INTERFACIAL ADHENSION OF PZT FERROELECTRIC THIN FILMS DETERMINED BY NANO-INDENTATION METHOD

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Summary We propose an elastic groundsill beam model with piezoelectric effect considered to assess the interfacial adhesion of ferroelectrics thin films, complemented and validated by nano-indentation fracture test on $Pb(Zr_{0.52}Ti_{0.48})O_3$ (PZT) thin films. It was observed from the load-indentation depth curves and atomic force microscopy (AFM) images that the fracture failure of PZT thin films induced by nano-indentations can be divided into three typical stages: no damage, bulging and spallation. The delamination of thin film systems was modeled as an interfacial crack propagation problem, with the energy release rate determined from the elastic groundsill beam model. For PZT thin films deposited on single Si substrate with thickness of 350nm and 450nm, the energy release rates per unit new crack area are in the range of 3.399~52.432J/m^2 and the phase angles are constant of 13.357^0.

INTRODUCTION

$Pb(Zr_xTi_{1-x})O_3$ (PZT) thin film is an important ferroelectric system with potential applications in MEMS, but the conventional models can not be used to analyse the interfacial adhesion and indentation fracture of ferroelectrics thin films accurately because of the piezoelectric effect. The description of fracture toughness of a poled piezoelectric ceramic is very complicated, involving fracture toughness measured perpendicular and parallel to the poling direction [1,2]. In this paper, we propose an elastic groundsill beam model to evaluate the interfacial adhesion of PZT thin films, characterized by energy release rate per unit crack area in nano-indentation test when the largest indentation depth is close to the film thickness. The piezoelectric effects are fully considered in the model. We conducted nano-indentation fracture test on PZT films deposited on single Si substrate by metal organic decomposition (MOD), using a cube corner indenter and atomic force microscopy (AFM). The spallations of the thin film were observed when indentation depths exceeded 13% of the film thickness and when the diamond indenter penetrated through the thin film into the substrate. Interfacial delamination is approximately consisted of the classical mode I and mode II cracks and the corresponding mode II crack dominates the interfacial crack in the ferroelectrics thin films.

THEORETICAL MODEL

We present a general theory for the axisymmetric indentation of transversely isotropic piezoelectric solids, governed by the following equations

$$\begin{cases}\sigma_{ij}=c_{ijkl}\varepsilon_{kl}-e_{kij}E_k\\ D_i=e_{ikl}\varepsilon_{kl}+\in_{ik}E_k\end{cases}\quad\begin{cases}\sigma_{ij,j}=0\\ D_{i,i}=0\end{cases}\quad\begin{cases}\varepsilon_{ij}=(u_{i,j}+u_{j,i})/2\\ E_i=-\phi_{,i}\end{cases}\qquad(1)$$

where, $\sigma_{ij}, D_i, \varepsilon_{ij}, u_i, E_i$ and ϕ are stress, electric displacement, strain, displacement, electric field and electric potential, respectively, and $c_{ijkl}, \varepsilon_{ik}, e_{kij}$ are elastic, dielectric and piezoelectric constants, respectively. The normal indentation of a piezoelectric thin films was considered as shown in Figs.1(a), where h, H, 2α and P_0 are thin film thickness, substrate thickness, apex angle and indentation load, respectively, and 2c is the largest imprint made on the surface of the film by the indenter. Combined with the assumption that the indenter is an insulator and does not reach the substrate, we have the relationship between the indentation load P_0 and the largest imprint 2c derived by Giannakopoulos and Suresh [3]. Because both the film thickness h and indenter contact region size c are in the nanometer scale, the approximations $\frac{\partial u_r}{\partial z}=\frac{\Delta u_r}{h}=\frac{u_r(r,0)}{h}$, $\frac{\partial u_z}{\partial r}=\frac{\Delta u_z}{c}=\frac{u_z(c,0)}{c}$, $E_z=-\frac{\partial\phi}{\partial z}=-\frac{\phi(r,0)}{h}$ are given. Using the above relationships and boundary conditions, the stress σ_{rr} in the indentation region of the thin film can be obtained as,

$$\sigma_{rr}=c_{11}\varepsilon_{rr}+c_{12}\varepsilon_{\theta\theta}+c_{13}\varepsilon_{zz}-e_{31}E_z\qquad(2)$$

We then present an elastic groundsill beam model as shown in Figs.1(b) to describe the indentation fracture characteristics of thin films, where the indentation depth is assumed to be close to the film thickness. When $\eta=\frac{h}{H}\to 0$, the constitutive equations of film and substrate are respectively given as

