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Journal of the European Ceramic Society 24 (2004) 1677–1680
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Poling and bending behavior of piezoelectric multilayers based on $Ba(Ti,Sn)O_3$ ceramics

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Abstract

Monolithic ceramic samples with up to four layers of $BaTi_{1-x}Sn_xO_3$ with different amounts of Sn were prepared. The gradient of the chemical composition has to be transformed into a gradient of piezoelectric properties by a poling process. The hysteresis loops of polarization P(E) and strain S(E) were measured and compared with calculated loops. We assumed that the polarization is constant in all layers. Based on this, the polarization of the multilayer system and the electric field strength in each layer in dependence on the applied voltage can be derived. In a first approximation, the effective strain S_3 of the system is determined by the displacement of each layer which depends on the appropriate electric field strength in this layer. The deflection at the end of the bending device (length 15 mm) was measured with an equipment based on a capacitive detector. Monolithic and glued samples with different Sn-contents of the layers were investigated. Maximum values of the bending deflection of about 0.02 μm/V were found.

Keywords: $BaTiO_3$ and titanates; Ferroelectric properties; Functionally gradient materials; Piezoelectric properties

1. Introduction

Usually, piezoelectric bending actuators are designed as an unimorph with one piezoelectric active layer or a bimorph with two piezoelectric active layers, which are mechanically joined by a glue layer.[1]

On the other hand, in the last years monolithic ceramic bending actuators were produced. These actuators are based on Functionally Gradient Materials (FGM) with a one dimensional gradient of the piezoelectric activity. Compared to conventional uni- and bimorph, FGM based bending actuators have some advantages. First, due to their relatively simple preparation they may reduce production cost. Second, it is possible to overcome the problems connected with the glued layer like peeling off or cracking. Third, the smooth gradient of piezoelectric activity can reduce internal mechanical stresses and extend the lifetime and improve the reliability of piezoelectric bending devices.[2]

FGM bending actuators are made of monolithic ceramics with a one dimensional gradient of the chemical composition. The poling process transforms the chemical gradient into a gradient of the piezoelectric coefficient d_{31}.

Here, actuators were prepared based on $BaTi_{1-x}Sn_xO_3$ (BTS) ceramics with different amount of tin ($0.075 \leqslant x \leqslant 0.15$). The piezoelectric properties have a maximum at a tin content of 7.5 mol% and decrease strongly with increasing amount of tin. Otherwise, the dielectric coefficient ε_{33} increases with increasing tin content.

2. Sample preparation

$BaTi_{1-x}Si_xO_3$ ceramics ($0.075 \leqslant x \leqslant 0.15$) were produced by classical mixed-oxide technique. The sintering was performed at 1400 °C for 1 h with a heating and cooling rate of 10 K min^{-1} to obtain coarse-grained ceramics with an average grain size about 80 μm.

Monolithic multilayer structures with a gradient of the tin content were prepared by successive pressing of the corresponding powder. They consist of two, three

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doi:10.1016/S0955-2219(03)00496-5

and four layers and are called in the following bimorph, trimorph and 4-morph, respectively. The chemical composition and configuration of the layers are shown in Table 1. The layers are named with BTSx, where x is the amount of tin in mol%.

During sintering the monolithic samples bend due to different thermal expansion coefficients of $BaTi_{1-x}Si_xO_3$ ceramic layers. In particular, a strong bending effect was obtained for the bimorph structure. Than higher the number of layers than lower the curvature of the sample. The 4-morph structure is nearly unbowed.

Additionally, two model structures were prepared for the varification of the modeling: a conventional glued bending device and a wire connected system, where the layers were connected only electrically. The wire connected system ideally corresponds to modeling assumptions, because no mechanical stress is induced by different remanent strain of the layers during the poling process. The influence of this stress can be estimated with the glued samples. The model structures were made up of ceramic sheets with the same chemical composition and configuration as in the monolithic samples. All investigated samples had the same dimensions, length L = 15 mm, thickness H = 1.2 mm and width W = 4 mm.

