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Hybrid powder-sol–gel PZT thick films on metallic membranes for piezoelectric applications

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Abstract

A hybrid sol–gel-powder method was used to process 3–6 μm thick lead zirconate titanate films via spin-coating a Ni-base alloy and silicon substrates. The films were crack free and possessed a fine microstructure. High effective piezoelectric coefficients, d_{33}, of 250 pm/V were obtained from piezoelectric hysteresis loops using a laser vibrometer. Crack free sensor membranes were obtained from these film heterostructures via laser beam micromachining, and their resonance behaviour was studied. Metallic membranes showed the best characteristics where deflections of up to 120 nm at resonance could be obtained at an excitation voltage amplitude of 0.5 V. Characterization of the membranes in terms of output current and voltage was also performed, and show the suitability of such heterostructures for piezoelectric sensor applications.

Keywords: Sol–gel processes; Dielectric properties; PZT; Membranes; Sensors

1. Introduction

Multifunctional materials based on the solid solution $PbTiO_3$–$PbZrO_3$ (PZT) exhibit high piezoelectric properties which make them promising candidates for many sensor and actuator applications.[1] These properties may even be amplified via using composite structures such as PZT/metal, e.g. unimorph, flex-tensional actuators[2] and Moonie actuators,[3] which have been shown to lead to high deflection amplitudes.

Modern devices require small, miniaturized structures which can be easily produced via thin film technology, and in fact the last decades have seen a plethora of work dealing with thin PZT films for micro-sensor and actuator applications.[4–7]

In the present paper a hybrid powder-sol-gel processing route[8–10] is employed in order to fabricate thick PZT films on metallic foils. New results on the piezoelectric properties of such films are presented, and perspectives for their use as resonant membranes for sensor applications are shown. Metallic substrates are of great interest for many applications including embedded capacitors[11] and sensors for automotive applications.[12] Although a recent interest has been devoted to ferroelectric thin films on metallic substrates,[11,13,14] no work has been found on the application of PZT thin film/metal heterostructures for piezoelectric applications.

2. Experimental

The thin films were processed using the route outlined in previous work.[9,10] First a PZT powder (PZ21, mean powder size 0.6 μm, Ferroperm-piezo, Denmark) was milled for 24 h and subsequently dispersed in a PZT sol with the stoichiometry $Pb_{1.1}(Zr_{0.52}, Ti_{0.48})O_3$ (PZT52/48). The substrates used are thin silicon wafers (100 μm thick) and 75 μm thick Inconel 750 (Goodfellow, Germany) foils. The substrates were first coated with four buffer layers of $(La_{0.8}, Sr_{0.2})MnO_3$ (LSMO) with a total thickness of 200 nm. The precursor solution for LSMO was prepared according to the procedure depicted in.[14] Crystallisation of LSMO was conducted at 800 °C for 20 min. The PZT films were processed by means of spin-coating. After pyrolysis and sintering for 30 min at 800 °C, the films were infiltrated with the PZT52/48 sol and annealed at 700 °C for 30 min to give dense films. The microstructure of the films was investigated by means of scanning electron microscopy (SEM) and X-ray diffraction.

Platinum top electrodes were sputtered onto the film surface to provide the electrical front contacts. For piezoelectric measurements, a circular front electrode with a diameter of

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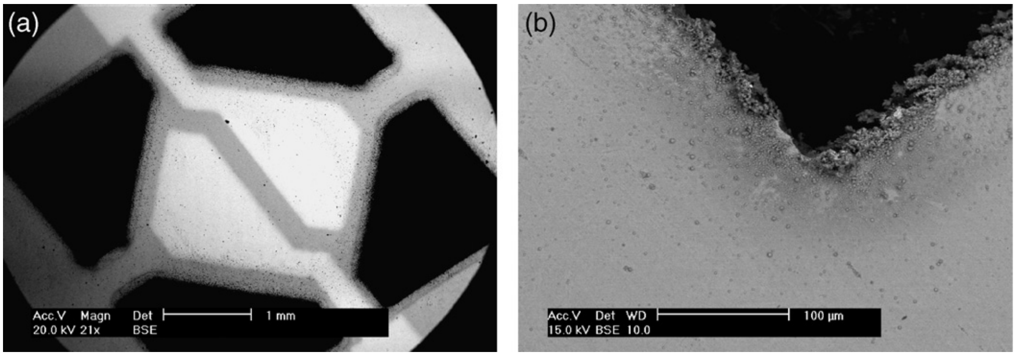

Fig. 1. SEM micrograph of the PZT/Inconel membrane at low magnification (a); (b) details of the microstructure at the laser cut edge and adjacent areas.

2 mm was used. The samples were measured in the unpoled state.

