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Elastic modulus, hardness and fracture behavior of $Pb(Zn_{1/3}Nb_{2/3})O_3$–$PbTiO_3$ single crystal

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Abstract

The deformation, crack initiation, fracture behavior and mechanical properties of (0 0 1)-oriented single crystal of $Pb(Zn_{1/3}Nb_{2/3})O_3$–7% $PbTiO_3$ (PZN–7% PT) in both unpoled and poled states have been investigated by using nanoindentation, micro-indentation and three-point bending experiments. Nanoindentation experiments revealed that, unlike typical brittle materials, material pile-ups around the indentation impressions were commonly observed at ultra-low loads. The elastic modulus and hardness were also determined by using nanoindentation experiments. The critical indentation load for crack initiation, determined by using micro-indentation experiments, is 0.135 N for unpoled samples, increasing to 0.465 N for the positive surface (crack propagation direction against the poling direction) of poled samples but decreasing slightly to 0.132 N for the negative surface (crack propagation direction along the poling direction) of the poled samples. Indentation/strength (three-point bend) test showed a similar trend for the "apparent" fracture toughness, giving 0.36 $MPa\sqrt{m}$ for unpoled samples, increasing to 0.44 $MPa\sqrt{m}$ for the positive surface of poled samples but decreasing to 0.30 $MPa\sqrt{m}$ for the negative surface of poled samples. Polarized light microscopy and scanning electron microscopy were used to study the material adjacent to the indentations and the fracture surfaces produced by the three-point bend tests. The results were correlated with the various fracture properties observed.

Keywords: Indentation; Piezo-materials; Single crystal; Fracture; Deformation; Cracks

1. Introduction

Relaxor-based ferroelectric single crystals, such as lead zinc niobate–lead titanate, $Pb(Zn_{1/3}Nb_{2/3})O_3$–$PbTiO_3$ (PZN–PT), and lead magnesium niobate–lead titanate, $Pb(Mg_{1/3}Nb_{2/3})O_3$–$PbTiO_3$ (PMN–PT), solid solution single crystals, have recently attracted considerable attention due to their extremely high dielectric and piezoelectric properties [1–8]. These single crystals have been touted as the preferred materials for future high-performance devices such as underwater projectors, hydrophones, medical ultrasound imaging transducers, sensors, actuators, etc. In these applications, the material experiences combined electrical and mechanical loadings. Moreover, actuation forces may act as crack driving sources, eventually leading to failure of the device. Thus, the investigation of deformation, cracking and fracture behavior of relaxor single crystals under the influence of mechanical and electrical loadings is of practical importance.

Although the deformation, cracking and fracture behavior of piezo-ceramics, such as PZT ceramics, have been studied extensively and well documented in literature [9–22], only very few works related to these issues have been done on single crystal piezo-materials [23–25]. As new devices involving these single crystals are being explored, the mechanical issues will become more and more important in the design and fabrication. This paper presents recent studies on deformation, crack initiation and fracture behavior of PZN–7% PT single crystal, both in unpoled and poled states, by means of the nano- and micro-indentation techniques and three-point bend tests. The elastic modulus, hardness, critical indentation load for crack nucleation and the "apparent" fracture toughness of the PZN–PT are determined.

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2. Materials and experimental work

2.1. Materials

PZN–7% PT single crystal grown using the high-temperature flux technique [26,27] (Microfine Materials Technologies Pte Ltd., Singapore), were used in the present work. Plate samples of 7.0 mm (L) × 2.5 mm (W) × 0.4 mm (T) in dimensions were diced from (0 0 1) wafers with [1 0 0] as length and [0 1 0] as width. Both unpoled and poled samples were investigated. The poled samples were prepared by first coating the opposite (0 0 1) faces of the samples with nichrome and gold–palladium in a high vacuum coater, followed by poling in silicone oil at room temperature under an electric field of 0.9 kV/mm. In this work, the specimen surface connected to the positive polarity of the power source is referred to as the negative surface while that connected to the negative polarity as the positive surface, and the poling direction as the thickness direction pointing from the negative surface to the positive surface.

