COMPUTER SIMULATION OF MICROSTRUCTURE PROCESSES BY FERROELECTRIC CERAMIC SINTERING AND FRACTURE

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ABSTRACT

Computer simulation of the microstructure and strength processes taking place in PZT ceramics by sintering and following fracture due to growing macrocrack is developed. As shielding macrocrack mechanism it's considered the phase transformation of the secondary phase inclusions in the vicinity of the crack tip caused by growing macrocrack stress field. Estimations of fracture energy alterations are received and condition of the catastrophic crack growth is discussed.

KEYWORDS

Recrystallization, shrinkage, spontaneous microcracking, phase transformations, macrocrack propagation

INTRODUCTION

Creation of new ferroelectric ceramics (FC) demands of investigations revealing causes of strength and fracture toughness variations. Microcracking and phase transformations taking place in the FC to render a main effects on the parameter alterations. As rule, the optimization strength problem is divided on the some separate studies, namely: 1) investigation of the spontaneous microcracking by cooling; 2) crack interactions with defect distributions; 3) evaluation of the crack resistance caused by microcracking or (and) phase transformations near growing macrocrack. However a material optimization demands a joint study of these problems.

The aim of this report consists in discussion of catastrophic crack growth condition and in estimation of fracture energy change in dependence on the presspowder initial porosity, intergranular cracking and phase transformations on a basis of the computer simulation.

Computer model corresponds to the next processes: 1) microstructure formation by sintering; 2) spontaneous cracking by cooling; 3) macrocrack propagation taking into account an energy absorption thanks to phase transformations near crack.

CERAMIC SINTERING AND COOLING MICROFRACTURE

We used the physical model of gradient sintering for PZT ceramics (Belyaev et al., 1989). It was considered the first basic problem for quasilinear equation of heat conductivity with FC microstructure formation, together (Karpinsky and Parinov, 1992). It was supposed that sintering front with unit thickness was performed as a two-dimensional lattice containing 1000 cells with characteristic size Δ=10μm arranged in square pattern. Every cell corresponded to powder granular or pore. We considered a presspowder sample in the furnace. It was assumed that the temperature distribution in the furnace T depended on one coordinate X and consisted of a constant and linear temperature curve. Initial porosity C_p in the sample was defined a priori. Computer model contained next steps: 1) study of the thermal front propagation and definition of the sintering region; 2) powder recrystallyzation model in this region; 3) microstructure shrinkage.

Spontaneous microcracking of grain boundaries was modeled by cooling. Above the Curie temperature the ferroelectrics have a cubic structure and the microfracture is caused by the considerable thermal gradients, only (Karpinsky and Parinov, 1992). Here we neglected by effect of elastic anisotropy (Tvergaard and Hutchinson, 1987). Below the Curie point the residual stresses are appeared due to the deformation phase mismatches and thermal expansions between adjacent grains (Pisarenko, 1987). Deformation phase mismatch is a main cause of the FC microcracking by cooling in this temperature interval. Computer models of microfracture have been developed earlier (Karpinsky and Parinov, 1991; Karpinsky et al., 1992; Parinov, 1992).

MACROCRACK IN THE CERAMIC

Growing macrocrack in the FC has been investigated on a basis of the graph theory by Karpinsky and Parinov, 1991. In this paper the different fracture mechanisms (intergranular, transgranular and mixed fracture) were considered and a crack interactions with microcracks, pores and grain phases were investigated, too. Karpinsky et al., 1992; Parinov, 1992 studied an influence of the microcrack process zone on the various microstructure and strength parameters. A real crack trajectory corresponded to the minimal trajectory length (see Fig.1) Effective surface fracture energy γ_o, connected with fracture toughness K_{1c} by formula $K_{1c}=\sqrt{2E\gamma_o}$, was given by