The piezoelectric measurements were conducted using a self-made experimental set-up including a computer controlled laser vibrometer (Polytec OFV 353 sensor head and OFV 3001 vibrometer controller), lock-in amplifier, frequency generator and voltage source. The vibrometer was operated in the most sensitive velocity range (1 mm/s/V). The position of the laser spot was in the centre of investigated front electrode.

Based on mechanical simulation using a commercial software (FEMLab) The membrane and electrode configurations were designed as illustrated in Fig. 1. The aim was to limit resonance to few defined modes. The membrane was then cut from the coated Inconel foil using laser micromachining under nitrogen atmosphere.

3. Experimental results and discussion

The films microstructures and phase compositions were shown in previous studies[9,15] to be pure perovskite and polycrystalline with no preferential orientation. Films deposited on different substrates show fine and very similar microstructures. The AFM root mean square surface roughness was shown to lie in the range between 50 and 60 nm.

The laser cut edges shown in Fig. 1a and b are rather rough, with a recast zone of approximately 10 μm. Molten and redeposited drops in the vicinity of the edges my also be seen. The thin films were fortunately free from cracks, and this attests to the mechanical stability of these thick film heterostructures.

The dielectric and ferroelectric properties of PZT thick films on Inconel have been reported elsewhere.[15] The dielectric constants obtained at 1 kHz are 740 for PZT on Inconel and 770 on Si. The dielectric loss, tan δ, was lower on Si with a value of 0.057 compared to 0.13 on Inconel.

The piezoelectric properties were characterized in the unpoled state. Fig. 2 shows the piezoelectric hysteresis for both silicon (a) and Inconel (b) membranes. Prior to piezoelectric characterization a frequency scan has been performed in order to locate resonances. All measurements were conducted at 3 kHz, outside the resonance regions (see below). As can be seen hystresis loops of the films on Inconel could be obtained in the DC voltage range of ±100 V, Fig. 2a. The films on Si suffered break-down at DC voltages higher than 20 V, so that saturated hysteresis loops could not be obtained, Fig. 2b.

The effective piezoelectric coefficients, $d_{33\text{eff},}$ obtained from the linear portion of the hysteresis curves are 258 pm/V and 250 pm/V for PZT on silicon and Inconel, respectively. However, the difference in the piezoelectric behaviour of both membranes is evident. While a high remnant strain is obtained on Inconel, characteristic for the shape memory effect described by Uchino,[1] the film on silicon is characterized by a slim and unsaturated hysteresis loop with a small remnant strain. The Inconel membrane shows a specific behaviour. Forward bias leads to saturation of strain at a voltage around 40 V. However, as the bias voltage

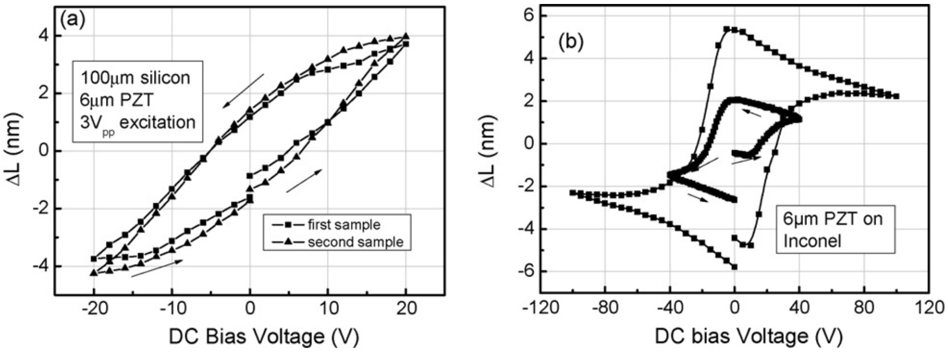

Fig. 2. Piezoelectric hysteresis loops obtained for 6 μm PZT films on a 100 μm silicon membrane (a and b) on 75 μm Inconel. The measurements were conducted at a frequency of 3 kHz and a driving voltage amplitude of 1.5 V.

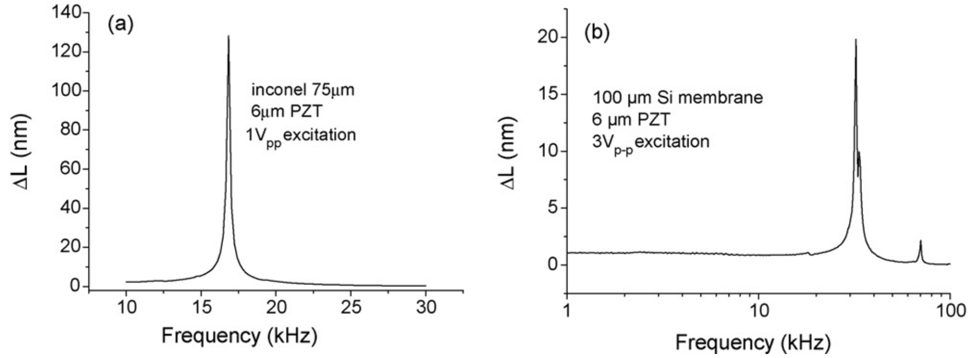

Fig. 3. (a and b) Resonance behaviour of PZT/Inconel and PZT/Si membranes showing the fundamental modes.

is reversed the strain increases and culminates at 0 V (remnant strain). Poling effects which increase the density of switched domains might account for this effect which merits to be investigated in more details.