3. Poling behavior

Here, the basic ideas of the modeling of the poling behavior of ferroelectric multilayer structures are briefly presented. The modeling is described in Ref. 3 in detail. The aim of the modeling is to calculate the virgin P-E curve of a structure consisting of N layers with different ferroelectric properties. We assumed that the dielectric displacement D_3 is constant and the electrical conductivity is neglected. Thus, the polarization P_3 of neighboring layers is equal

$$P_3 = P_3^{(1)} = \ldots = P_3^{(i)} = \ldots = P_3^{(N)}. \tag{1}$$

The dependence of polarization on the applied voltage, respectively on the electric field strength $E_3^{(i)}$ in the layer i, strongly depends on the amount of tin. We used measured virgin loops P(E) of single ceramic sheets with a certain tin content and fitted the experimental data by two different polynomials $E_3^{(i)} = f(P_3)$ for increasing and decreasing the electric field, respectively. The modeling of both these parts is carried out separately. Thus, for example with N layers we got a system of N equations. Furthermore, we can derive the virgin loop P(E) of the multilayer system using the condition

$$U_{\mathrm{appl}} = \sum_i E_3^{(i)} h^{(i)} \tag{2}$$

The virgin loop P(E) of the layer with the lowest induced or spontanous polarization was measured up to an electric field strength of about 2 kV/mm. We assumed that the material was completely poled at this electric field and the polarization reached saturation. Because the polarization in the other layers could not be higher than in the layer with the saturation polarization, the remaining layers should be not have been completely poled. In Fig. 1, the virgin loops of the single layers and the calculated P(E) curve of the bimorph are shown.

Moreover, the electric field strength in each layer in dependence on the applied voltage can be calculated. This allowed us to derive the virgin loop of the strain S_3 parallel to the electric field. Virgin loops of single layers were measured up to the maximum electric field in this layer calculated by Eq. (2). The experimental data were fitted by polynomials $S_3^{(i)} = f(E_3^{(i)})$. In a first approximation, the effective strain S_3 of the system is determined by the displacement of each layer. Using Eq. (2) and the following condition

$$S_3 = \frac{1}{H} \sum_i S_3^{(i)} \left(E_3^{(i)} \right) h^{(i)} \tag{3}$$

we derived the dependence of the strain S_3 on the applied voltage. Here, H is the thickness of the whole system and $h^{(i)}$ the thickness of layer i. Fig. 2 illustrates the good agreement of this modeling with experimental results.

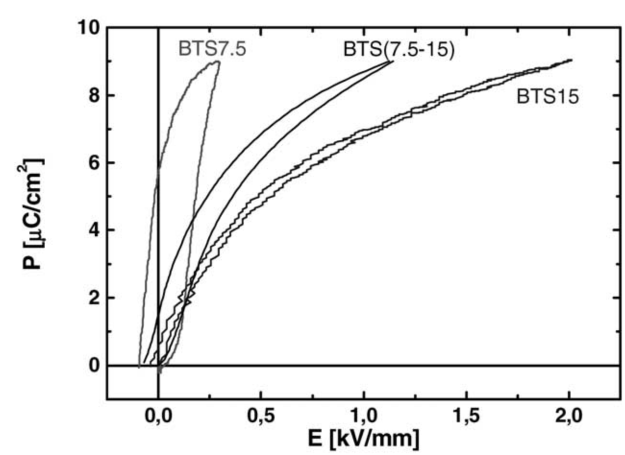

Fig. 1. Virgin loops of single ceramic layers using to fit E(P); calculated virgin loop of the corresponding bimorph (BTS7.5–BTS15).

Table 1
Configuration of the bending actuators

Bimorph	BTS7.5-BTS15
Trimorph	BTS7.5-BTS12.5-BTS15
4-morph	BTS7.5-BTS10-BTS12.5-BTS15

4. Bending behavior

4.1. Modeling

In general, the deflection of a bending device depends on the difference of the dilatation of the layers lengthwise. Both, the piezoelectric and dielectric coefficients of the layers influence the dilatation. First, the piezoelectric effect defines the strain of the layer depending on the electric field strength. On the other hand, the value of the electric field in a layer depends on the dielectric coefficient.

