Where the electrode was etched this was done by the application of 35% Nitric acid for 5 min followed by 3 rinses in distilled water. Etching was not required for the samples studied using μCT.

Initial samples were attached to an alumina substrate and then cross sectioned using a diamond edge saw (Minitom, Struers) to a width maximum of 2 mm, in order to allow X-ray penetration. The alumina was purely to provide strength during cross sectioning and this process caused only very minor exterior damage at the edges, but allowed internal features to remain intact.

In later experiments the samples approximately 2 mm×5 mm×0.16 mm were made from commercial PZT. These

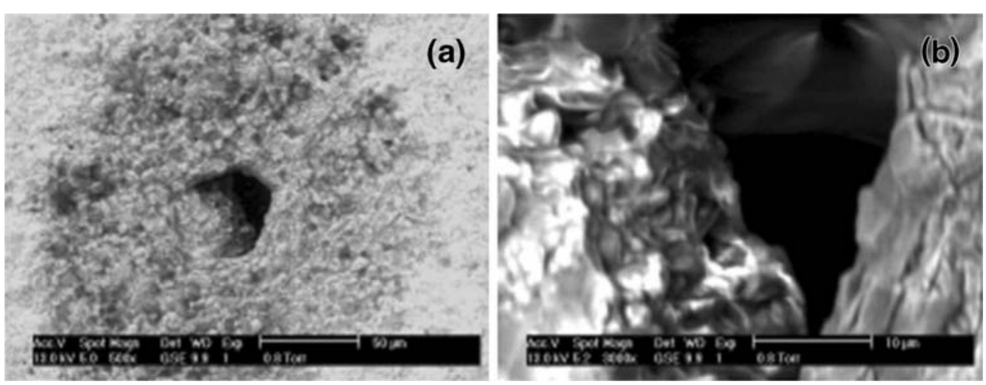

Fig. 2 SEM image of a discoloration region showing there is a corresponding surface defect, (**a**) 500× and (**b**) 3000× magnification

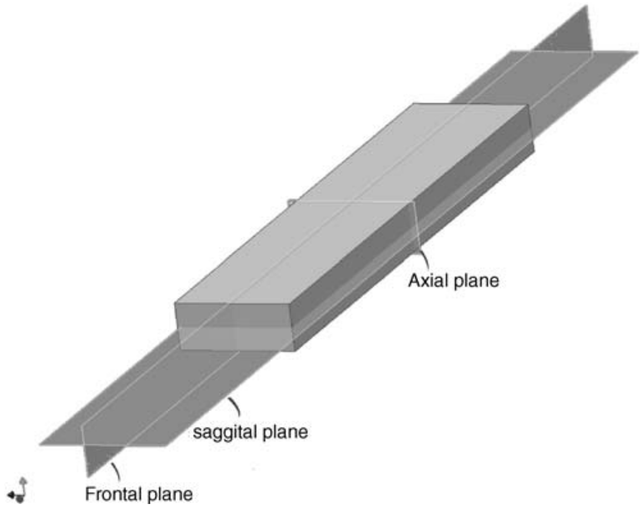

Fig. 3 The 3 planes: frontal, sagittal and axial, of which the μCT machine produces detailed information

samples were μCT scanned before and after temperature–humidity bias testing and the findings obtained compared.

3 Results and discussion

Figure 1 shows BFM images from a sample after test and a fresh control sample. After removal of the nickel electrodes discolored regions can be readily observed on the anodic surface of the ceramic compared to control samples (Fig. 1).

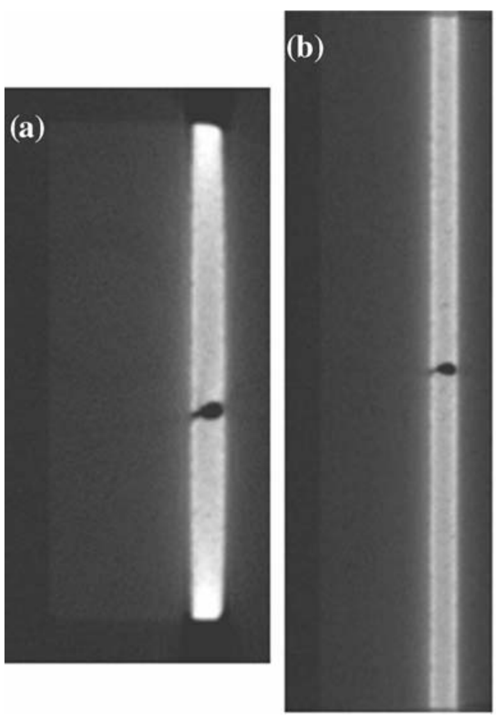

Fig. 4 μCT of the discolored region in Figure 2 showing the formation of a large void as seen from both the axial view (**a**) and the sagittal view (**b**). *Bar*=160 μm

Figure 2 is a photomicrograph of one of the discolored regions under ESEM. It can be seen that a surface hole or defect is related this region. Figure 2(a) shows clearly the region of discoloration surrounding the surface defect. Figure 2(b) shows that at high magnification (×3000) the internal formation of the defect cannot be resolved.