The resonance behaviour obtained using the laser vibrometer mentioned above is depicted in Fig. 3a for the Inconel and Fig. 3b for the silicon membranes. The samples were poled merely by the piezoelectric hysteresis measurements. The highest deflection is obtained for the Inconel membrane at its fundamental frequency of 18.84 kHz. An excitation voltage amplitude of 0.5 V was enough for a deflection amplitude of approximately 130 nm. In contrast, the properties of the PZT/Si membrane were found to be quite poor.

The output voltage and current curves associated with the resonance curve of PZT/Inconel are illustrated in Fig. 4. For sensor operation both voltage and current outputs are high, and can be used without complex electronics.

The electro-mechanical behaviour of the membrane has been modelled using FEM analysis. The modelling only represents the freely moving part of the membrane. The rest of the membrane is glued on a support plate. The idealisation in the modelling is such that the ends of the suspension beams are considered clamped. The eigenmodes are determined by the mechanical properties of the substrate alone, neglecting the thin active layer. In this case for an Inconnel membrane the first resonance is at 18 kHz and the second (and third due to a degeneracy) is calculated at 38 kHz. Fig. 5 shows that the first mode corresponds to the vibration of the membrane in the z-direction with maximum deflection at the centre of the membrane. This behaviour has been experimentally observed when mapping the deflection over the membrane surface (not shown).

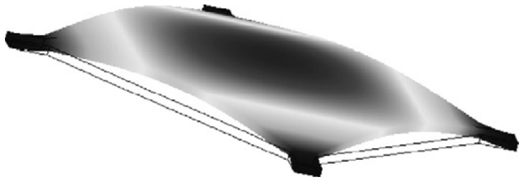

Fig. 5. Map of the deflection of the PZT/Inconel membrane for the first resonance at 18 kHz.

Finally, the structural-acoustic coupling of PZT coated membranes using the Finite Element Method (FEM) has been investigated using the software FEMLAB, which offers in its MEMS module the necessary models for simulating micromechanical structures. The modelling is intended to show the potential of such resonant membranes for pressure sensor applications.

The resonance frequency of the membrane is influenced by the density of the surrounding medium (and in consequence its pressure). In our simulation we couple a Boundary-Element-Method (BEM) of the acoustic problem to the FE model for the membrane itself. The coupling is such that the normal acceleration at a surface point is considered as an acoustic source and that the acoustic pressure acts as a force on the surface. The results show that the first resonance frequency is shifted by $-31.6\,\text{Hz}/(\text{kg/m}^3)$. Results of the simulation and experimental validation will be presented elsewhere.

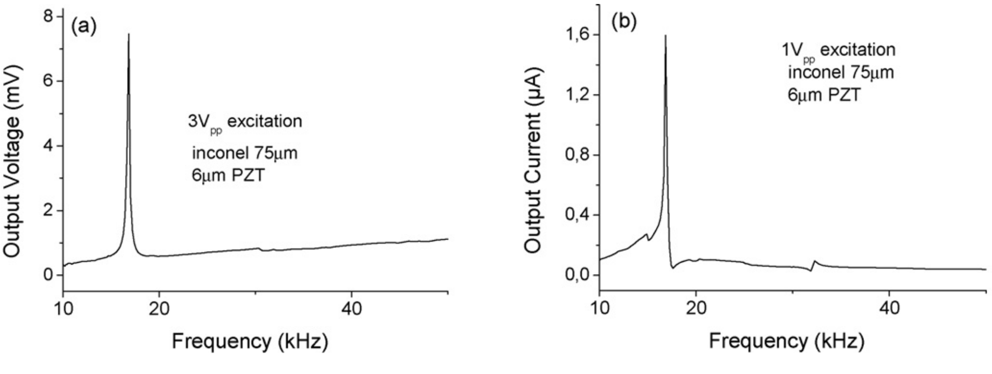

Fig. 4. Output voltage (a) and current (b) of the PZT/Inconel membrane as function of frequency.

4. Conclusion

It has been shown that crack free PZT thick films can be processed via a hybrid powder-sol-gel method on thin Inconel and Si substrates. The dielectric properties lie in the range of those obtained on platinized substrates, though the dielectric loss was found to be higher on Inconel. The piezoelectric properties are similar to those reported for flex-tensional actuators. The high remnant strains obtained resemble those of shape memory ceramics. Finally the potential of such heterostructures as resonant membranes for pressure sensor or other piezoelectric sensors is demonstrated.

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