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Fabrication and characterization of annular-array, high-frequency, ultrasonic transducers based on PZT thick film

D. Wang^{a, b, *}, E. Filoux^c, F. Levassort^d, M. Lethiecq^d, S.A. Rocks^e and R.A. Dorey^f

^aKey Laboratory for Micro/Nano Technology and System of Liaoning Province, ^bKey Laboratory for Precision and Non-traditional Machining Technology of Ministry of Education, Dalian University of Technology, Dalian, 116024, China

^cVernon S.A., 180 rue du Général Renault, 37038 Tours, France

^dUniversité François-Rabelais de Tours, GREMAN UMR7347 CNRS, 10 boulevard Tonnellé, 37032 Tours Cedex 1, France

^eCranfield University, Bedfordshire MK43 0AL, UK

^fUniversity of Surrey, Surrey, GU2 7XH, UK

Abstract

In this work, low temperature deposition of ceramics, in combination with micromachining techniques have been used to fabricate a kerfed, annular-array, high-frequency, micro ultrasonic transducer (with seven elements). This transducer was based on PZT thick film and operated in thickness mode. The 27 μm thick PZT film was fabricated using a low temperature (720 $^{\circ}\text{C}$) composite sol-gel ceramic (sol + ceramic powder) deposition technique. Chemical wet etching was used to pattern the PZT thick film to produce the annular array ultrasonic transducer with a kerf of 90 μm between rings. A 67 MHz resonance frequency in air was obtained. Pulse-echo responses were measured in water, showing that this device was able to operate in water medium. The resonance frequency and pulse-echo response have shown the frequency response presented additional resonance mode, which were due to the lateral modes induced by the small width-to-height ratios, especially for peripheral rings. A hybrid finite-difference (FD) and pseudospectral time-domain (PSTD) method (FD-PSTD) was used to simulate the acoustic field characteristics of two types of annular devices. One has no physical separation of the rings while the other has 90 μm kerf between each ring. The results show that the kerfed annular-array device has higher sensitivity than the kerfless one.

Keywords: PZT; thick film; ultrasonic transducer; annular array

1. Introduction

Ultrasound transducers are widely used today especially in the field of medical imaging systems. The growing demand of higher-resolution imaging requires desirable materials, fabrication techniques and design of structures for creating effective high frequency ultrasound transducers. Lead zirconate titanate (PZT) is a suitable material for ultrasound transducer because of its high piezoelectric constant, relative permittivity and electromechanical coupling factor [1]. High operating frequencies above 30 MHz are needed to achieve high resolution images for various applications such as ophthalmology, dermatology and intravascular imaging [2].

The thickness-mode resonant vibration frequency is inversely proportional to the thickness of the PZT structure [3]. Therefore, thicknesses of 10-100 μm are required to achieve sufficiently high frequencies and spatial resolution. Fabrication of the thick PZT film

represented a significant technical challenge using thin-film technology such as physical vapour deposition, chemical vapour deposition, sputtering or pulsed laser deposition, due to the slow deposition rates and high levels of stress generated during the process, which can lead to cracks in the film [4]. The use of conventional bulk ceramic processing with subsequent machining and bonding has a significant difficulty of producing film smaller than 100 μm , because of the high brittleness of the film at this thickness [5]. Other thick film fabrication techniques based on sintering of ceramic particles, such as screen printing, are limited by high temperature processing which can easily result in damage to the bottom electrode, substrate and PZT film itself [6]. In our previous work using a combination of conventional sol-gel processing and PZT powder processing thick films between 2 and 30 μm were fabricated using spin coating methods at lower temperatures (720°C), compatible with silicon [7].

The most commonly used ultrasound transducers are single elements [8]. However, the image field-of-view is limited when using this kind of transducers. To overcome this problem, multi-element transducers such as linear and annular arrays allow for the focal distance to be varied through delayed excitation of the array elements, which significantly increases the depth-of-field [9]. A simple type of multi-element ultrasonic transducer is a kerfless array, where there is no physical separation between elements [10]. However, crosstalk between adjacent elements can significantly degrade performances [11]. One effective way of reducing crosstalk is to mechanically isolate the individual elements [12]. The fabrication of kerfed multi-element arrays is challenging due to the dimensions of the kerfs required at high frequencies [13]. In this work a kerfed annular-array, high-frequency ultrasonic transducer with seven elements was designed and fabricated using the PZT composite slurry/sol deposition technique combined with micromaching fabrication technology. The performance of this device, including resonance behaviour and acoustic responses, was analyzed.

2. Material and methods

2.1 PZT sol and composite slurry

The PZT sol used in this work was prepared from the precursors of Lead (II) acetate trihydrate, Titanium (IV) isopropoxide, Zirconium (IV) propoxide, Niobium (V) ethoxide, Antimony (III) ethoxide, Manganese (II) acetate and 2-methoxyethanol (2-ME). The details of preparation procedures were described in our previous work [14]. The general formula of the sol used was: $\text{Pb}_{1.1}[\text{Nb}_{0.02}\text{Sb}_{0.02}\text{Mn}_{0.02}(\text{Ti}_{0.48}\text{Zr}_{0.52})_{0.94}]\text{O}_3$. The PZT thick film was formed through deposition of a composite slurry prepared from PZT sol and PZT powder. The PZT slurry was prepared by mixing PZT powder (Pz26, Ferroperm Piezoceramics, MEGGITT, Denmark), 2-ME based PZT sol, sintering aid of $\text{CuO}_2\text{-PbO}$ (4.7 wt%) and dispersant KR55 (Ken-React Lica 38, KenRich) in a nitrogen environment, then ball-milled on a roller for 24 h. In the PZT slurry the sol binds the PZT particles together and reduces the sintering temperature, due to the presence of nanoscale material, which in turn decreases lead volatilization [15]. The sintering aid enhances densification and leads to an increase in piezoelectric properties of the PZT film formed [16]. The composition of the PZT slurry is shown in **Table 1**.

