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Microcrack clouds in fatigued electrostrictive 9.5/65/35 PLZT

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Abstract

The formation of microcracks during cyclic bipolar fatigue is shown for purely dielectric electrostrictive 9.5/65/35 transparent lead–lanthanum–zirconate–titanate (PLZT). The optically transparent material allows for the direct observation of broken grain facets. The maximum reached polarisation decreases by 30% after 3×10^6 cycles. The formation of microcrack clouds at the specimen edges and at macroscopic cracks are shown. No cracks were found in sample regions without obvious flaws present. By estimating the cracked volume, comparisons with FEM from previous studys allow to calculate the influence of the observed cracking on the polarization. As a consequence, the observed microcracks are considered to be mainly responsible for polarisation degradation.

Keywords: Fatigue; Ferroelectric properties; Fracture; PLZT

1. Introduction

The fatigue behaviour of electroactive devices has been subject of many investigations (references can be found in Ref. 1). Many underlying mechanisms have been proposed, but the understanding of the particular contribution to the overall damage and their timely sequence remain open questions. Several authors state that microcracking is involved in damage.[2–4] It is particularly fatal as it lays the ground for macrocrack formation. It was shown that thermal depolarisation does not lead to complete recovery of switchable polarisation, supporting the assumption that some residual damage is present as microcracks.[3,4] Only very few publications deal with the fatigue behaviour of electrostrictive materials (e.g. Ref. 5).

Experimental evidence for the existence of microcracks in fatigued specimens was given by direct observation in the TEM[2] and SEM[6–8] and indirect measurement of elastic properties.[9] It is generally accepted that microcracks reduce the local electric field in their vicinity.[10,11] Computational simulations showed that the macroscopic polarisation is decreasing with increasing size of cracked area.[12] A direct measurement of the effect of a given microcrack density on the macroscopic response is difficult in ferroelectrics, since the superimposed degradation due to domain pinning is suggested to be also present in the fatigued material. Electrodes are known to serve as a starter location for the microcracking,[4] but the growth process is still unclear.

The material used in this investigation offers two major advantages: It is transparent, allowing for optical investigations also in the bulk and shows no hysteretic behaviour due to domain movement.

2. Experimental

Commercial hot pressed, 100% dense PLZT with the well characterised composition 9.5/65/35 was used.[13–15] At room temperature, this material is purely electrostrictive and transparent. The material was cut to pellets of 0.95 mm thickness, 10 mm in diameter. The flat surfaces were polished to a 1 μm finish. The specimens were investigated by optical microscopy prior to electroding. The flat surfaces were sputtered with Au/Pd of approx. 100 nm thickness leaving a rim of 250 μm uncovered to avoid arcing during high voltage application. For fatigue experiments, a 50 Hz sinusoidal electric field of 2 kV/mm maximum amplitude was applied for 3×10^6 switching cycles. The polarisation vs. electric field loop was monitored using a capacitor in series and visualised

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on an oscilloscope screen. To avoid arcing, the specimens were immersed in silicone oil. For further details on the cycling procedure, see Ref. 1.

3. Results

Fig. 1(a) shows the unfatigued hysteresis loop during the first switching cycles. It exhibits the almost linear electrostrictive behaviour with a minuscule hysteresis (at 50 Hz). The peak polarisation value is ~0.24 C/m^2. After 3×10^6 cycles, The polarisation peak value has degraded by 30% [Fig. 1(b)].

No obvious damage in any region within the specimen was optically seen before cycling. An image taken after cycling and electrode removal is shown in Fig. 2. Macroscopic cracks were found at the electrode–ceramic boundary pointing to the centre of the specimen (arrow in Fig. 2). All these cracks appeared within a ring of ~300 μm distance from the electrode edge (dashed line in Fig. 2). The cracks reached lengths of 50–80 μm with a spacing of ~100 μm between the individual cracks. Deviation from this regular crack pattern was only observed in the case of shape inhomogeneities of the electrode. Fig. 3 shows a higher magnification of one such microcrack cloud. In a region of ~10–20 μm around the macroscopic cracks, intensive intergranular microcracking was observed.

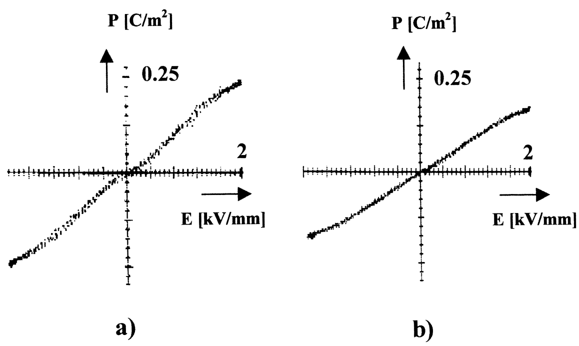

Fig. 1. (a) Unfatigued and (b) fatigued (3×10^6 cycles, 50 Hz, 2 kV/mm) polarisation loops in 9.5/65/35 PLZT.

