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Direct observation of domain switching and crack nucleation in a piezoelectric material

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Abstract

The effect of an electric field on domain switching and fatigue induced crack nucleation and growth in a piezoelectric material of nominal composition $Pb(Zr_{0.5}, Ti_{0.5})O_3$ has been investigated. The ceramic was subjected to localised static and cyclic electric fields, which were applied via pairs of closely spaced surface-mounted electrodes, while simultaneously imaging the microstructure in the SEM. Electric field–polarisation hysteresis loops were also collected from the local region using the same electrodes.

Domain wall mobility was observed above a threshold electric field strength, as was microcracking. Cracks were seen to nucleate at grain boundaries, and were sometimes associated with microstructural features, such as pores. Crack propagation was mainly intergranular, and occurred preferentially in a direction parallel to the local field direction. Transgranular fracture was also observed, with the crack path being influenced by interaction with domain boundaries. Factors affecting domain switching and crack propagation are discussed in the context of the locally applied electric field.

Keywords: C. Mechanical properties; C. Piezoelectric properties; D. PZT; Domains

1. Introduction

Bulk lead zirconate titanate (PZT) piezoceramics are widely exploited in various applications including: sensors, actuators, transducers, transformers and filters [1,2]. In operation, many of these devices are subjected to large electric fields that give rise to significant local mechanical stresses, resulting in fatigue and degradation of electromechanical properties [3–6]. In order to understand the underlying mechanisms controlling these processes, several workers have studied the fine-scale mechanisms of failure in piezoelectric materials under electrical and mechanical loading. Many researches have suggested that electrical and mechanical fatigue of piezoelectric ceramics is predominantly due to the effects of domain switching during electric field cycling [7–13]. Electrical fatigue, associated with a reduction in switched charge and induced strain [14], has been attributed to a range of microstructural and microchemical features including: grain size, pore distribution, and chemical segregation effects [15]. Under cyclic voltage loading, mechanical degradation by crack nucleation and growth mechanisms has also been reported to occur [4,15,16]. At low electric fields (E), below the coercive field (E_C), crack propagation is attributed to the effect of domain switching due to stresses arising from the localisation of the electric field around crack tips [15,17]. Above E_C, crack propagation is believed to be due to localised stresses arising from differences in electric field strength between the cracked and uncracked regions [15]. A study of through-thickness cracks in bulk PZT established a threshold value of E, corresponding to $0.797E_C$, below which crack growth did not occur, but above which the propagation rate increased steadily, allowing a phenomenological model for the relationship between field strength and rate of crack growth to be developed [18].

Crack-growth experiments have been carried out on barium titanate (BT) [8,19], lead magnesium niobate–lead niobate (PMN–PT) solid solutions [20,21] and PZT [18,22] using pre-cracked bulk specimens and cyclic electric fields. A general preference for crack propagation perpendicular to the electric field direction was observed in unconstrained samples. A TEM-based study of single crystal PZT found that microcrack

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nucleation occurred under both static and cyclic electric fields and noted an interaction between the preferred crack path and domain wall alignment [23].

In this study, SEM based local property measurement techniques have been used to measure electric field–polarisation hysteresis loops of localised regions within a PZT ceramic, on the 20 μm scale, using surface mounted electrodes, while making simultaneous in situ observation and characterisation of microstructural changes at the sample surface, including domain wall movements due to switching, and crack nucleation and growth under conditions of constant electric field and cyclic fatigue loading.

