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PZT multilayer films: Modelling polarization effects caused by microstructural inhomogeneities

J. Rödel*, W. Beckert[1], W.S. Kreher

Technische Universität Dresden, Institut für Werkstoffwissenschaft, 01062 Dresden, Germany

Abstract

PZT films on a substrate are of interest for application as a ferroelectric printing form. Owing to the difficult preparation conditions, such films contain inhomogeneities such as pores or glassy inclusions in the range 10–50 μm, which may lead to faulty printing properties. These effects are caused by a locally varying electric surface potential and depend on the size, depth and properties of the inhomogeneities. In this paper we present results of different electric field calculations, especially those for films containing pores. The model is based on a linear piezoelectric finite element analysis taking into account the variation of dielectric and remnant polarization properties. The deterioration of surface potential caused by the heterogeneities may be smoothed by an additional thin film surface layer.

Keywords: Defects; Electrical properties; Films; PZT

1. Introduction

PZT films on a substrate are of interest for application as a ferroelectric printing form.[1] As a consequence of the difficult preparation conditions, such as films contain inhomogeneities as pores or glassy inclusions in the range 10–50 μm.[2] In general, these defects can be described as non-polarizable inclusions, which are the source of inhomogeneous field distributions in the film. Inhomogeneous fields cause inhomogeneous surface properties, like locally varying electric surface potential, which may lead to faulty printing properties.[2] These effects depend on the size, depth and properties of the inhomogeneities.

The deterioration of surface potential caused by the heterogeneities may be smoothed by an additional defect-free thin film surface layer. More details about experimental investigations of multilayer systems for printing forms can be found in ref. 2. The aim of this work was to characterize the surface effects of microstructural inhomogeneities in order to understand the effects of multilayers on the polarization in the vicinity of inhomogeneities and to investigate numerically the effect of different boundary conditions at the free surface on the fields in the vicinity of defects.

In this paper we present results of different electric field calculations with particular attention paid to pores, which are generally considered the worst form of weakly polarizable inclusions. The model is based on a linear piezoelectric finite element analysis that takes into account the variation of dielectric and remnant polarization properties.

2. Finite element modeling

In order to solve the electromechanical field problem, a finite element model was developed. The problem is stated in terms of mechanical and electrical equilibrium equations, the definitions of strain (linear approximation) and electric field, the linear piezoelectric material law, and appropriate geometrical model and boundary conditions.

The field equations have been formulated in their axisymmetric form, and the problem was analyzed numerically using a flexible scripted finite element model builder and numerical solver (FlexPDE).[3] The main advantage of this software package is the flexibility in the statement

* Corresponding author. Tel.: +49-351-463-31414; fax: +49-351-463-31422.

E-mail address: roedel@tmfs.mpgfk.tu-dresden.de (J. Rödel).

[1] Present address: Fraunhofer Institut für Keramische Technologien und Sinterwerkstoffe, Winterbergstr. 28, 01277 Dresden, Germany.

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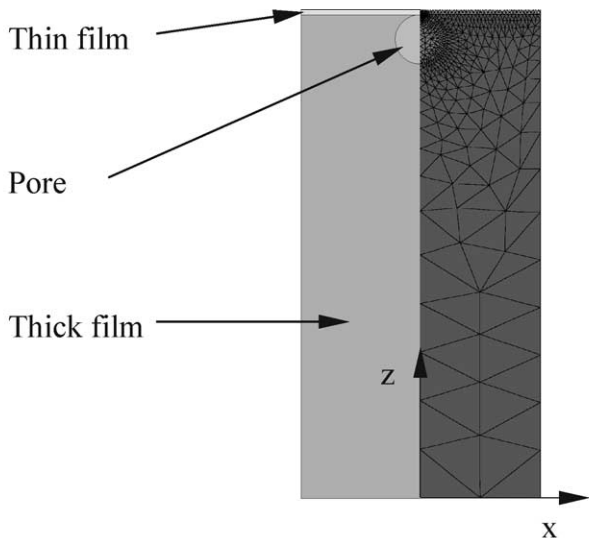

Fig. 1. Axisymmetric finite element model of a two layer system with a pore underneath the thin film.

of the field problem, which allows us to take into account piezoelectric coupling as well as remnant strain and remnant polarization.

