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Crack tip process zone domain switching in a soft lead zirconate titanate ceramic

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Abstract

Non-180° domain switching leads to fracture toughness enhancement in ferroelastic materials. Using a high-energy synchrotron X-ray source and a two-dimensional detector in transmission geometry, non-180° domain switching and crystallographic lattice strains were measured in situ around a crack tip in a soft tetragonal lead zirconate titanate ceramic. At $K_I = 0.71$ MPa m$^{1/2}$ and below the initiation toughness, the process zone size, spatial distribution of preferred domain orientations, and lattice strains near the crack tip are a strong function of direction within the plane of the compact tension specimen. Deviatoric stresses and strains calculated using a finite element model and projected to the same directions measured in diffraction correlate with the measured spatial distributions and directional dependencies. Some preferred orientations remain in the crack wake after the crack has propagated; within the crack wake, the tetragonal 001 axis has a preferred orientation both perpendicular to the crack face and toward the crack front.

Keywords: Ferroelectricity; Fracture; Ceramics; Toughness; X-ray diffraction (XRD)

1. Introduction

The inherent brittleness of ferroelectric ceramics is a structural liability that leads to crack initiation at defects and stress concentrations such as pores and electrode and substrate interfaces. However, non-180° domain switching in the frontal zone and crack wake lead to a rising R-curve behavior, or an increase in toughness with crack extension [1–10]. Using various techniques, recent work has elicited the region in which domain switching occurs or the "switching zone" in ferroelastic materials, the size of which is related to the toughness enhancement. In $BaTiO_3$ ceramics, Nomarski interference contrast and atomic force microscopy have been employed to measure local surface depressions that result from the strain associated with ferroelastic switching perpendicular to the sample surface [8,9]. In electrically poled lead zirconate titanate (PZT) ceramics, Lupascu and co-workers showed that the change in potential energy can be mapped spatially surrounding the crack tip using a liquid-crystal display [2,6]. Employing X-ray diffraction, Glazounov et al. [5] measured the intensity ratio change of certain diffraction peaks as a function of distance from the crack face. Hackemann and Pfeiffer [7] have also demonstrated that domain orientations perpendicular to the sample surface can be measured around the crack tip using a small beam size from laboratory X-rays in reflection geometry.

However, non-180° domain switching within the plane of the sample has yet to be reported, and it is this plane that exhibits a complex stress distribution contributing to the toughness enhancement. The directionally dependent

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domain switching distributions around the crack tip are discussed here using the well-known Mode I elastic stress profiles. In plane-stress Mode I loading, elastic stress distributions as a function of radial coordinates (r, θ) are given as [11]

$$\sigma_{XX} = \frac{K_I}{\sqrt{2\pi r}}\cos\frac{\theta}{2}\left[1-\sin\frac{\theta}{2}\sin\frac{3\theta}{2}\right] \qquad (1)$$
$$\sigma_{YY} = \frac{K_I}{\sqrt{2\pi r}}\cos\frac{\theta}{2}\left[1+\sin\frac{\theta}{2}\sin\frac{3\theta}{2}\right]$$
$$\tau_{XY} = \frac{K_I}{\sqrt{2\pi r}}\cos\frac{\theta}{2}\sin\frac{\theta}{2}\cos\frac{3\theta}{2}$$
$$\sigma_{ZZ} = \tau_{YZ} = \tau_{XZ} = 0$$

Fig. 1 illustrates the crack orientation in both Cartesian (X, Y) and radial coordinate systems.

The present work is motivated by a desire to better understand the directionality of domain switching near a mechanically loaded crack tip. To this end, the behavior of local crystallographic orientations are examined at an array of points spanning the switching zone. A useful quantity for illustrating the in-plane directional variations as a function of position relative to the crack tip (as well as providing clear comparisons between experimentally measured and modeled data) is the stress projection

$$\sigma_{\vec{n}}(\eta) = n_i(\eta)n_j(\eta)\sigma_{ij}, \qquad (2)$$

where $\vec{n}(\eta)$ is a unit vector with an in-plane direction with respect to the sample coordinate system shown in Fig. 1. The scalar $\sigma_{\vec{n}}(\eta)$ represents a normal stress, i.e. the component of the traction vector acting on a surface with normal $\vec{n}$ in the direction $\vec{n}$. As a consequence of the measuring convention used (see Fig. 1 inset), $\sigma_{\vec{n}}(\eta)$ is equivalent to the σ_{YY} component transformed to a new coordinate system by a clockwise rotation of η degrees about Z [12]. Note that Eq. (2) also implies an in-plane antipodal symmetry described by $\sigma_{\vec{n}}(\eta) = \sigma_{\vec{n}}(\eta + 180°)$.

