Evaluation of Toughness Anisotropy on Doped PZT Ceramics as a Function of Load and Temperature

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Abstract. The crack growth resistance behaviour of a doped lead zirconate titanate (PZT) ceramic as a function of loading conditions has been investigated over a temperature range between room temperature (RT) and 400 °C, i.e. above the Curie temperature. The stress-induced switching process has been assessed under compression tests on both pre- and post-indented poled specimens. Non-poled samples have been used as reference material. The change in crack length depending on the poling state before and after the tests has been measured to account for the ferroelectric/ferroelastic behaviour of the PZT material. A corresponding fracture mechanics analysis has revealed toughness anisotropy in the specimens with respect to the poling direction. Experimental findings show the effect of temperature in the domain switching process which can be enhanced by additional mechanical loading, leading to partially depolarisation of the PZT material. The effect of poling orientation coupled with temperature and mechanical stress is discussed. A combination of temperature and mechanical stress has been found to induce a full depolarisation of the PZT material below the Curie temperature.

Introduction

The outstanding piezo-electrical properties of lead zirconate titanate (PZT) based ceramics are widely employed in the fabrication of actuators used in applications that require precision displacement control or high generative forces, i.e. precision mechano-electronic and semiconductor devices. In particular, multilayered piezoelectric actuators, referred to as MPAs, are currently used to control modern fuel injection systems [1, 2]. The rapid operation of MPAs with response times of the order of milliseconds results in low power consumption, high precision control and less noise compared to the common electromagnetic actuators. The functionality of these components is associated with the loading conditions as well as with the actuator design and thermo-electro-mechanical reliability in service.

MPAs are designed as a stack of thin piezo-ceramic layers which are separated by metallic electrodes. In such way high electric fields causing large elongations can be reached with relatively low voltages [3]. The corresponding strains are of the order of 0.1 % (for a typical stack length of about 40 mm the elongation is 40 μm). This elongation can be attained in a very short time, allowing the accurate flow of fuel into the combustion chamber. The effectiveness of such a process (low consumption, reduction of emissions) relies on the reliable functionality of the MPA. In addition, although these MPAs operate under externally applied compressive stresses, failure of components in service has been reported associated with the propagation of cracks within the

electrode-ceramic multilayered structure [4]. In this regard, the relatively large displacements and large forces within the MPA along with the combined thermal, electrical and mechanical loadings yield nonlinear effects which may lead to degradation of the performance of the MPA [5, 6]. Therefore, the investigation of the initiation and subsequent growth of cracks in piezo-ceramic materials is of primary importance and has been the focus of many researchers. Considerable work has been done to analyse the behaviour of monolithic piezo-ceramics to determine the crack growth resistance with respect to defined electrical boundary conditions [5, 7-11]. The combination of electrical and mechanical loads, acting in critical regions of the MPA, yields a different crack growth resistance when applied in service compared to that exhibited by bulk ceramics.

The motivation for this work is to assess the fracture resistance of a doped PZT piezo-ceramic material as a function of applied thermo-mechanical loads over a temperature range between room and Curie temperature. This aims to simulate, to some extent, the real thermo-mechanical behaviour of the PZT ceramic material in MPAs during service. In doing so, the indentation microfracture (IM) method is employed to analyse the crack growth resistance of poled PZT ceramic material as a function of loading conditions. The stress-induced switching process has been assessed on poled specimens at different temperatures. Non-poled samples have been used as reference material to account for the effect of temperature in the depolarisation process. Additionally, mechanical loads have been applied in compression at different temperatures to attain the influence of the combined thermo-mechanical loading on the depolarisation of PZTs. The change in crack length, depending on the remnant poled state of the specimens after thermo-compressive loading conditions, has been measured to account for the toughness anisotropy of the piezo-ceramic material.

Experimental

Material of study

A commercial soft doped PZT ceramic with a composition near the morphotropic phase boundary has been used in these experiments. The material was designed as a stack of thin piezo-ceramic layers sintered at 1100°C in lead-enriched atmosphere. The sintered plates were ground to the finished shape and prismatic bar specimens of dimensions $4 \times 3 \times 10$ mm³ were cut from the plate. CrAg-electrodes were then deposited by a sputtering process onto the small surfaces for poling purposes. Two sides of the specimens were polished with diamond paste up to 1 μm for a better identification of the indentation cracks. The specimens were then poled longitudinally (along the largest dimension) with an electrical field of 2 kV/mm. Some specimens were kept “as sintered”, i.e. in the non-poled state, for comparison studies.