The model geometry is shown in Fig. 1. The model consists of two layers of different piezoelectric materials. The thickness of the thick film was assumed to be 100 μm. The thin film thickness was varied between 100 nm and 15 μm. In all cases presented here, a spherical pore of 10 μm in diameter was located directly at the interface between thick and thin films.

The material properties (see Section 3) are assumed to be homogeneous within each region. As the model is not able to consider nonlinearities like ferroelastic or ferroelectric switching, the material properties and remnant polarizations are assumed to be given in advance and fixed during the calculation of fields.

In all cases, the bottom surface was set to zero potential, which is equivalent to a grounded electrode. The conditions at the inner boundary (symmetry axes) were set according to axisymmetry. As the model geometry represents a detail of a large layer system, at the outer boundary (right in Fig. 1) the radial component of the dielectric displacement (normal to the boundary) was set to zero. In order to represent the mechanical boundary conditions in a film deposited on a rigid substrate, we have fixed the displacements at the bottom surface in the radial (x) and axial (z) directions and at the left and right boundaries in the radial direction. The free upper surface was assumed to be stress free.

The electrical boundary conditions at the free surface in experiments and in the real printing process are not well known. For that reason we have analyzed the problem using different well defined boundary conditions in order to study the effects of pores and shielding of defects by defect free thin films. In Sections 4.1 and 4.2 we present results where the electric potential at the free surface was assumed to be homogeneous and set to zero. This case is equivalent to an electroded and short circuited surface. In the printing process, the polarization of the film is mainly caused by applied surface charges, so in Section 4.3 we have studied the fields under this boundary condition.

3. Material properties

The thick and thin film layers differ in their properties. In our calculations we have assumed a thick film material equivalent to a PZT-PMN ceramic material (Sonox P51, CeramTec, Lauf, Germany). For the thin film we have assumed properties of undoped morphotropic PZT 53/47.[4] The properties are summarized in Table 1. Most important are the differences in the dielectric constants, which cause electric field concentrations in the thin film. A remarkable difference in the polarization behavior between thick and thin film can be observed.[2,5] Thin films need a much higher electric field for poling, and saturation can only be achieved with difficulty.[5] For this reason, the polarization in the thin film was set to 0.1 C/m^2. As we do not have reliable data for the remnant strains of thick and thin films, these variables are set to zero. In cases where we considered unpoled material (Section 4.3), the piezoelectric constants and remnant polarization are also set to zero.

4. Results and discussion

4.1. Effects of pores

The effects of pores and other weakly polarizable inclusions can be characterized under conditions of constant surface potentials. We have assumed that the ferroelectric thick film and thin film are homogeneously poled within each region (0.15 and 0.1 C/m^2, respectively). Under these conditions inhomogeneous electric

Table 1
Material properties of poled PZT-PMN thick film and PZT 53/47 thin film[2,4,5]

Property		Thick film	Thin film
Dielectric constants	$\epsilon^{\gamma}_{33}/\epsilon_0$	1100	534
	$\epsilon^{\gamma}_{11}/\epsilon_0$	1100	307
Elastic constants (GPa)	c^E_{11}	111	134.9
	c^E_{12}	73	67.9
	c^E_{13}	79	68.1
	c^E_{33}	112	113.4
	c^E_{44}	29	22.2
	c^E_{66}	19	33.5
Piezoelectric constants (C/m^2)	e_{31}	−3.8	−3.9
	e_{33}	9.7	10.22
	e_{15}	9.6	10.54
Remnant polarization (C/m^2)	P^r_3	0.15	0.1

fields are caused by the differences of the material properties (polarizations and dielectric constants) between the two layers and between ferroelectric material and the pore. In the absence of the defect, the internal fields would be homogeneous within each region and depend on the dielectric properties and the relative thickness of the two layers only. Results of numerical field calculations have been reported in ref. 2.

In order to quantify the effect of pores in the vicinity of the free surface, we have plotted the dielectric displacement in the direction normal to the surface. Fig. 2 shows the decrease of electric field and displacement at the surface due to the presence of the defect.