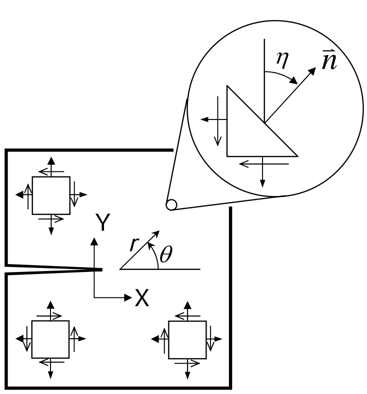

Fig. 1. Schematic of crack geometry. Parameters r and θ define a physical position (X, Y) with respect to the crack. Rotation angle η from the Y-axis corresponds to the projection direction $\vec{n}$ at each individual X, Y position. Differences in the directions of normal and shear strains in different regions relative to the crack tip are illustrated.

It is understood that ferroelastic domain switching is caused by the deviatoric stresses [13–15]. The projected deviatoric stress, $s_{\vec{n}}(\eta)$, is given by

$$s_{\vec{n}}(\eta) = \sigma_{\vec{n}}(\eta) - \delta_{ij}\sigma_{ij}/3 \qquad (3)$$

where δ_{ij} is the Kronecker delta. Using Eqs. (1)–(3), $s_{\vec{n}}(\eta)$ is calculated as a function of position (X, Y) surrounding a crack tip for an applied stress intensity factor of $K_I = 0.71\,\mathrm{MPa\,m^{1/2}}$. Spatial distributions of $s_{\vec{n}}(\eta)$ for $\eta = 0°$, 30°, 60°, and 90° are shown in Fig. 2. Given that these deviatoric stresses induce domain switching and that their spatial distributions change with angle in the plane of the sample, it is hypothesized that the domain switching behavior in these directions of the sample will be highly correlated with these distributions.

Because the techniques used in prior crack tip switching zone measurements have no in-plane directional resolution, we present a new approach by which the directional dependence described above is resolved. High-energy X-rays can penetrate through several millimeters of most materials and therefore provide a powerful X-ray transmission technique by which to characterize in-plane behavior. When position sensitive area detectors are employed, high-energy synchrotron X-rays enable the capture of the entire ring of scattering vectors associated with each Debye–Scherrer cone for a single sample position [16–18].

Because the Bragg angles for most lower-order reflections in these materials are typically 5° or less for high-energy X-rays ($\lambda < 0.25$ Å), the cones of scattering vectors lie nearly parallel to the X–Y plane of the sample. Therefore, all scattering vectors for each reflecting plane $\{hkl\}$ are treated as lying in the X–Y plane of the sample in this analysis. In this geometry, the normal lattice strains can then be extracted from the experimental data using

$$\varepsilon_{hkl}(\vec{n}) = \frac{d_{hkl} - d^{\circ}_{hkl}}{d^{\circ}_{hkl}}, \qquad (4)$$

where d_{hkl} and d°_{hkl} are the measured mean crystallographic lattice spacing for strained and unstrained crystallographic orientations such that $\vec{n}$ is parallel to the $\{hkl\}$ pole. These measured strains may, in turn, be related analytically to an average projection of the underlying strain tensors in the same crystallographic domain in a manner analogous to Eq. (2).

In this work, we combine high-energy synchrotron X-ray diffraction with a two-dimensional detector to map both the preferred orientation induced by ferroelastic domain switching and $\{hkl\}$ lattice strains in the plane of a compact tension specimen at stress intensity factors approaching and exceeding the initiation toughness. The directionally dependent in-plane domain switching behavior in a soft PZT ceramic is thereby resolved and discussed in the context of the complex stress state at the crack tip.

2. Experimental procedure

A soft Nb-doped $Pb(Zr_{0.52}Ti_{0.48})O_3$ (PZT) ceramic with a composition near the morphotropic phase boundary