ANISOTROPY OF FRACTURE TOUGHNESS OF PIEZOELECTRIC CERAMICS

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ABSTRACT

A model has been suggested to explain the observed nature of anisotropy of PZT- and barium titanate-based piezoceramics, based on an assumption of pressure-induced occurrence of a domain reorientation zone near the crack tip.

KEYWORDS

Fracture toughness, elastic anisotropy, domain reorientation, pressure-induced zone, crack tip, deformation energy.

INTRODUCTION

B.Jaffe, Cook and H.Jaffe (1971) discovered that piezoelectric ceramics features a transversal-isotropic symmetry of elastic properties. Fracture toughness of materials of such a symmetry class depends, according to results of Sih, Paris and Irwin (1965), upon the crack propagation direction and is characterized by two fracture energy values: Γ_r in the plane containing the symmetry axis and Γ_z in the perpendicular plane. The fracture toughness of piezoceramics has been determined by Freuman, McKinney and Smith (1974) and Bruce, Gerberich and Koepke (1978) by means of traditional fracture mechanics, suitable for isotropic ceramic materials, without taking into account the nature of the anisotropy and also a feasible influence of the piezoelectric effect. Because of that, this data on fracture toughness are contradictory. Also there is no common opinion about the microstructural mechanism, responsible for the fracture toughness anisotropy of the piezoceramics.

Materials, Experimental Procedure and Results

Two lead zirconate-titanate solid solutions-based (composition I) being barium-doped, and the other niobium- and tungsten-doped (composition II), and also a titanate barium-based composition doped with calcium (composition III), were taken for examination.

All the compositions are tetragonal below the Curie point, but feature different ferroelectric rigidities: composition II is ferroelectrically soft while compositions I and III are of an intermediate ferroelectric rigidity.

Since anisotropy of elastic properties and coupling of electrical and mechanical fields are typical for piezoceramics, they were accounted for in stress intensity factor (SIF) calculations in order to obtain a correct value for the fracture toughness. The presence of piezoeffect lowers the SIF value for both directions of crack propagation as compared to its value for an elastic medium. However, this lowering is insignificant and may be neglected in the SIF calculations for double torsion specimen.

Experiments on determining the piezoceramic fracture toughness by double torsion method were conducted on the "Instron TT-k 1126" testing set. For every crack propagation direction the critical SIF value was determined on 5 to 7 specimens. The tests were conducted in air at t° = 19 to 21°C and relative humidity of 70 to 80%.

In determining the fracture toughness by the indentation of the Vickers diamond pyramid formulas were used which relate the load applied to the indenter and the length of radial cracks at the impression with K_c as was done by Evans and Charley (1976).

The results of K_{IC} measurements by both methods are given in the Table.

TABLE 1 Results of SIF Measurement

Material	Double torsion: MPa x $m^{1/2}$		Indentation: MPa x $m^{1/2}$		Fracture energy, J/m²	
	K_{Ic}^{rz}	K_{Ic}^{zr}	K_c^{rz}	K_c^{zr}	Γ_r	Γ_z
Composition I	1.35	-	1.50	0.70	10.7	3.8
Composition II	-	-	1.82	0.77	22.2	6.4
Composition III	1.52	1.38	-	-	-	-

