

# Processing of a multilayer bender type actuator

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## Abstract

A PZT material with high  $d_{31}$  was developed and the processing of a bending type multilayer actuator by stacking and co-firing platinum electroded tape cast layers was investigated. The thermocompression step, as well as the binder burnout heating profile, proved to be critical and a tempering process was introduced that prevents the microcracks formation during densification. All the processing steps were optimized and prototype actuators made of 11 layers, each layer being 50  $\mu\text{m}$  thick, were fabricated. The final microstructure was studied as well as the no-load deflection at increasing driving voltage. © 2001 Elsevier Science Ltd. All rights reserved.

**Keywords:** Actuators; Multilayers; PZT; Tape casting

## 1. Introduction

The development of multilayer actuators of the bending type, i.e. using the transversal  $d_{31}$ , is attracting increasing attention<sup>1–3</sup> as the processing technologies to reduce the layers thickness and the electrode design<sup>4</sup> are improved so as to let them work at the usual operating voltages. As the driving voltage is lowered by the reduction of the layer thickness, it has to be reduced to few tens of microns, still remaining the length and width of the order of centimetres. It implies that some steps of the process play a critical role as regards the flatness, geometrical tolerance and mechanical performance<sup>5</sup> under operation, the lower the layer number, the higher the deformation.

The aim of this work was the development of the whole process for the production of multilayer actuators at the laboratory scale, the investigation of the critical steps of the processing and a preliminary characterization of the deformation performance.

## 2. Experimental procedure

A PZT powder of the composition  $\text{Pb}(\text{Zr}_{0.54}\text{Ti}_{0.46})\text{O}_3$ , properly doped with a combination of isovalent ( $\text{Sr}^{2+}$ ),

donor ( $\text{Nb}^{5+}$ ) and acceptor ( $\text{Y}^{3+}$ ) dopants to improve the piezoelectric strain constant  $d_{31}$ , as reported in Table 1, was developed following the conventional solid state reaction of the starting oxides and carbonates, carefully mixed and calcined at 850°C for 4 h. The average particle size of the starting PZT powder was 0.9  $\mu\text{m}$ .

For the tape casting slurry formulation, an azeotropic mixture of methyl-ethyl-ketone (MEK) and ethanol (EtOH) (66:34 vol.%) was used as dispersing medium while powder deflocculation was assured by addition of glycerol trioleate (GTO). Two types of binders [polyvinylbutyral (PVB) with different molecular weights (Butvar B-98, MW 55,000 and Butvar B-76, MW 105 000)] and buthyl-benzyl-phthalate (BBP) (Santicizer-160) as plasticizer were employed. The slurry composition is reported in Table 2. The PZT powder to non volatile organic volume ratio was 1.05.

The internal electrodes were based on a conventional screen printing platinum paste.

The slurry was prepared by ball milling the powder with the dispersing medium (20 h), the deflocculant and the first binder, prolonged of further 20 h upon addition of the second binder and 4 h more with the plasticizer. After de-airing, tape casting was carried on with a laboratory bench working with a stationery doctor blade system. The blade height was changed in order to obtain green tapes with thickness in the range 70–250  $\mu\text{m}$ ; the carrier (consisting of a polyester film) speed was fixed at 300 mm/min. Drying was carried out at room temperature without any air flowing.

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Discs were cut from the tape, electrodes were deposited by screen printing the platinum paste then they were stacked and put into a die cavity heated to 75°C. An external pressure of 18 MPa was applied and held for 15 min, then a tempering treatment at 80°C for 15 min followed.<sup>6</sup>

The multilayers were heated with a controlled temperature ramp (6°C/h from room temperature to 500°C) and sintered at 1200°C for 1 h, placed on a ZrO<sub>2</sub> disk and covered with an Al<sub>2</sub>O<sub>3</sub> crucible and sealed with pack (PbZrO<sub>3</sub> added with 5 wt% ZrO<sub>2</sub>) to held a constant PbO activity at the sintering temperature. The multilayers were finished, external electrodes were deposited and fixed with a thermal treatment at 750°C for 15 min; the poling process was finally carried out into silicon oil at 120°C under a dc field of 3 kV/mm for 40 min.

The sintered microstructure of the polished perpendicular sections of the interfaces was investigated by scanning electron microscopy (SEM) coupled with an energy dispersive X-ray spectrometer (EDS). The flexural mode deformation of the actuator versus the applied voltage was determined quasi-statically with the aid of a microscope.