2. Experimental

$(Pb_{0.965}La_{0.01}Sr_{0.02})(Zr_{0.5}Ti_{0.5})O_3$ powder was prepared by a standard mixed oxide route using GPR grade starting powders. The sample composition was chosen to lie within the tetragonal phase region of the PZT phase diagram, close to the morphotropic phase boundary (MPB). Green compacts were uniaxially pressed at 400 MPa, and sintered in oxygen at 1225 °C for 2 h within a closed alumina crucible, which was covered with $PbZrO_3$ powder to limit lead loss. A cross-section of the sintered specimen was cut, ground flat and polished with diamond paste, followed by lapping with a colloidal silica suspension to give a strain-free surface suitable for imaging and electron backscattered pattern (EBSP) analysis. An array of 80 μm square Pt electrodes, with 20 μm separation, was sputtered onto the polished surface through a photolithographically applied lift-off mask. The sample was mounted in a Phillips XL30 FEGSEM, and observed using backscattered electron (BS) imaging under low energy beam conditions (8 kV, 1.5 nA) to prevent beam-induced specimen damage. Grain size was calculated using SEM images and a standard linear intercept method, applying a geometrical factor of 1.56 [24]. The orientation of twin planes and selected grain boundaries was determined by EBSP analysis using a HKL EBSP system mounted on the same SEM. Because of the low *c/a* ratio and the resolution limitations of EBSP indexing, crystal directions were calculated according to a cubic unit cell. Electrical contact to the surface electrodes was made using micromanipulator controlled tungsten probes and a custom-built voltage source was used to generate an electric field within the sample between pairs of adjacent electrodes. DC fields were allowed to stabilise for 60 s prior to imaging, and images were collected with the electric field 'on'. AC voltage cycling was carried out at 20 Hz. Each series of cycles was terminated with an identical poling pulse to ensure that images were collected under identical conditions to allow consistency of observation. Spatially localised polarisation–electric field hysteresis loops were collected via the same electrodes using proprietary software.

3. Results and discussion

3.1. Microstructure

The density of the ceramic was measured using the Archimedes method and found to be 98% of theoretical. There is some fine residual porosity, principally at triple points but also occasionally extending along grain boundaries. The grains are equiaxed, with a mean size of 6 ± 2 μm. Twinning is readily observed within the grains, although subsequent images contain significant noise due to the low-energy imaging conditions that were adopted in order to prevent beam-induced sample damage.

3.2. Static field behaviour

Fig. 1(a) shows a BS image of a PZT grain, located between a pair of electrodes situated 20 μm apart and located to the left and the right of the viewed area. The edge of the right-hand electrode is visible at the side of the image. Microstructural changes occurred throughout the material due to the application of an external electric field: we will use the example of this grain to illustrate in detail the typical behaviour. The twin plane orientations within this grain were indexed using a combination of EBSP and trace analysis, and are indicated in the figure, being a mixture of {1 1 0} and {1 0 0} types. A localised electric field was generated in the sample by applying a voltage across the electrodes. The magnitude and polarity of the applied electric field is defined here by the potential of the right-hand electrode (the edge of which is visible in the image) relative to the left-hand electrode, with the resultant electric field being oriented horizontally across the image. For voltages in the range 0 V to −130 V, the microstructure appeared to remain unchanged. However, when the voltage was increased to −135 V there was significant domain wall movement, bringing the twin configuration shown in Fig. 1(b) into contrast. The twin planes within this grain are now all of the {1 1 0} type and are aligned approximately perpendicular to the electric field direction. The associated strain has also resulted in the nucleation and propagation of an intergranular crack in a direction parallel to the electric field. This new twin arrangement was retained after the electric field was removed, although the crack was observed to close slightly, indicating some strain relaxation.

The electric field direction was then reversed by applying +135 V to the right-hand electrode. This led to further domain wall movement, and the formation of the twin microstructure shown in Fig. 1(c). The twin planes in contrast are now a mixture of {1 1 0} and {1 0 0} types, and generally lie at about 45° to the electric field direction. The intergranular crack has propagated further and forked along two grain boundaries, both of which lie at about 45° to the electric field direction. On removing the electric field, the twin microstructure immediately relaxed to that shown in Fig. 1(d). The fine twins that are now in contrast lie predominantly on {1 1 0} planes. There is also a prominent {1 1 1} twin running through the centre of the grain. It was possible to switch repeatedly between the twinning patterns shown in Fig. 1(b)–(d) by applying −135 V (Fig. 1(b)), removing the field (Fig. 1(b)), or by applying +135 V (Fig. 1(c)) and then removing the field (Fig. 1(d)). We note the twin arrangement formed when −135 V was applied, with {1 1 0} twin planes aligned perpendicular to the electric field, is stable to the removal of the electric field while the configuration formed in the same grains by reversing the electric field, where the twin planes predominantly lie at 45° to the field direction,