$$\gamma_o = L\,\gamma_b/h \qquad (1)$$

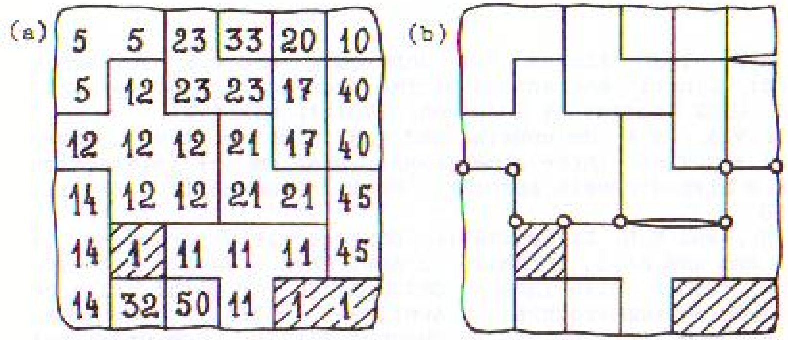

Fig. 1. Example of a macrocrack propagation in the FC model sample (a) the represantation of FC structure fragment in the computer; (b) a macrocrack propagation in the microstructure (circles), spontaneous cracks are shown on the grain boundaries, porosity are shaded.

where E is Young's modulus; γ_b is the grain boundary energy; h is the sample width. It was carried out for rectilinear intergranular boundary $\gamma_o=\gamma_b$.

Other crack resistance mechanism is studied in this report. It's connected with existence of the phase transformations caused by the internal stresses near growing macrocrack. Crack growth in the FC is defined by its domain structure and by existence of many phases. Studies of crack propagations through domain boundaries have been shown a necessity of calculation them parameters for prognosis of FC strength and fracture toughness (Karpinsky et al., 1981). Domain reorientation influence on the fracture toughness was investigated by Pisarenko, 1987 on the structure model in the vicinity of a crack tip. Analogous phase transition processes are typical for ceramics reinforced by partially-stabilized zirconia particles (PSZ) (Porter et al., 1979). In these materials it's possible a phase transformation from tetragonal to monoclinic crystal structure under stresses. Fracture toughness alterations are caused by energy absorption by the phase transformation (PT) zone surrounding growing crack tip. It's known that PZT ceramics have two phases in the morphotropic transitions regions (MTR), namely: tetragonal (T) and rhombohedral (R) ferroelectric phases. Volume fraction phase correlations are defined by technological heterogeneities and by fluctuations of the component concentrations. They are caused by electrical fields and by mechanical stresses (Isupov, 1980).

Stresses in the vicinity of the crack tip in ceramic of MTR boundary composition can to initiate the phase transitions in the ceramic grains. We use the energy balance method (Cherepanov, 1974) for qualitative evaluation of the crack unstability conditions at PT zone presence. Then for stressed sample is carried out

$$\delta \mathcal{E} + \delta W = \delta J \qquad (2)$$

where $\delta\mathcal{E}$, δW, δJ are the selected elastic energy; absorbed energy owing to phase transformation and surface energy growth due to the crack propagation on the length $2\delta l$, accordingly. We consider infinite, elastic, plane, single phase body with finite crack by length $2l$, growing under monotonically increasing load σ.

Suppose that the phase transformation energy $W(l)$ is equal to sum of the own strain energy for secondary phase inclusions and interaction energy of inclusions with elastic crack field (Grekov et al., 1987)

$$W(l) = W_0 \int_{\Omega} C(\bar{r})d\bar{r} + \int_{\Omega} C(\bar{r})W_{int}(\bar{r})d\bar{r} \qquad (3)$$

Here Ω the transformed zone and the boundary of this region given by equation

$$R_{\Omega} = K_1^2\cos^2(\varphi/2)(1+\sin(\varphi/2))^2/(2\pi\sigma_{1c}^2) \qquad (4)$$

where $C(\bar{r})$ is the secondary phase fraction in Ω. Here K_1 is the stress intensity factor; $\bar{r}=\{r,\varphi\}$ are crack-tip polar coordinates the transformed boundary; σ_{1c} is the critical stress.

Then we define a dependence between W and l. It's caused by dependence of the PT zone size and secondary phase density on a crack length $2l$ and on a value of the σ. Hence, it's important a physical basis of the PT zone boundary definition and inclusion concentration in its, caused by stress fields at crack tip. So, it was found by Isupov, 1981 a volumetric phase correlation in PZT ceramic at condition of the mechanical stress homogeneity and supposition of an isotropic distribution of the orientation crystallographic direction angles in grains relatively to infinite force direction. Ceramic grain has rhombohedral or tetragonal symmetry at uniaxial loading. The choice is defined by maximal projection of the lattice distorsion vector on tensile loading direction. Thus, it was proposed that the phase transformation condition in PZT ceramics is determined by tensile stress directed along of the elementary cell edge and exceeding a critical stress.

$$\sigma_1\cos^2\alpha \geq \sigma_{1c} \qquad (5)$$

where σ_1 is the main maximal stress near crack tip

$$\sigma_1 = (K_1/\sqrt{2\pi r})\cos(\varphi/2)(1+\sin(\varphi/2)) \qquad (6)$$

α is the minimal plane angle between direction of the σ_1 and $\{100\}$ at given point.