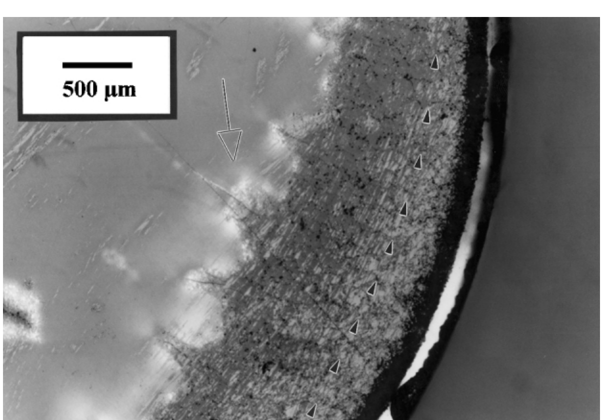

Fig. 2. Microcrack clouds (arrow) grown from the electrode edge into the material after 3×10^6 bipolar cycles at 2 kV/mm. The dashed line indicates the edge of the electrode which was removed for optical investigation. The cracked area is opaque and therefore appears darker than the uncracked regions.

Microcracks were exclusively observed near macrocracks or the specimen/electrode edge. Fig. 4 sketches the regions of micro- and macrocracking. Neither macroscopic nor microscopic cracking was found at the centre bulk or under intact electrodes in the specimen centre.

4. Discussion

It is known that any microstructural defect like surface unevenness,[4] pores[16] or already existing cracks may serve as a starter for fatigue cracks. Internal flaws sufficiently enhance the externally applied stress fields to locally initiate cracking. In hot pressed electrostrictive PLZT no internal flaws are present. Despite high mechanical stresses under application of high electric fields due to grain misorientation no optically detectable cracks are observed in the interior of the PLZT before or after cycling. Even though 9.5/65/35 is elastically isotropic for low fields, anisotropy develops at elevated fields, because the

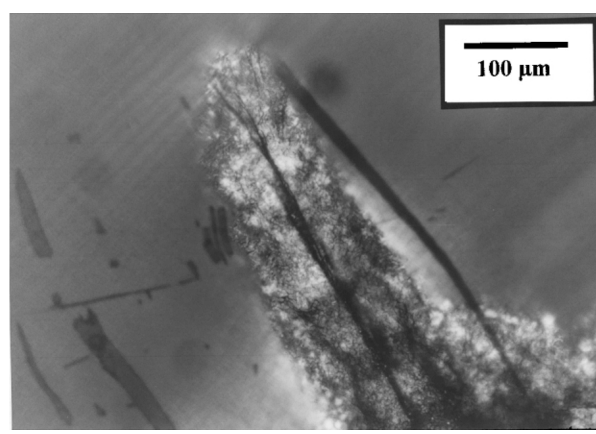

Fig. 3. Microcrack clouds surrounding an already existing macrocrack. This detail shows broken grain facets. No indication for intragranular fracture was observed.

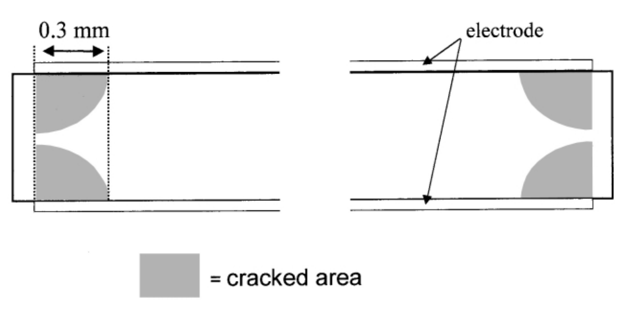

Fig. 4. Schematic drawing of the sample cross-section view, showing the appearance of the cracked area (darkened areas). A width of 0.3 mm for the cracked area was determined.

induced polarisation and strain cannot arbitrarily orient in the rhombohedral grains. The rim of non electrode-covered material left for insulation purposes results in a region of material that is not subjected to electric fields. This imposes macroscopic constraints inducing macroscopic stresses at the electrode edge, which is known to be the initiation site of primary cracks.[17] Subsequently, a flaw will permit material in its vicinity to grow or shrink more, because it experiences less constraints by its environment and local field enhancement. The additional strain directly translates into additional elastic stresses at some other points in the vicinity, because no switchable domain system is present which could accommodate such local stresses and constitute some toughening. Thus, once a flaw enhances local stresses, the process perpetuates and microcracks form dense clouds around the initial defect.[18]

The microcrack density can be used to estimate the effect on the macroscopic response of the specimen. Fig. 4 shows a cross-section view of the fatigued specimen indicating the microcracked region. An average width of 0.3 mm for the cracked zone was determined from the micrographs, corresponding to a cracked volume fraction of 8% of the entire electrically active sample. Kim and Jiang used a computational simulation for estimating the effect of microcracking on the maximum achievable polarisation in a ferroelectric material.[19] They used a linear piezoelectric material law neglecting ferroelectric domain switching, so the results can be transposed on our electrostrictive material to some extent. They showed that the ratio of cracked and uncracked volume and the reduction of macroscopic polarisation are linearly related and that 2.5% of cracked volume is responsible for 10% of polarisation loss. Following that argumentation, 30% loss, as observed in our study, can be achieved by 7.5% cracked volume, in excellent agreement with our observation. However, the agreement may be partly fortuitous. Kim and Jiang[19] modelled a completely random microcrack distribution throughout the sample which may provide a different microcrack–microcrack interaction than our case. Therefore, other phenomena may still have to be considered. Nevertheless, the absence of domains and hysteretic behaviour points to a lesser influence of electrochemical processes, in contrast to fatigue in ferroelectrics.

The intention of the present study is twofold: First, the presented direct observation of formation of microcrack clouds in the vicinity of obvious flaws like electrode edges or precracks might give new insights in the basic questions like microcrack initiation and microcrack propagation not only in electroactive materials. Secondly, FEM studies predicting the influence of microcracks on the polarisation in electroactive material find at least partial experimental confirmation.

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