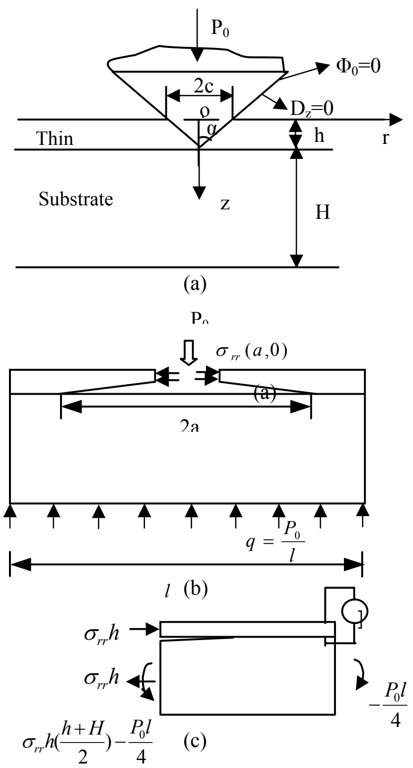

Fig. 1

$$\sigma_{rr}=\left(c_{11}-\frac{c_{13}^2}{c_{33}}+\frac{e_{13}^2}{\in_{33}}-\frac{c_{13}e_{13}e_{33}}{c_{33}\in_{33}}\right)\varepsilon_{rr}\quad 0\le z\le h.\qquad \sigma_{rr}=\frac{8\mu_2}{\kappa_2+1}\varepsilon_{rr}\quad h\le z\le h+H \tag{3}$$

which is derived from the composite beam theory, with $\kappa_2=3-4\nu_2$, ν_2 and μ_2 being the Poisson抯 ratio and shear modulus of substrate, respectively.

We consider the interface crack problem as shown in Figs.1(c) using the theory of composite beam. The interface intensity factors K_1 and K_2 reduce to the classical mode I and mode II factors K_I and K_{II},

$$K_{\rm I}=\frac{1}{\sqrt{2}}\left[Ph^{-1/2}\cos\omega+2\sqrt{3}Mh^{-3/2}\sin\omega\right],\quad K_{\rm II}=\frac{1}{\sqrt{2}}\left[Ph^{-1/2}\sin\omega+2\sqrt{3}Mh^{-3/2}\cos\omega\right] \tag{4}$$

where the angle ω is a function related with the Dundurs' parameters α and β [4]. The energy release rate per unit new crack area can be then written in terms of the complex stress intensity factor,

$$G=\frac{c_1+c_2}{16\cosh^2\pi\varepsilon}|\mathbf{K}|^2,\quad \mathbf{K}=K_I+iK_{II},\quad \varepsilon=\frac{1}{2\pi}\ln\frac{1-\beta}{1+\beta} \tag{5}$$

where, ε is the bimaterial constant.

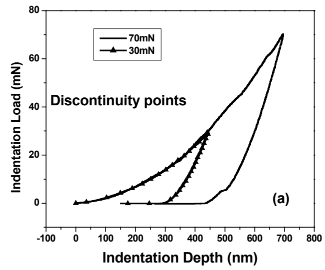

Fig.2

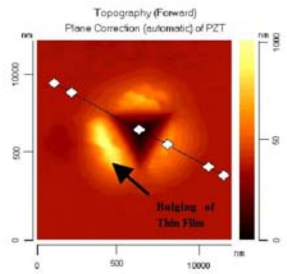

Fig.3

EXPERIMENTAL

The indenter was driven perpendicularly into, then out of, PZT thin films prepared by MOD. In order to understand indentation fracture process of PZT, we used three types of peak indentation loads to induce the fracture of the samples and recorded the corresponding load-indentation depth curves. Each indentation experiment consisted of loading, holding, and unloading completely. After the unloading, associated crack patterns were examined by AFM. The typical load-displacement curves and AFM image of indentations made at indentation loads are given as Fig.2 and Fig.3.

To determine the parameters for the theoretical model, we obtain elastic, piezoelectric and dielectric constants of PZT thin films from the reference [3]. The interfacial crack length 2a, the largest imprint 2c at the peak indentation load P_0, and the residual depth could be obtained from the AFM image. As a result the interfacial adhesion of PZT thin films could be determined from our theoretical model given as equation (5).

EXPERIMENTAL RESULTS AND DISCUSSIONS

From the load-indentation depth curves and AFM images, the different failure process can be observed. Summarizing the experimental observation, one concludes that each discontinuity indicates the point where the nano-indenter was driven from one medium into another, and the propagation of interfacial cracks increases with increasing indentation load.

After determining the radius of the residual indent mark region *c*, the interface intensity factors K_1, K_2, and energy release per unit new crack area G can be calculated by equations (4) and (5). The energy release rate versus the interfacial fracture area is shown in Fig.4, which showed that the energy release rate decreased as the interfacial crack length increased.

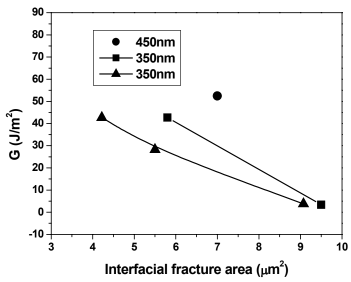

Fig.4

CONCLUDING REMARKS

We proposed a model to measure interfacial adhesion in ferroelectric thin films. An elastic groundsill beam model taking into account the piezoelectric effect is developed to describe the interfacial crack for ferroelectrics thin films when the indentation depth is close to the film thickness. The model was validated by nano-indentation tests made on PZT thin films, where the elastic modulus and hardness of PZT thin films were determined from load-indentation depth curves, and energy release rate and stress intensity factors were calculated.

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