DISCUSSION

The results of fracture toughness measurements for cracks directed parallel and perpendicular to the axis of residual polarization, obtained by both double torsion and indentation methods, coincide. Meanwhile the fracture toughness data obtained by Bruce, Gerberich and Koepke (1978)are contrary to those obtained in the present study. The cause of this discrepancy is the use of an incorrect formula for SIF calculations. The dependence of the piezoceramic fracture toughness upon the direction of crack propagation with respect to the axis of residual polarization can be attributed to two possible mechanisms. The investigation mentioned above used the traditional approach which consists in a direct influence on K_{IC} of internal stresses arising from 90° domain reorientations during polarization. In this case the true fracture toughness is assumed as being constant and the deviations are explained as the result of the internal stress field effect upon the crack. Rice and others (1978) found that a similar mechanism results in a change of the fracture energy of ferroelectric ceramics in a ferroelectric phase as compared to that in a paraelectric phase. However, the deformation of piezoelectric grains at polarization of ferroelectric ceramics in contrast to that at a phase transition, is of a directional nature. Polarization results in an elongation of the piezoceramic element in the field direction (z-axis) which should give rise to compressive internal stresses along this axis and suppress the crack development along the perpendicular direction (r-axis) as compared to that along the z-axis. As our experiment gives an opposite result the interpretation of the observed character of fracture toughness anisotropy on the basis of a simplified analysis of the internal stress distribution is unacceptable. An interaction between the crack and the predominantly polarization-oriented 90° domain walls may be regarded as another feasible mechanism. The domain wall, being a twin boundary, forms an obstacle for crack development and inhibits its propagation in the z-direction on the one hand, whereas, being a defect concentrator it promotes the crack movement in the perpendicular direction on the other one. Such a mechanism explains correctly the observed anisotropy of the critical SIF of piezoceramics, but is opposed by at least two facts indicating against it. Firstly, according to the data of investigators mentioned above, no essential difference between the structures of fracture surfaces along and across the axis of residual polarization has been found. These results of LTZ piezoceramics microstructure studies indicate only the decrease in concentration of domain walls in the material, but not a change in their preferential orientation. Secondly, the character of the K_{IC} anisotropy observed by Kovalev (1981) occurs both at intragranual and at intergranular types of fracture (PZI-based composition). In the last case the interaction with the grain substructure cannot be used to explain the difference in the K_{IC} values for r- and z- directions of crack development. Both above mechanisms are one-sided whereas, in ferroelectric ceramics the internal stress mechanisms and the interaction with the substructure are closely related with each other and their influence cannot be separated.

The response of ferroelectric ceramics to external effects is determined by the mobility of domain structure of grains. It is well known that deformation in this case occurs through domain processes, namely through displacement of 90° domain walls under stress. Near the tip of the propagating crack a region of high stress concentration exists, where pressure-induced structural transitions of the twinning type are feasible. Consider a model of structural transition of a piezoceramic grain under a stress near the crack tip. For simplicity, we will neglect the temperature dependence and relaxation nature of the domain process-induced deformation and also the associated electroelastic effects.

Suppose that after the polarization process some piezoceramic grains are oriented along the field direction by a total elimination of 90° domain walls, i.e. they are monodomain ones. The rest of the grains, where no 90° domain switching occurred,

can be in following states: (a) field direction-oriented before the polarization; (b) reoriented by a 180° switching without deformation; (c) remain in the previous randomly oriented positions. Thus (in the first approximation) the polarized ceramics may be treated as a composite material where grains not deformed by polarization form a matrix, whereas monodomain grains, field-direction-oriented by elimination of 90° domain walls, are metastable inclusions. Assuming the domain processes initiated by the crack to be the cause of origination of the fracture toughness anisotropy of the ferroelectric ceramics in a polarized state, we consider a model of a feasible structural transition of a polarization direction-oriented monodomain grain. The feasibility of such a stress-induced process and the assessment of its influence upon the fracture toughness magnitude may be calculated adopting the procedure used by Evans and Heuer (1980) for modeling a phase transition of metastable zirconium oxide inclusions near the crack tip.

Since the strains associated with the displacement of a 90° domain wall are a twinning shear, they can be calculated according to a simple scheme of Esaklul, Gerberich and Koepke (1980). For a plane-strain case we obtain that grain strains in a structural transition from a metastable (monodomain) to a stable (polydomain) state are

$$\varepsilon_{11} = 0, \quad \varepsilon_{22} = 0, \quad \varepsilon_{12} \approx \frac{c-a}{c+a} \tag{1}$$

Considering the thermodynamical condition of a structural transition of a single grain, we have

$$\Delta G = -\Delta G_0 + \Delta U_1 \leq 0 \tag{2}$$

where ΔG is the change in free energy of the "inclusion-matrix" system and $\Delta U_1 = \Delta U + W$ is the deformation energy, accumulated in the structural transition, which includes the internal stress energy ΔU caused by strains (1) because of grain constraint in the matrix and the work W of external crack-promoted stresses on strains accompanying the structural transition. Apart from the deformation energy formula (2) includes also a parameter $\Delta G_0 > 0$ which corresponds to the energy absorbed by the structural transition of the grain in a free state. While ΔG_0 at a phase transition is the difference of chemical free energies of the phases, here we take ΔG_0 as equivalent to the grain twinning energy. The magnitude of ΔG_0 depends upon the ferroelectric rigidity of the composition. In the absence of external stresses we obtain from inequality (2) that

$$\Delta U \leq \Delta G_0 \tag{3}$$

This is the condition for a spontaneous structural transition which shows that for grains satisfying condition (3) the structural transition from a monodomain polarized state to a polydomain depolarized state will occur in the absence of external stresses. This is observed by Jaffe, Cook and Jaffe (1971) in practice directly after polarization. For other metastable grains the G value lies within the range