Thermo-mechanical testing

Indentation tests were performed in both poled and non-poled specimens using a Vickers indenter (Zwick GmbH, Ulm, Germany) up to a maximum load of 9.8 N and holding time of 10 s. At least 3 indentations were placed along the polished surface of each specimen in a way that the resulting cracks were parallel or perpendicular to the longitudinal direction, as illustrated in Fig. 1. The cracks were then measured with an optical microscope (Olympus Austria GmbH, Vienna, Austria), before conducting the thermo-mechanical tests, in order to have a reference crack length value. After the tests, new indentation cracks were performed in the tested specimens and the crack lengths measured to be compared with the initial indentation crack lengths.

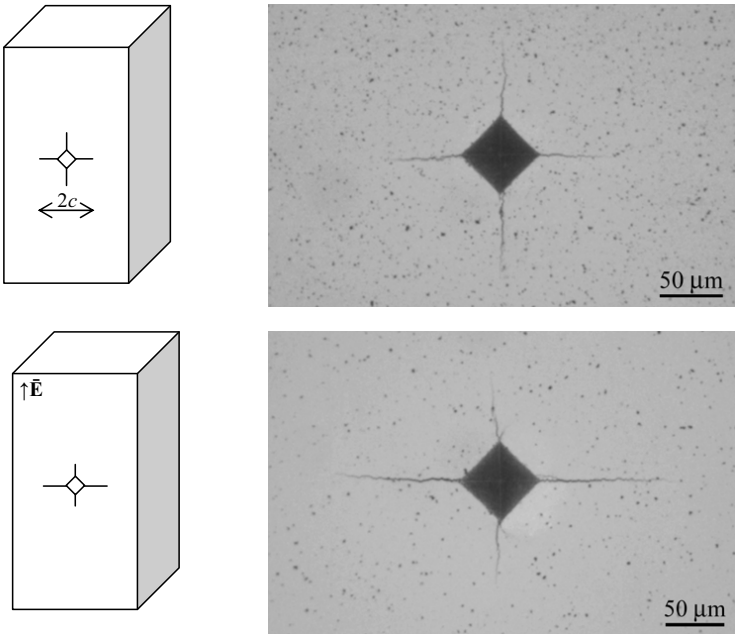

Figure 1. Indentation cracks parallel and perpendicular to the longitudinal direction in a) non-poled specimens and b) poled specimens, before the thermo-mechanical tests.

The influence of the temperature and the mechanical loading in the crack growth resistance of PZTs was assessed by means of a temperature chamber (Carbolite GmbH, Ubstadt-Weiher, Germany) coupled to a universal testing machine (Messphysik Materials Testing GmbH, Fürstenfeld, Austria). The temperature tests were performed following a ramp with a heating rate of 2°C/min and a dwell of 30 minutes at the aimed temperature. The selected testing temperatures were 25 °C (RT), 75 °C, 150 °C, 300 °C, and 400 °C.

The compression tests were performed at a loading rate of 0.5 mm/min and the maximal load was maintained for 1 min. The specimen was set in the machine with the longitudinal axis (poling axis) parallel to the loading axis. In order to investigate the pure effect of temperature in the switching of domains in poled specimens, the tests were also performed without mechanical stress (i.e. 0 MPa); non-poled specimens were also tested at the same temperatures for comparison. Further tests were performed applying mechanical stresses of -25 MPa and -50 MPa, based on realistic loading scenarios of MPA devices in service.

The combination of temperature and mechanical stress (compression) was assessed with individual compression tests (i.e. -25 MPa and -50 MPa) for each selected testing temperature.