Although ESEM is useful in visualizing surface abnormalities, any internal changes are not visible unless the sample is sectioned. To overcome this drawback this research applied μCT to non-destructively visualize any changes to the internal structure. This technique provides the user with detailed information in all of the structures 3 planes defined here as frontal, sagittal and axial which are shown schematically in Fig. 3.

Figure 4(a) and (b) show the μCT visualization of the surface pore described using ESEM previously (Fig. 2). It can be seen that a surface hole or defect is related to this region. Control samples do not show linked surface and internal void formations. However, it is impossible to confirm whether or not the void has grown during testing without before and after scans.

It is proposed that this structural change is caused by partial discharges [11–13] within the ceramic, with the

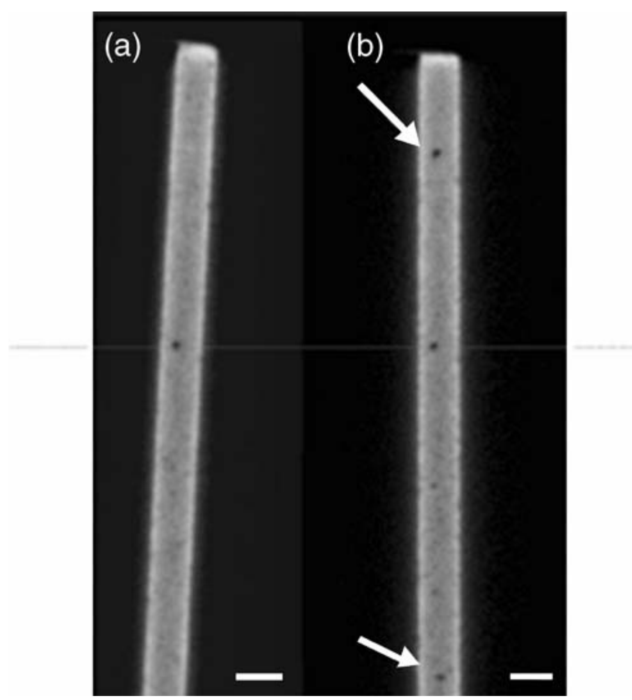

Fig. 5 μCT sagittal cross -sectional image of the same PZT actuator sample showing an internal pore whose dimensions have not significantly changed (*line*) and the appearance of other internal voids (*arrows*) when comparing scans from (**a**) before environmental testing and (**b**) after 48 h at 55 °C, 90 RH and biased at 1.6 kV/mm d.c. *Bar*=160 μm

propensity for discharge being increased by the ingress of water through surface defects [14].

The destructive nature of partial discharges in dielectric voids is well known [11]. It has been shown in insulators such as PZT that if there is an air filled crack or pore within the ceramic then the electrical field across the structural flaw can be enhanced approximately 1000 times higher than the applied electric loading [12]. Under such conditions the electrons in the pores will be accelerated ionizing the atmospheric gases and the produced ions accelerated towards the negatively charged side of the flaw. In this situation the air is said to be partially discharged. This process can continue in an avalanche fashion until the surfaces charge has been countered by a layer of oppositely charged particles. Before this can happen there is the possibility of a streamer-type discharge caused by the difference in velocity of the electrons and the ions [13]. The effects of this type of discharge on the bulk of the material have been shown to be heavily influenced by the water vapor content of the discharged gas [14], with high humidity causing an increase in the formation of microchannels [14]. If this type of discharge was forceful enough then it is proposed that it would expel part of the ceramic away from the main structure leaving the surface flaw and underlying void which is visualized under μCT (Figs. 2 and 4).

To examine this in more detail a subsequent experiment took smaller samples (2 mm×5 mm×0.16 mm). These samples were scanned prior to temperature–humidity bias testing. The samples were rescanned after testing and the images obtained compared for structural alterations.

Figure 5 shows a void that has formed within the sample during the production process, and gives a comparison between the same region on a sample before and after testing. The photomicrographs clearly show that μCT can distinguish an increase in the number of internal voids present within the ceramic during temperature–humidity bias testing. Figure 5 also indicates that the dimension of the large void that was present prior to testing has remained relatively constant.

The change in ceramic resistance of PZT actuators under temperature–humidity bias testing has been described previously [10]. However the assessment of any internal structural change has not been investigated and is extremely difficult if not impossible to perform using conventional light and electron microscopy procedures without destroying the features under investigation. CT offers a solution to this problem and can provide a non-destructive method of visualizing changes within the body of a device.

Although the resolution of this technique (μCT) is limited by the power of the accelerating voltage to a few microns, it is planned that further work would look to apply synchrotron radiation [15, 16] to tested samples in order to obtain sub-micron resolution levels and allow a greater insight into internal structural changes caused by environmental factors.

4 Summary

The conclusions of this research are:

- μCT allows the non-destructive visualization of internal defect formation and growth in small PZT ceramic parts.
- Unlike conventional microscopy techniques μCT is able to provide information on the evolution of internal structural changes over time.
- For the soft PZT ceramics tested structural changes such as the increase in the number and size of pores and appearance of microcracks occur within the ceramic during temperature humidity bias testing.

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