$$0 < \Delta G_0 \leq \Delta U \tag{4}$$

Considering the terms in (2) reduced to a unit volume, we obtain the limiting condition of a structural transition as

$$\Delta U + K_I W(\rho) = \Delta g_0 \tag{5}$$

where $\Delta U = U r^{-2}$, $\Delta g = \Delta G_0 r^{-2}$ are specific energies of deformation due to internal stresses and of twinning, respectively; $W(\rho) = W/K_I r^2$ is the specific work of external stresses from a crack with $K_I = 1$, situated at a distance ρ from the centre of the grain undergoing the transition; and r is the grain radius. Solving (5) for ρ, we obtain in the first approximation the assessment of the size of the zone of inelastic deformation caused by domain processes at the crack tip.

The value of ΔU for ferroelectric ceramics of specific composition as well as the form of the $W(\rho)$ function can be calculated by a numerical procedure of the finite element method. To this end, we make use of a classical inclusion-matrix calculation scheme. The stressed state in the vicinity of an inclusion is caused by the assignment inside the V_1 region of initial strains $\bar{\varepsilon}_{ij}$ corresponding to a twinning shear (1). The work W of crack stresses in the structural transition process is readily calculated by the following scheme. In calculating the V region by means of isoparametric finite elements as Zienkiewicz suggested (1971) we assign in the region integration points singular initial stresses caused by the crack presence at a distance ρ from the inclusion centre:

$$\bar{\sigma}_{ij}(\rho_s) = \frac{K_I f_{ij}(\theta_s)}{\sqrt{2\pi \rho_s}} \tag{6}$$

where ρ_s and θ_s are polar coordinates of the integration point with respect to the crack tip and f_{ij} are determined via trigonometrical functions. Then the deformation energy of the finite element model can be found by numerical integration of the expression

$$U_1 = \frac{1}{2}\int_V \sigma_{ij}(\varepsilon_{ij} - \bar{\varepsilon}_{ij})\,2V + \int_V \bar{\sigma}_{ij}\,\varepsilon_{ij}\,dV \tag{7}$$

where the first and the second terms are ΔU and W respectively. Calculating the deformation energy of the finite element model with initial stresses (6) by means of (7) with variable distance ρ, we obtain discrete values of the $W(\rho)$ function. To simplify the further procedure, we select the function as

$$W(\rho) = \frac{C}{\sqrt{\rho - \rho_0}} \tag{8}$$

which adequately approximates the discrete values obtained by the finite element method. The form of the function calculated for two cases of crack propagation in a piezoceramic specimen relative to the direction of residual polarization P_0 is shown in Fig.1. It follows from equations (5) and (8) that

$$\sqrt{\rho - \rho_0} = \frac{C K_I}{\Delta U(\xi - 1)} \tag{9}$$

where parameter $\xi = \Delta g_0 / \Delta U$ and, according to (4), $0 < \xi < 1$. It is seen that the closer is the grain to the spontaneous transition ($\xi \to 1$) the longer is the distance ρ. The analysis (9) indicates that the structural transition of the grain at the crack tip is possible only at $W < 0$, when the deformation energy U_1 decreases because of the work of initial

stresses (6) caused by the crack presence.

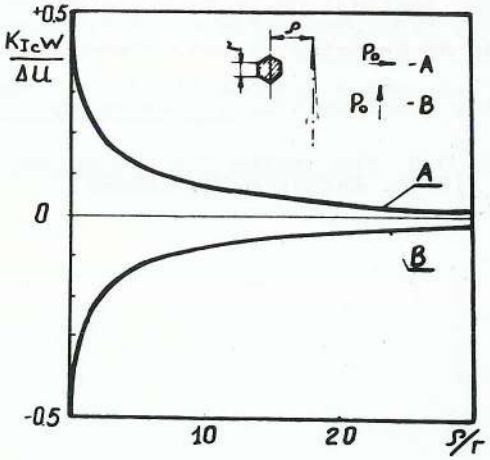

Fig. 1. Variation of work W of crack stresses at structural transition of grain vs ρ/r ratio for two directions of crack propagation a) across P_o b) along P_o for composition I(^{o}r=2.5мm)