It was supposed that the FC had rhombohedral symmetry. Then it's existed the value σ_{1c} which depend from rhombohedral distorsion value of lattice or from ceramic composition situation at the phase diagram relatively of morphotropic boundary. In general case a direction of the σ_1 don't coincide with edge of rhombohedron because ceramic grains are orientated randomly. Rhombohedral distorsion of PZT pseudocubic lattice is only a few (less 30′) (Yaffe et al., 1971). Then we considered a simple model, where a ceramic was performed by the randomly orientated grain distribution in a cubic matrix before loading. Analogously to Isupov, 1981 it was supposed an existence of the homogeneous distribution of grain orientations independent from neighbours. This supposition neglected by natural ceramic anysotropy which took place due to preparation processes.

We proposed a texture absence in FC sample and used a problem about cube misorientation (Mackenzie and Thomson, 1957; Mackenzie, 1958). Then a misorientation angle distribution density for crystallographic direction $\{100\}$ is equal to

$$dP(\alpha) = (8/(5\pi))\sin(\alpha)(\pi^2/32 - (1/\sqrt{2})\tan(\alpha/2))d\alpha, \quad 0 \leq \alpha \leq \beta$$

$$dP(\alpha) = 0, \quad \alpha > \beta, \quad \cos\beta = 2/3 \qquad (7)$$

A secondary phase concentration $C(\bar{r})$ is given by

$$C(r,\varphi) = (1/d)^3\int_0^{\theta} dP(\alpha), \quad \theta = \arccos(\sqrt{\sigma_{1c}/\sigma_1}) \qquad (8)$$

where d is the mean grain size. Combining expressions (7) and (8) one can to obtain

$$C(r,\varphi) = (1/d)^3(\pi(1-\sqrt{\sigma_{1c}/\sigma_1})/20 - (8/(5\pi\sqrt{2}))(\arccos(\sqrt{\sigma_{1c}/\sigma_1}) - \sqrt{1-\sigma_{1c}/\sigma_1})) \qquad (9)$$

then an addition to δJ caused by phase transformation energy is equal to

$$W(l) = W_0\int_0^{\pi}\int_0^{R_\Omega} C(r,\varphi)r\,dr\,d\varphi + \int_0^{\pi}\int_0^{R_\Omega} W_{int}(r,\varphi)C(r,\varphi)r\,dr\,d\varphi = A\sigma^4 l^2 \qquad (10)$$

In the (10) W_0 and W_{int} were defined in form (Kunin, 1975)

$$W_0 = 2d^3(1+\nu_0)\mu_0 M^2/((1-\nu_0)(1-2(1-2\nu_0)\chi_1/(3(1-\nu_0)(\chi_0+\chi_1))));$$

$$W_{int} = VM_0\sigma_{ii}; \quad \sigma_{ii} = (2K_1/\sqrt{2\pi r})\cos(\varphi/2);$$

$$M_0 = M/(1-2(1-2\nu_0)/(3(1-\nu_0))); \quad M = \Delta V/(3d^3)$$

where χ is the compression modulus; ν is Poisson's ratio; μ is the shear modulus; V is the elementary cell volume; ΔV is the variation of the V by phase transition; indices 0,1 are corresponded to matrix and inclusion; A is the constant depending from average quasiisotropic elastic constants of the inclusion and matrix and difference of elementary cell volumes

(Shermergor, 1977). It's noted that from expressions (7)-(9) an existence of two phases in PT zone to follows simultaneo- usly by condition: $\sigma_{1c} \leq \sigma_1(\bar{r}) \leq 2.25\sigma_{1c}$. Then a value σ_1 is equal to infinity at $r \to 0$ from condition (6) but in reality σ_1 reaches a defined value σ_m ($\sigma_m \gg \sigma_1$). If $\sigma_m > 2.25\sigma_{1c}$ then crack tip is surrounded by T-phase fully initiated by internal mechanical stresses. For simplicity we limited a consideration by dilatation caused owing to volumetric vari- ation mismatches of both phases. Then we returned to (2) and proposed an ordinary expression

$$\delta G = \sigma^2 \pi l\, \delta l/(2E_o) \tag{11}$$

where E_o is Young's modulus for matrix. Hence it follows a balance energy equation

$$2A\sigma^4 l\, \delta l - \sigma^2 \pi l\, \delta l/(2E_o) = -4\gamma_b\, \delta l \tag{12}$$