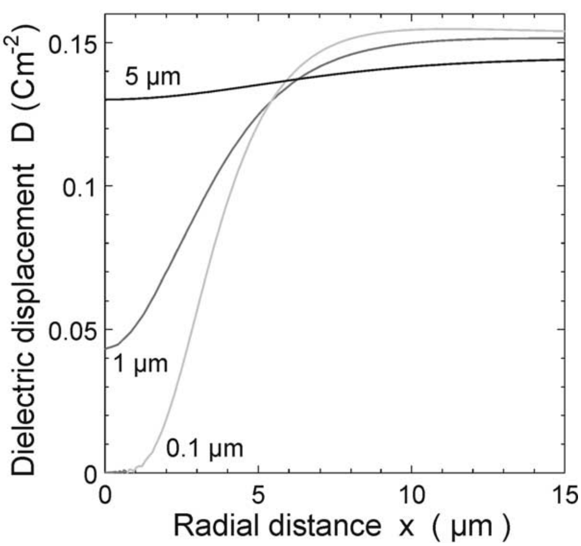

Fig. 2. Dielectric displacement at the free surface for different thin film thicknesses (0.1, 1 and 5 μm).

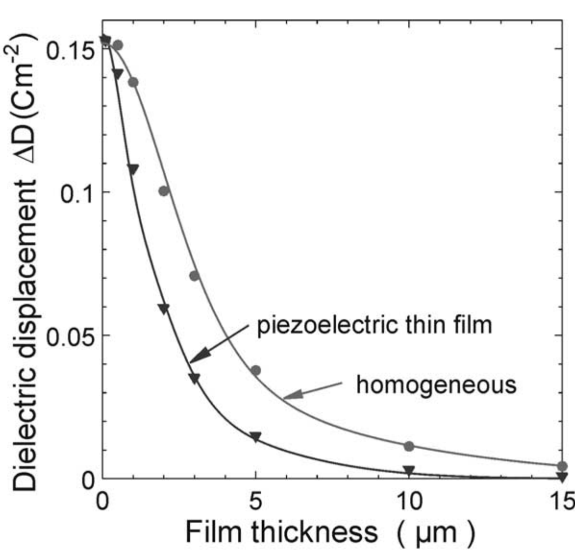

Fig. 3. Differences in the dielectric displacement at the surface as a function of the thin film thickness. Results are shown for shielding by a piezoelectric thin film compared to a homogeneous thick film material.

4.2. Effects of thin film layers

The effects of pores and other weakly or non-polarizable inhomogeneities can be reduced by defect free surface layers. The dielectric displacement on the surface for different thin film thicknesses is shown in Fig. 2. In Fig. 3 the maximum variation of the dielectric displacements is plotted as a function of the film thickness. The results are shown for shielding by a piezoelectric thin film and are compared to the case of a homogeneous thick film material. As expected, the thin film is more effective than the homogeneous material because of the lower dielectric constants. Due to this difference the electric field is concentrated in the thin film and the effects of inhomogeneities underneath the interface are decreased.

4.3. Effects of surface charges

In order to study the internal fields caused by surface charges, the model was extended by adding a (vacuum) layer with a virtual polarization of 0.15 C/m^2, which is equivalent to adding free surface charges (schematically shown in Fig. 4). Such surface charges would generate high electric fields in the ferroelectric layers. Without considering the effect of pores, these fields would exceed 20 and 10 kV/mm in the thin film and thick film, respectively, and result in poling of the films. Therefore, in the next step of the calculation, polarizations of 0.13 and 0.1 C/m^2 are assumed. As shown in Fig. 5, the remnant polarization decreases the electric fields and results in a relaxation of the potential energy. However, the internal fields in the vicinity of defects are again inhomogeneous. An appropriate measure of the surface properties for these boundary conditions is the surface potential. In our case, a difference of almost 50 V is observed at the surface of a 1 μm thin film above a 10 μm pore (Fig. 6). This inhomogeneous surface potential would cause additional effects like migration of charges, resulting in inhomogeneous charge distributions. These effects, however, have not been considered in our model yet.

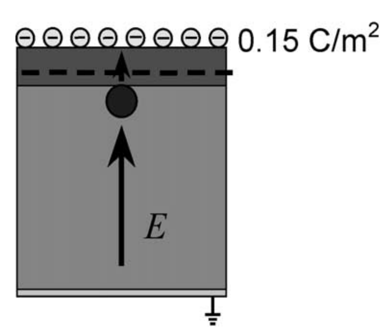

Fig. 4. Schematic figure of the field problem caused by surface charges.