We assumed, that the elastic properties of the investigated BTS ceramics do not depend on the amount of tin. Consequently, the deflection δ at the end of a one side fixed actuator can be calculated by the theory of Marcus[4]

$$\delta = \frac{L^2}{2}\frac{\int_{-h/2}^{h/2} d_{31}(z)E_3(z)z\,dz}{\int_{-h/2}^{h/2} z^2\,dz}. \quad (4)$$

The electric field in the layer i depends on the applied voltage U_{appl} and the dielectric coefficients ε_{33} of all layers in the following manner

$$E_3^{(i)}(z) = \frac{U_{appl}}{h^{(i)}}\left(\varepsilon_{33}^{(i)}\sum_j^N \frac{1}{\varepsilon_{33}^{(j)}}\right)^{-1}. \quad (5)$$

4.2. Experimental results

The poling process was optimized to get best piezoelectric properties of the single layers. A DC-voltage was applied for 5 seconds. All samples were poled at room temperature. The wire connected system was used to determine the piezoelectric and dielectric coefficients of the poled layers. The measurements were done at least 24 h after poling. Then the layers were glued in order to produce an bending actuator, too. In Table 2 the data of the 4-morph system were shown. These results were used for calculation of the bending deflection by Eqs. (4), (5).

The sample was fixed at one side, the bending deflection was measured with a capacitive deflection sensor at the free end. A sinusoidal voltage of 137 Hz was used, much lower than the mechanical resonance frequency of the bending actuator. A maximum voltage of about 100 V was applied to the sample. The electric field strength can be higher in some layers, because the dielectric coefficients are quite different. In Table 2 the values of the electric field of each layer in a 4-morph system are shown, calculated for an applied voltage of about 100 V. The values has been derived by Eq. (4).

A bipolar voltage was applied to the actuator and the average value of the positive and negative maximum deflection was calculated. In Fig. 3, the maximum deflection of the wire connected poled sample is seen to increase nonlinearly with increasing voltage higher than 40 V/mm. Samples which were glued before poling and monolithic samples show a linear dependence of the > average maximum deflection on the applied voltage.

Otherwise, the deflection of all structures depends at higher voltages nonlinearly on the applied voltage. Moreover, the bending loops are asymmetrical (Fig. 4). The deflection strongly increases in the direction of the poling field. In the opposite direction the bending deflection is much smaller. This nonlinear effect is weaker for monolithic and glued samples which were already connected during poling.

Table 2
Material properties of single layers after poling, electric field strength in these layers at an applied voltage of 100 V (4-morph, wire connected poled)

Tin content mol%	d_{31} pm/V	d_{33} pm/V	$\epsilon_{33}^T\ \epsilon_0$	E V/mm
7.5	−117	188	4120	136
10.0	−50	125	4880	115
12.5	−30	50	13,170	43
15.0	−2	5	14,560	39

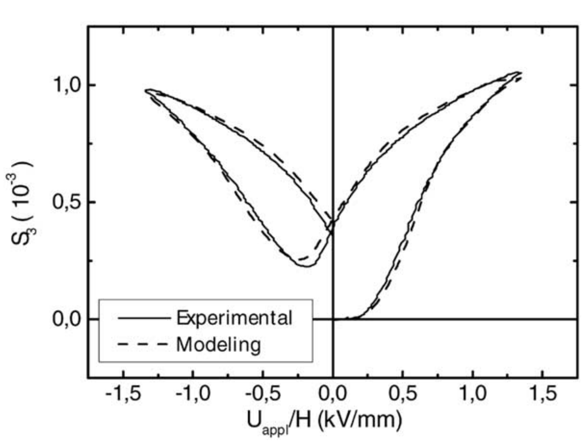

Fig. 2. Measured and modeled virgin loop $S_3(E_3)$ of a monolithic trimorph.

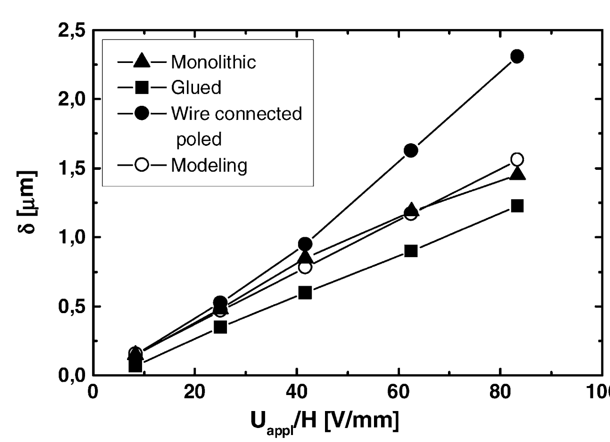

Fig. 3. Average value of the maximum deflection in dependence on an applied bipolar voltage for trimorph systems.