### 3. Results and discussion

The sintered multilayers dimensions were 26 mm long, 12 mm wide and thickness depending on the number

Table 1  
Main dielectric and piezoelectric properties of the bulk material

Coupling factors	$k_{33}$	0.711
	$k_{31}$	−0.391
	$k_p$	0.69
Piezoelectric charge coefficients (10 <sup>−12</sup> m/V)	$d_{33}$	447
	$d_{31}$	−242
Relative dielectric constants (at 100 Hz)	$\epsilon_{33}^T$	2334
	$\epsilon_{11}^T$	2926
Mechanical quality factor	$Q_m$	71
Density <sup>a</sup> (10 <sup>3</sup> kg/m <sup>3</sup> )	$\rho$	7.52

<sup>a</sup> Density of isopressed pellets (150 MPa) sintered at 1200°C for 1 h.

Table 2  
Composition of the tape casting slurry

Component	Concentration (vol. %)
PZT powder	18.9
MEK/EtOH	64.8
GTO	1.9
PVB-1 (Butvar B-98)	2.4
PVB-2 (Butvar B-76)	8.0
BBP (Santicizer-160)	4.0

and thickness of the layers, a typical value being 0.57 mm for an 11 layer sample, each layer being 50 µm thick. The sheet obtained by tape casting shows a rough upper side, while the lower surface is rather smooth and glossy so the electrodes were screen printed on the rough surface to improve their adhesion.

Thermogravimetric analysis (TGA) performed on a green multilayer with internal platinum electrode (Fig. 1) revealed that the organic additives of the screen printing paste start to burn out at 50°C while the main degradation of the binders and plasticizer occurs in the 200–300°C temperature range and is completed at 420°C. The DTA curves show three main exothermic peaks at 280, 325 and 450°C assignable to the decomposition of the organic compounds, rupture of the side and principal chains, followed by the degradation of the cyclic structures. These data stress the importance of a slow debonding from room temperature to 500°C. This was a critical step to assure a good uniformity and the conductivity to the platinum layer as a consequence of the good metal interconnection as shown in Fig. 2. The mor-

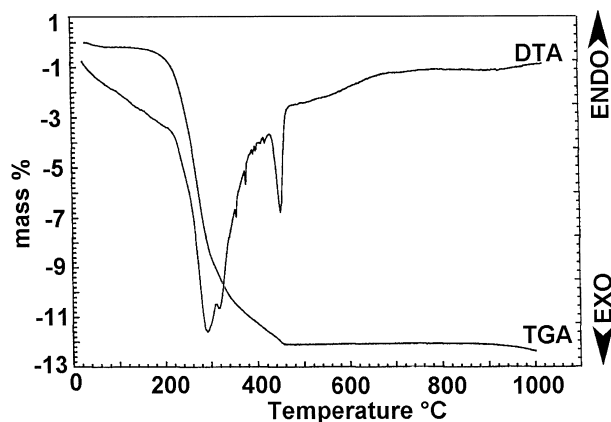


Fig. 1. DTA and TGA analysis of the green electroded multilayer.

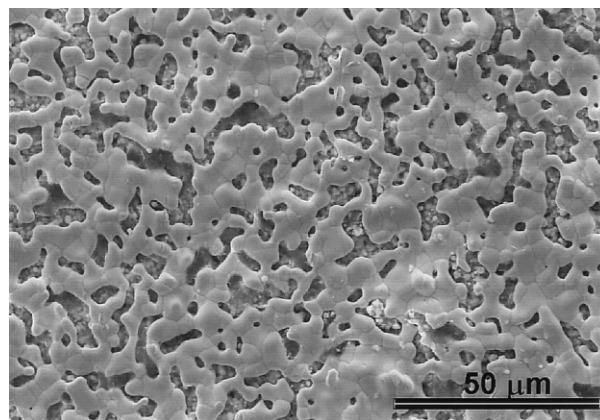


Fig. 2. SEM morphology of an external layer of platinum after debonding and sintering.