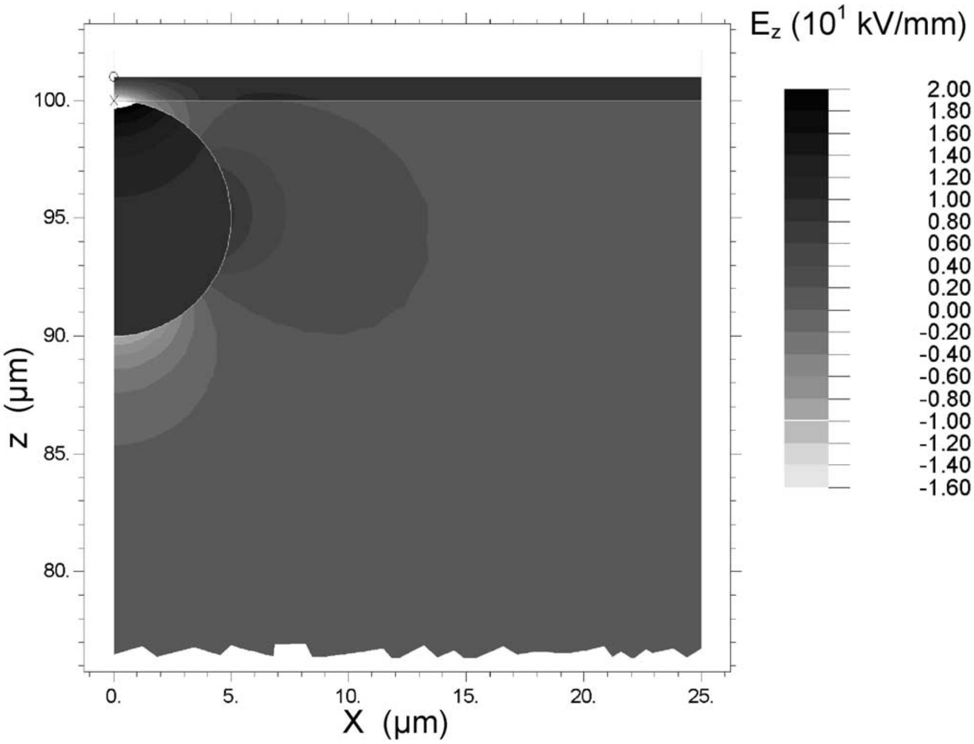

Fig. 5. Electric field in the normal (z) direction in homogeneously polarized films (see text). Fields are caused by surface charges of 0.15 C/m^2.

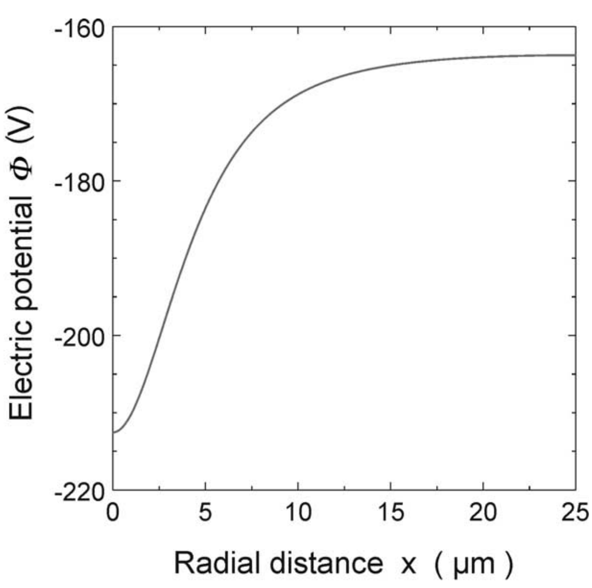

Fig. 6. Electric potential at the surface in the vicinity of a pore below the thin film. The potential is caused by constant surface charges (0.15 C/m^2) and homogeneously poled layers.

5. Conclusion

Pores, as the worst case of non-polarizable inclusions, and shielding by defect-free thin films have been studied by a linear finite element model including piezoelectricity and remnant polarization. The model provides a quantitative assessment of inhomogeneous electric fields and dielectric displacements. Numerical results of the internal fields in the vicinity of pores are given for different boundary conditions.

Our calculations revealed that a thin film thickness of the same order of magnitude as the defect size is necessary to eliminate the inhomogeneity effects at the surface.

Electric fields caused by surface charges result in an inhomogeneous surface potential. In real systems, this would cause additional migration and inhomogeneous distribution of charges and toner particles or would result in inhomogeneous polarization, which presently are not considered in our model. Thus, further development of a nonlinear model for the evolution of inhomogeneous polarization is necessary to gain more insight into the physical processes in ferroelectric films with defects.

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