Results and discussion

Crack growth resistance evaluation

The crack growth resistance was evaluated for every testing condition following the relation proposed by Anstis *et al.* using the IM method [12], by measuring the length of the indentation cracks both parallel ($2c^{//}$) and normal ($2c^{\perp}$) to the longitudinal axis of the specimen:

$$K_{\mathrm{Ic}} = \chi \cdot \frac{P}{c^{3/2}} \tag{1}$$

Where χ is a constant related to the shape of the indentation crack, P is the indentation load and c is half the length of the measured indentation crack, as depicted in Fig. 1.

In order to estimate the χ constant, using the IM method, the value of K_{Ic} (intrinsic to the material) should be known. Assuming an "as-sintered" state for the PZT material, five single edge V-notched beam (SEVNB) specimens were prepared for the determination of K_{Ic} (see Fig. 2). During the notching procedure, cracks developed at the notch tip, yielding an effective single edge pre-cracked beam (SEPB) configuration.

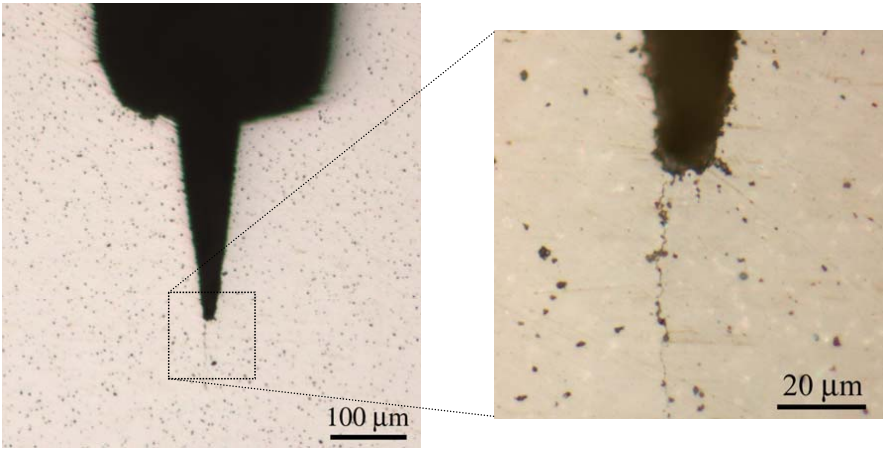

Figure 2. Pre-notched PZT bulk specimen for fracture toughness determination. A crack has developed during the notching process, yielding a single edge pre-crack beam (SEPB) configuration.

Four point bending tests (outer and inner spans of 30 mm and 15 mm, respectively) were performed in 40 x 3.5 x 4 mm^3 specimens following the norm standards ENV-843-1 [13] and the fracture toughness was evaluated according to [14]:

$$K_{\mathrm{Ic}} = \sigma_f \cdot Y \cdot \sqrt{a} \tag{2}$$

Where σ_f is the failure stress (in MPa), a is the crack length (in m) and Y is a geometric factor defined for an edge crack and given by [15]:

$$Y(\delta) = \frac{1}{(1-\delta)^{3/2}}\left[1.9887 - 1.326\delta - \frac{\delta(1-\delta)}{(1+\delta)^2}(3.49 - 0.68\delta + 1.35\delta^2)\right]; \quad \delta = a/W \tag{3}$$

A fracture toughness value K_{Ic} of 1.09 ± 0.08 MPam$^{1/2}$ was obtained. Thus, taken this value as the toughness value for the PZT material, the parameter χ was determined using Eq. (1) by measuring the crack length of an indentation load of 9.8 N, resulting in a value of 0.105 ± 0.008.

Temperature effect on poled and non-poled specimens

The effect of temperature on the crack growth resistance of poled and non-poled PZT specimens is presented in Fig. 3. The fracture toughness calculated with Eq. (1) for the non-poled specimens remains constant after heating the specimen up to the testing temperatures (reaching a maximal difference of 3.5% between RT and 400 °C) and isotropic (i.e. same crack length parallel and normal to the longitudinal axis). On the other hand, for the poled specimens, a clear toughness anisotropy can be appreciated as a function of crack orientation (i.e. parallel or normal to the poling axis). The maximum and minimum toughness values are reached at room temperature in direction parallel and normal to the poling axis respectively, resulting in $K_{Ic}^{//}$ = 1.51 ± 0.02 MPam$^{1/2}$ and $K_{Ic}^{\perp}$ = 0.62 ± 0.01 MPam$^{1/2}$. As the temperature increases, $K_{Ic}^{//}$ and $K_{Ic}^{\perp}$ vary slightly leading to smaller and higher toughness values respectively, reaching a minimum and a maximum values after the Curie temperature of $K_{Ic}^{//}$ = 0.99 ± 0.02 MPam$^{1/2}$ and $K_{Ic}^{\perp}$ = 1.00 ± 0.02 MPam$^{1/2}$ respectively.