In (12)we selected a solution by which σ was the positively and to decreased to zero at $l \to \infty$

$$\sigma = (\pi/(8AE_o) - (\pi^2/(64A^2E_o^2) - 2\gamma_b/(Al))^{1/2})^{1/2} \tag{13}$$

As it follows from (13) for sufficiently little crack sizes $l < l_c = 128A\gamma_b E_o^2/\pi^2$ any great external stress don't lead to catastrophic crack growth. Thus, safe cracks exist in the PZT in model limits. Analogous rezult but corresponding to dislo- cation plastic zone was received by Vilman and Mura, 1979. This results have interesting physical sense. In PT zone the tetragonal inclusions situated in more dense rhombohedral matrix to create the compressing stresses causing a macro- crack shielding. For short cracks $l < l_c$ energetics show a mo- re advantageous initiation of phase transformations near crack tip to compare with its catastrophic growth at any great stresses. Calculation gives a more high critical load to compare with Griffith's formula for crack length $l > l_c$. In particular, a critical load in our model for crack length $2l_c$ have factor $\sqrt{2}$.

Further, expressions (2), (11) and (13) give

$$\delta W = (\pi^2 l(1-(1-l_c/l)^{1/2})/(16E_o^2A) - 4\gamma_b)\, \delta l, \quad \text{at } l \geq l_c \tag{14}$$

hence $\delta W = 4\gamma_b\, \delta l$ at $l = l_c$ but at $l \gg l_c$ we decomposed an ex- pression $(1-l_c/l)^{1/2}$ on degrees l_c/l and to limited by three terms of the expansion

$$\delta W = (l_c/l)\gamma_b\, \delta l \tag{15}$$

In (14) an evaluation was received in supposition about mini- mal applied stress (the Griffith's stress) causing a catast- rophic crack growth. As it's seen from (14) and (15) a crack

length increase corresponds to expended energy decrease on the phase transformation initiation. It's noted that received es- timations have qualitative character, since they don't take into account a growing crack dynamics.

The secondary phase influence on the effective surface energy γ_p was estimated taking into account a small deflection the crack trajectory. We integrated an expression (15) on l in integration limits $[l_c, L]$ and result was divided on sample width h

$$\gamma_p = (l_c/h)\gamma_b \ln(L/l_c) \tag{16}$$

DISCUSSION OF CALCULATIONS

Results of computer simulation the PC properties in dependen- ce on initial porosity C_p were received. They are shown in the Table 1. Shrinkage koefficient $k_\Delta = n_{op}/(n_{op}+n_{cp}+n_{cr})$ grows with C_p. Here n_{op}, n_{cp}, n_{cr} is the cell number of the open, closed porosity and grain phase, respectively. However, dec- rease of the grain size R_c to compensates k_Δ increase.

Table 1. Computer simulation results.

Properties	Initial porosity, C_p, %					
	0	10	20	30	40	50
k_Δ	0.00	0.02	0.04	0.14	0.28	0.43
R_c/R_c^o	1.00	0.94	0.90	0.84	0.83	0.79
f_∞	0.12	0.11	0.10	0.10	0.10	0.09
γ_o/γ_b	1.25	1.00	1.00	0.82	0.88	0.90
γ_p/γ_b	0.17	0.17	0.17	0.16	0.15	0.15

This results to cause the close values of the closed porosity for all considered cases of the porous PC. Grain size decrea- se establishes a decrease of the spontaneous cracking fracti- on f_∞. Effective fracture energy γ_o/γ_b has not a monotonical dependence on C_p in phase transformation absence at the vici- nity of crack tip. It's reached minimum at the C_p=30%. Analo- gous discrepant dependence of the fracture energy on grain size has been observed by Pisarenko, 1987. Calculations for l_c gave the value 10µm. Then an influence of the phase trans- formations on the fracture energy γ_p/γ_b was approximately equally for all values of the C_p and they represented a small contributions in common fracture energy values.

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