2.2. Nano- and micro-indentation experiments

Nanoindentation experiments were performed on the (0 0 1) surface of the PZN–PT samples (Nano Indenter XP^{TM}, MTS Corporation, USA) with a Berkovich diamond indenter tip, to investigate the deformation behavior of the PZN–PT crystal at ultra-low loads and to determine the elastic modulus and hardness of the crystal. The samples were oriented such that one of the edges of the indent was along the [1 0 0] crystal direction, i.e., parallel to the length of the sample, as indicated by indent (a) in Fig. 1. The load was applied at a constant strain rate of 0.05 s^{-1} until the maximum load was reached. The maximum load was held for 10 s followed by unloading at the same rate (of 0.05 s^{-1}). Maximum loads of 50, 30, 20, 10, 5 and 1 mN were used. Ten indents were made per maximum load condition on each sample. The deformation behavior around the indentation sites was studied using the scanning electron microscope (SEM) (JSM6700F, JEOL, Japan). The elastic modulus and hardness of the PZN–PT samples were determined using conventional nanoindentation analysis [28]. It is well-known that both the elastic modulus and hardness of single crystals showed anisotropic behaviors

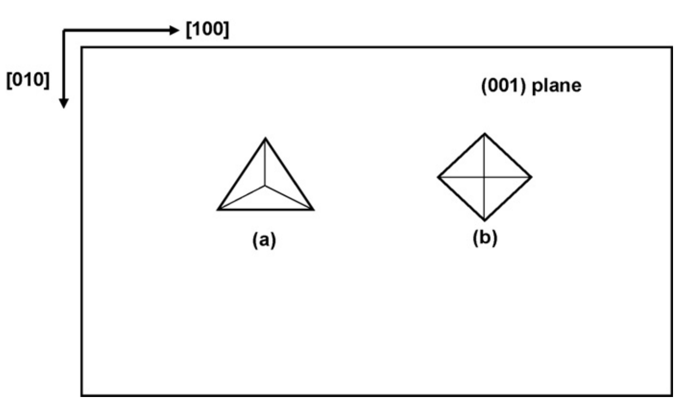

Fig. 1. Schematic diagram showing the orientations of the (a) Berkovich and (b) Vickers indentations on (0 0 1) plane of the single crystal.

depending on the crystal orientations. However, for simplicity's sake, the anisotropic nature of elastic modulus and hardness of PZN–PT will be ignored in our analysis.

For the micro-indentation experiment, PZN–PT samples were loaded mechanically using a Micro-Force Tester [Model 8848, Instron Ltd., USA] by pressing a Vickers diamond indenter tip against the (0 0 1) surface. The samples were oriented such that the diagonals of the Vickers indents were along [1 0 0] and [0 1 0] crystal directions of the sample, as indicated by indent (b) in Fig. 1. The loading, holding and unloading duration were 20 s each and the load was applied by a displacement-controller at a rate of 0.001 mm/s. Immediately after loading the indentation impressions were examined with an optical microscope and micrographs of the indents were taken within 2 min to minimize the effect of the environment on crack growth. The critical indentation load for crack initiation, the crack length and orientation were determined from the micrographs.

Both unpoled and poled samples were tested by nano- and micro-indentation experiments. The nanoindentation (10 indents) and micro-indentation experiments (20 indents) were conducted on both the positive and negative surfaces of the poled samples. The indentation on the positive surface was against the poling direction, whereas the indentation on the negative surface was along the poling direction. Cross-polarized light microscopy was used to investigate the effect of mechanical and electrical loadings on domain structures and their influence on the cracking behavior in both poled and unpoled samples.

2.3. Three-point bend tests

Due to size limitation of the specimens (7 mm in length), three-point bend tests were carried out on both unpoled and poled samples. One surface of the test samples was first pre-cracked with a Vickers indenter; the indentation load used for pre-cracking was higher than the critical indentation load (to be discussed in Section 3.2) in order to produce a crack (or cracks) at the center of the surface on the specimen. During indentation, the sample was oriented so that the diagonals of the Vickers indentation were aligned along the length and width of the test sample. Then, the pre-cracked sample was loaded under three-point bending condition until it fractured.

Due to the small size of the samples used, all three-point bend tests were performed using a non-standard micro-bending fixture having a support span of 5 mm. All the tests were carried out on a universal testing machine [Model 5543, Instron Limited, USA] with the pre-cracked surface of the sample under the tensile bending stress during the test. A displacement rate of 0.5 mm/min was applied until the sample fractured. After the test, the fracture surfaces were examined using the SEM. Ten specimens of each type of samples (unpoled, positive surface and negative surface of poled samples, respectively) were tested.

The indentation load, flexural strength, elastic modulus and hardness (determined by nanoindentation experiments) were used to calculate the "apparent" fracture toughness of the specimens [29,30]. The term "apparent" fracture toughness is used here to denote that the test technique used is not a standard one, although similar techniques have been used frequently in frac-