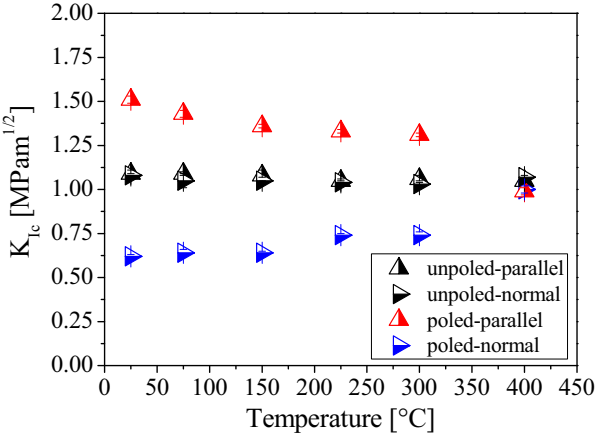

Figure 3. Crack growth resistance measured in poled and non-poled PZT specimens as a function of temperature. Whereas the fracture toughness remains constant and isotropic (i.e. same crack length parallel and normal to the longitudinal axis) in the non-poled specimens after the testing temperatures, an effect of the temperature in the toughness of the poled specimens can be appreciated, leading to partly depolarisation.

The change in fracture toughness with temperature is associated with the domain switching process activated by the thermal loading. It can be inferred from Fig. 3 that from temperatures above 75 °C some of the domains (initially oriented parallel to the longitudinal/poling axis) may have recovered their original orientation (depolarisation effect), thus affecting the initial crack growth resistance of the material. It can be also seen that the decrease in toughness in the parallel direction is counterbalanced by an increase in the normal direction. Above the Curie temperature, the material is no longer polarised, and thus the toughness measured after 400 °C is almost equal to the toughness of a non-poled material, i.e. $K_{Ic} = 1.09 \pm 0.02$ MPam$^{1/2}$.

Thermo-mechanical effect on poled specimens

The effect of mechanical loading (compression along the longitudinal axis) combined with the temperature effect on the crack growth resistance of poled PZT specimens is presented in Figs. 4 and 5 for a mechanical stress of -25 MPa and -50 MPa respectively.

The crack growth resistance measured in poled PZT specimens after a mechanical stress (compression) of -25 MPa as a function of temperature shows a slightly change in the toughness values for temperatures between 25 °C and 150 °C (Fig. 4). In this regard, a significant change can be clearly seen for higher temperatures. For instance, at temperatures around 300 °C, the toughness value in parallel and normal direction, i.e. $K_{Ic}^{//} = 1.04 \pm 0.02$ MPam$^{1/2}$ and $K_{Ic}^{\perp} = 0.99 \pm 0.03$ MPam$^{1/2}$ respectively, is practically the same as that of the non-poled specimens (Fig. 1). The material has almost been brought to a state similar to the "as-sintered" state. Another interesting effect, which can be inferred from Fig. 4, is that for temperatures above the Curie point (e.g. 400 °C), the applied compressive stress along with the high temperature lead to a kind of mechanical polarisation in direction normal to the longitudinal axis, yielding opposite toughness anisotropy (as compared with the initial poling state) in the material.