phology of the multilayer structure is shown in Fig. 3. Fig. 3(a) reveals that the internal electrodes are well defined and joined to the bulk PZT layers which thickness is about 50  $\mu\text{m}$ . The structure results homogeneous and only few small pores of the size of about 3  $\mu\text{m}$  are present. The morphology of the fracture surface shown in Fig. 3(c) evidences well faceted grains with a rather spread size distribution in the range 1–10  $\mu\text{m}$  and a prevailing intergranular fracture. The XRD diffraction pattern evi-

dences the presence of almost pure tetragonal phase. The joining of the electrode to the PZT layer, investigated by SEM and EDS analysis (Fig. 4) confirms that the electrode is well interconnected to the PZT layer [Fig. 4(a)] and no platinum diffusion takes place, as can be seen from the comparison of the analysis of the bulk near to the electrode and the electrode itself (Fig. 4(b)). In fact no platinum trace was found in the bulk PZT at 0.5  $\mu\text{m}$  distance from the electrode.

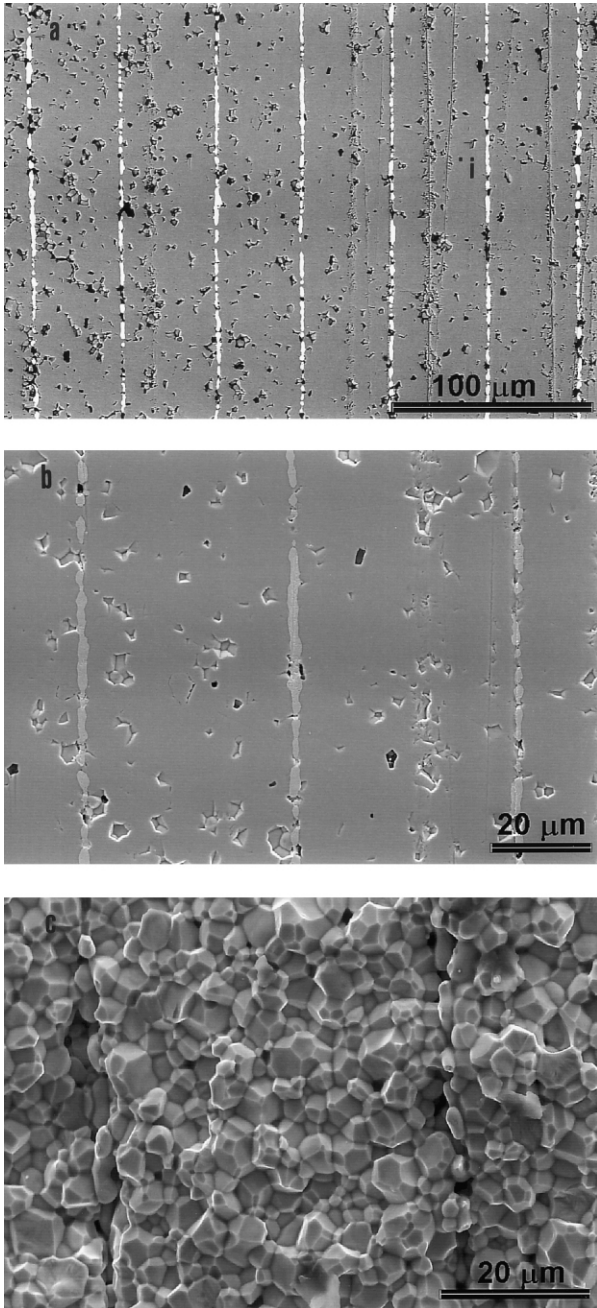


Fig. 3. SEM morphology of the cross-section of the sintered multilayer: (a) BSE image; (b) polished surface; and (c) fracture surface, at increasing magnifications.