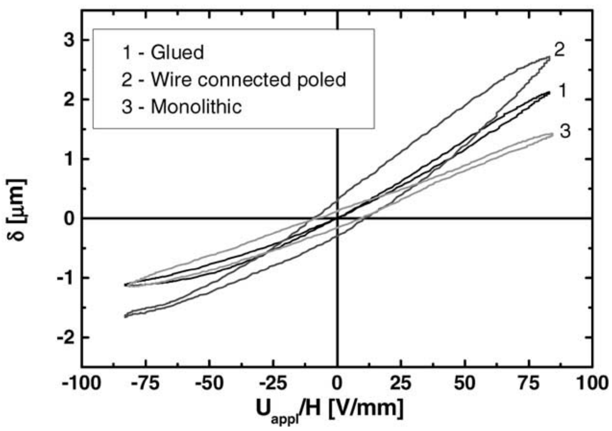

Fig. 4. Bipolar bending loops of monolithic and model actuators at an applied voltage of about 100 V.

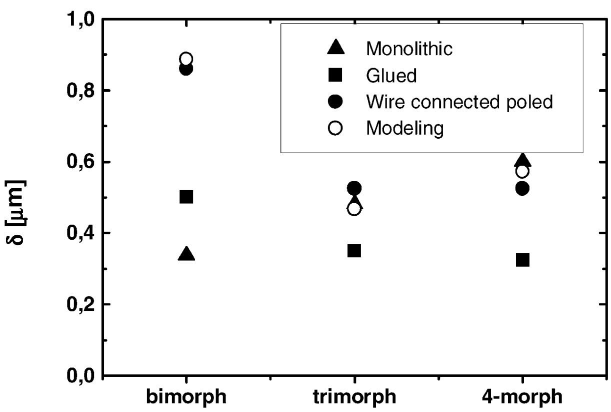

Fig. 5. Deflection of model structures and monolithic bending devices in dependence of the number of layers at an applied voltage of 30 V.

In Fig. 5, the experimental results were compared with the modeling. Only data measured at small voltages (30 V) were used, where the nonlinear effects vanished. The data of the wire connected poled sample were in the best agreement with the modeling. The bending of monolithic samples with more than two layers can also be described well with the analytical approximation. We suppose that the differences of the monolithic bimorph are associated with the strong bending of this sample by the sintering.

A lower deflection of structures was obtained, which were first glued before being poled. This may be due to mechanical stresses perpendicular to the electric field induced by the remanent strain after poling. We suggest, that the mechanical stress caused by the clamping influenced the poling degree of the layers and thereby the piezoelectric and dielectric coefficients. The monolithic samples are ceramics with a smooth transition of amount of tin between layers. The mechanical stress should be much lower in such devices based on FGM.[2] In addition, the influence of the glue while poling is not clear.

5. Summary

Monolithic $Ba(Ti,Sn)O_3$ ceramics with a gradient of the amount of tin were prepared and poled. The remanent polarization P_r after poling was slightly higher than in the model structures. We assumed that this was due to smooth transition of the amount of tin between neighboring layers in the monolithic ceramics. We found a good correspondence with the results of the modeling. Although, the maximum electric field strength in layers with higher spontaneous polarization (BTS7.5) is much lower during poling, the layers have high piezoelectric coefficients. The deflection of the bending actuators is quite linearly at small driving voltages and can be described with an analytical approximation. Higher voltages in poling direction produce an increasing of the deflection. However, the deflection of the actuator decreases at the negative electric field. In spite of minor differences in P_r between monolithic and model structures an excellent consistency of the bending properties was found. Monolithic bending devices based on FGM are not inferior to comparable glued actuators. Otherwise, the bending deflection of about 0.02 µm/V is much lower than conventional devices with middle electrodes where the layers are poled in opposite direction (0.11 µm/V).[5]

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