For the case of poled PZT specimens, the mechanical stress (compression) of -50 MPa applied along the direction of poling yield a significant depolarisation effect even at low temperatures. Similar to the previous case, at temperatures ca. 225 °C, the toughness value in parallel and normal direction (i.e. $K_{Ic}^{//} = 1.04 \pm 0.02$ MPam$^{1/2}$ and $K_{Ic}^{\perp} = 1.04 \pm 0.01$ MPam$^{1/2}$ respectively) coincides with the toughness of the non-poled specimens (Fig. 1); the material has been depolarised. From this temperature (i.e. >225 °C) and due to the relative high compressive stresses (-50 MPa), the toughness anisotropy reverses, yielding higher crack growth resistance in direction normal to the longitudinal axis (Fig. 5). Opposite to the previous case, this phenomenon can be already observed below the Curie temperature. For temperatures beyond the Curie point (e.g. 400 °C), the toughness normal to the longitudinal direction can reach values up to $K_{Ic}^{\perp} = 1.75$ MPam$^{1/2}$.

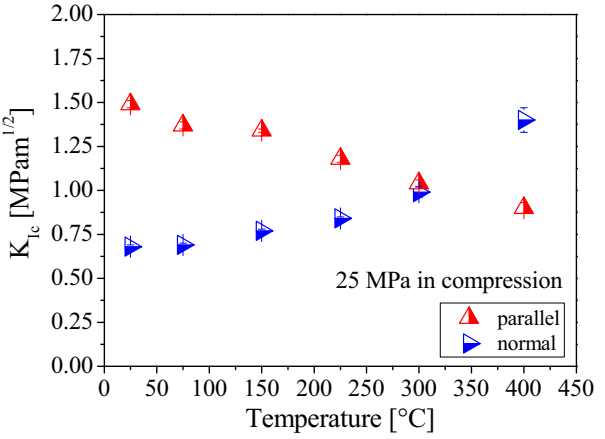

Figure 4. Crack growth resistance measured in poled PZT specimens after a mechanical stress of -25 MPa (compression) as a function of temperature. Although a slightly change in the toughness values can be appreciated for temperatures between 25 °C and 150 °C, the most significant change is clearly seen at 300 °C. At this temperature, the toughness value in parallel and normal direction is practically the same as that of the non-poled specimens. Above the Curie temperature the toughness anisotropy is reversed.

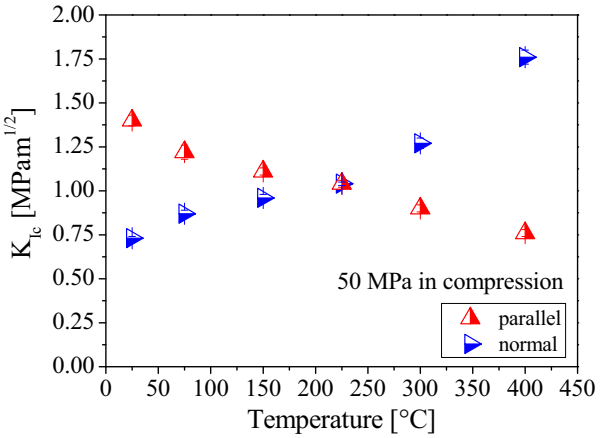

Figure 5. Crack growth resistance measured in poled PZT specimens after a mechanical stress of -50 MPa (compression) as a function of temperature. The mechanical stress applied against the direction of poling yield a significant depolarisation effect even at low temperatures. The crack growth resistance of the PZT reverses already for temperatures above 225 °C, reaching values up to 1.75 MPam$^{1/2}$ for 400 °C.

Summary

The toughness anisotropy of a commercial soft PZT material has been determined using the IM method as a function of temperature and mechanical stress on electrically poled specimens. The toughness values reached at room temperature in direction parallel and normal to the poling axis, respectively, resulted in $K_{Ic}^{//} = 1.51 \pm 0.02$ MPam$^{1/2}$ and $K_{Ic}^{\perp} = 0.62 \pm 0.01$ MPam$^{1/2}$, as compared with the toughness of a non-poled specimen, $K_{Ic} = 1.09 \pm 0.02$ MPam$^{1/2}$, taken as a reference. The increase in temperature leads to a light, gradually depolarisation of the material, until the Curie temperature is reached and the material is full depolarised. The additional mechanical compressive stress enhances such depolarisation effect with the temperature, yielding to a full depolarised material even below the Curie point. It has been shown that the combination of thermo-mechanical stresses influences the crack growth resistance of a poled PZT material, what should be considered for the structural reliability of MPAs against crack propagation.

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