From Fig.1 it follows that the supposed transition of the grain is favored only by that crack which propagated along the direction of residual polarization. In the vicinity of its tip, a zone of an additional energy dissipation due to stress-induced structural transitions is formed. The crack movement in the perpendicular direction will, on the contrary, inhibit the domain processes in the nearest grains, and the fracture will occur without additional energy dissipation. The size of the inelastic deformation (twinning) zone in the first case can be calculated by formula (9). No such zone will appear in the second case. Thus, qualitative result predicted by the model agrees with that observed experimentally. The use of formula (6) for numerical calculation of the width of the domain reorientation zone restricts the region of a correct determination within the framework of the model, adopted here. As Evans and Heuer (1980) indicate, the use of formula (6) is valid for $\rho \ll a$ where a is the crack length. Since in experimental fracture toughness determinations by the traditional methods of fracture mechanics (as in the present paper) the crack length greatly exceeds the characteristic dimension of the microstructure this condition is fulfilled. On the other hand, the finite element calculation of the work of crack stresses $\bar{\sigma}_{ij}$ on the structure transition strains ε_{ij} for $\rho \gg r$ may lead to an increase of the numerical integration error because of high $\bar{\sigma}_{ij}$ gradients. Therefore, ρ can correctly enough be determined in the $r < \rho \ll a$ interval.

The fracture energy rise due to the formation of additional energy dissipation zone at the crack tip can be calculated by a simple technique, similar to that used by Evans and

Heuer (1980). Denoting by Γ_z the fracture energy in the absence of this zone, in our case for the plane perpendicular to the axis of residual relaxation, we can write

$$\Gamma_r = \Gamma_z + \Delta U^* \cdot \rho \cdot p \tag{10}$$

where Γ_r is the fracture energy for the polarization plane. The energy ΔU^* absorbed in a single event of the structural transition of a grain of radius r is

$$\Delta U^* = \Delta U r^2 (1 - \xi) \tag{11}$$

If f denotes the share of unstable grains in piezoceramics, then p = 0.385 f/r^2 for hexagonal grains. Since $K_{Ic}^{rz\,2} = 2\Gamma_r S_{rz}^{-1}$, we obtain

$$\Gamma_r / \Gamma_z = \frac{1}{1 - \varphi} \tag{12}$$

where

$$\varphi = \frac{0.77 \cdot f \cdot c^2}{\Delta U (1 - \xi) S} \tag{13}$$

The f value can be chosen as a part of possible polarization-promoted 90° domain switchings.

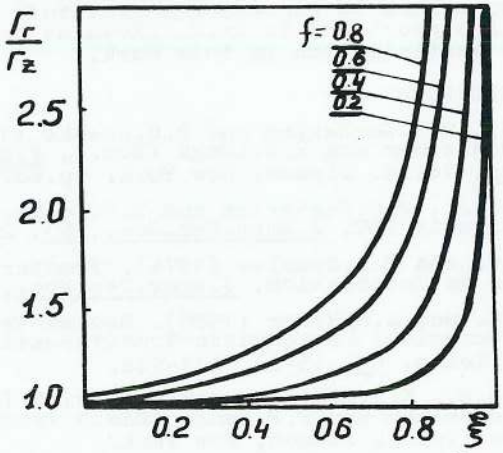

Fig. 2. Ratio of fracture energy Γ_r per Γ_z vs parameters ξ and f for PZT.

Fig. 2 illustrates dependence (12) for different ξ and f. The lack of data on the Δg_o value makes it difficult to compare quantitatively the model-predicted behavior with that observed. Analysing (12) with an account for (12) makes it possible to determine the degree of various factors upon Γ_r/Γ_z ratio. It is obvious that for ferroelectrically softer compositions with a relatively high parameter ξ values the difference between the fracture energies Γ_r and Γ_z will be more pronounced. This fact corresponds to the results of fracture energy measurements by the indentation method for compositions I and II. A similar conclusion can be made also for f, namely, the more polarization-promoted reorientations of 90° domains occur, the stronger will show up the anisotropy of

fracture toughness.

CONCLUSIONS

The measurements of fracture toughness of polarized ferroelectrical ceramics were conducted by double torsion and Vickers pyramid indentation methods with account for the anisotropy of elastic properties. The finite element calculations showed that in determining K_{Ic} by conventional methods of fracture mechanics the piezoeffect may be neglected with an insignificant error. However, the disregard of elastic anisotropy leads to qualitatively wrong results. The polarization process is shown to lead to different values of fracture toughness with respect to the axis residual polarization of a piezoceramic element. The fracture toughness along the axis of residual polarization is higher than that in the perpendicular plane. To explain the observed differences a model is suggested, based on an assumption of the occurrence of a stress-induced domain reorientation zone near the crack tip.

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