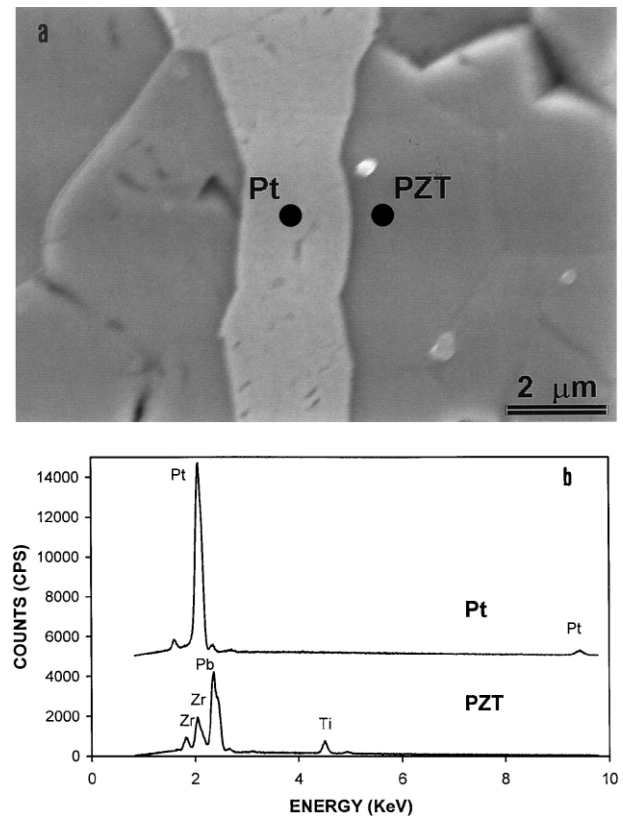


Fig. 4. SEM morphology of the internal electrode area (a) and EDS analysis (b) of the platinum electrode and of the bulk material 0.5  $\mu\text{m}$  away from the electrode.

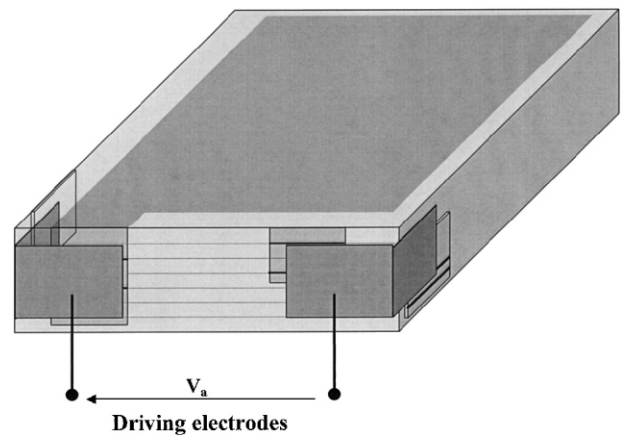


Fig. 5. Final configuration of the multilayer  $d_{31}$  bender actuator, including the external electrodes arrangement.

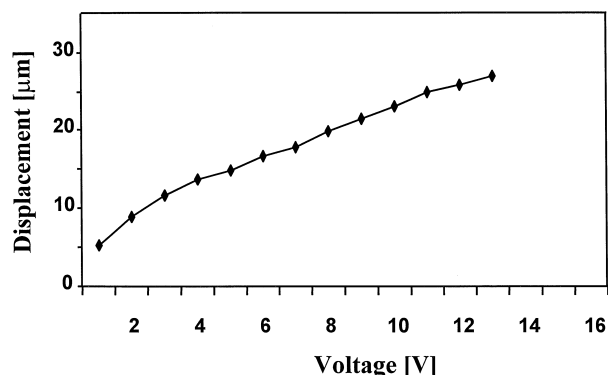


Fig. 6. No-load deflection versus applied voltage for the 11 layers actuator.

In the initial development stage, the samples resulted broken either after debonding or sintering. The introduction of a tempering step inhibited the crack growth during the sintering stage, that had resulted detrimental for the integrity of the heterogeneous multilayer architecture. In fact, the soaking of the stacked samples at a temperature slightly higher than that of the thermo-compression step promoted the recovery of the internal stresses introduced by the stacking under pressure. They came mainly from the differences in the softening of the composition of the organic additives used for the tape casting slurry and for the screen printing paste at the low temperature heating, associated to the differences in the dilatometric performance of the bulk PZT and platinum at the sintering temperature.

To investigate the relation between the displacement and the applied voltage, the 11 (50  $\mu\text{m}$  thick) layers prototype actuator was contacted with wires, clamped in a sample holder (see Fig.5) and the no-load deflection was measured optically as a function of the increasing voltage as reported in Fig. 6.

#### 4. Conclusions

The whole process for the production of a multilayer bending actuator was developed starting from sheets obtained by tape casting. It resulted into uniform layers well interconnected with the co-fired internal electrodes. The optimisation of each heating treatments resulted critical, especially the introduction of a tempering step allowed to attain good mechanical performance upon application of a low voltage field. These preliminary results confirm that all the processing steps were properly developed. To achieve improvements of the performance, the thickness of the layer will be further reduced.

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