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Table 1 Composition of PZT slurry

PZT slurry	Quantity
PZT sol	30 ml
PZT powder	45 g
Dispersant KR55	0.9g
Cu ₂ O/PbO	0.3105/1.926g
Zirconia balls milling media	200 g

2.2 Fabrication of the annular array

Figure 1 shows the procedures for creating the PZT thick-film, annular-array ultrasonic transducer. The 60 nm ZrO₂ diffusion barrier was formed on the silicon substrate by spin coating of a Zirconium isopropoxide sol and crystallization in a furnace, prior to the deposition of the Ti/Pt electrode and the PZT film, to prevent Pb, from the PZT layer, diffusing into the silicon wafer to form Lead silicates. Patterned bottom and top electrodes of Ti/Pt (8 nm/100 nm) were deposited using photolithographic lift off techniques with RF magnetron sputtering. During the formation of the PZT thick film, the substrate with patterned bottom electrode was spin-coated (2000 rpm for 30 s) with 2 layers of composite slurry followed by 6 layers infiltration of diluted PZT sol (50 vol% in 2-ME). This process was repeated 6 times, leading to a final film thickness of about 27 μ m.

Figure 1. Schematic illustrating the fabrication route of the PZT thick film, annular-array, ultrasonic transducer.

The PZT thick film was patterned by masking and selectively exposing the PZT to chemical etchants. The photoresist and etching solutions were AZ4562 (Clariant UK Ltd., Horsforth, Leeds, UK) and HCl (4.5 wt%)/HF (0.5 wt%)/H₂O (95 wt%), respectively. The PZT was etched in a solution of HF/HCl at 60 °C for 10 minutes. The patterned PZT thick film was then sintered at 720 °C for 20 min prior to depositing the Ti/Pt top electrode. The SiO₂ and Si materials underneath the active PZT thick film structure were removed using RF reactive ion etching (RIE, 30 minutes) and deep reactive ion etching (DRIE, 140 minutes). Each element was poled at 8.4 V/μm at 200 °C for 5 min.

2.3 Electro-acoustic characterization

The electrical impedance of each element was characterized using a HP4395 spectrum analyzer (Agilent Technologies, Inc., Santa Clara, CA, United States) to determine the resonance frequency and the electromechanical coupling coefficient (thickness mode) of the individual rings. Performances of the transducer were also evaluated through measurement of the acoustic field radiated by each element. They were excited using an AVTEC AVG-3B -C impulse generator (Avtech Electrosystems Ltd., Ottawa, Canada) with a 50 MHz impulse and the acoustic response was measured in water using a broadband needle hydrophone with a 40 μm-large active area (Precision Acoustics Ltd., Dorchester, UK).

3. Results and discussion

3.1 Structure of the annular array

The annular-array transducer was composed of 7 rings with a mechanical separation of 90 μm at the surface of the PZT film, and an outer diameter of approximately 2 mm. Each individual electrode had an area of 0.212 mm^2 to ensure electrical impedance matching. **Figure 2a** shows an overview of the 7-ring transducer with the top ground electrode faintly visible as a vertical line in the lower half of the device. One can see that some PZT ceramic remains between rings in some areas, particularly near the base of the kerfs where it is difficult for the etchant to enter and waste product to be removed. During the etching process an intermediate product of PbClF will be formed [17]. The residue of PbClF can act as a barrier that affect the etching uniformity, particularly in the narrow place such as the PZT residue near the base of kerfs observed in this work. The individual electrodes and connection lines are clearly visible from the backside of the PZT film (**Figure 2b**), where the silicon has been removed by DRIE.

Figure 2. Scanning electron micrographs showing the 7-ring annular-array transducer: (a) front view where the PZT has been etched and (b) backside where the silicon has been removed by DRIE and the individual bottom electrodes are visible

Figure 3 shows the homogeneous microstructure of the PZT thick film after etching. The PZT film exhibited slight, sub-micrometer surface cracking (**Figure 3a**), but these cracks were non-connecting such that the top electrode deposited on the PZT surface was not disrupted and all of its area remained active. The cracks also do not extend through the entire thickness of the PZT film, as can be seen in **Figure 3b**, such that no element was electrically shorted. The porosity of this PZT thick film is ~13% which was deduced from the scanning electron microscopy micrographs of film fracture cross section. The variation of the density of a material with porosity can be considered linear. The relationship between density of the PZT thick film and its porosity can be presented by the following equation [18].

$$\rho = \rho_0(1 - P) \quad (1)$$

Where ρ is the density of the PZT thick film, ρ_0 is the bulk density of PZT, and P its porosity. According to the bulk density of the materials used in this work (density of PZ26 is 7700 kg m^{-3}), the density of the PZT thick film formed in this work is $\sim 6700 \text{ kg m}^{-3}$.

Figure 3. Scanning electron micrographs showing the surface (a) and fracture cross section (b) of the PZT thick film fabricated.

3.2 Acoustic performances

Figure 4 shows the electrical impedances as a function of frequency for each element of the 7-ring array in air, measured using a spectrum analyzer. The parallel resonance frequency occurs at about 67 MHz for all elements. For the outer elements, lateral resonance modes appear and corresponding resonance frequencies are closer to the thickness-mode resonance. This increase in the energy of the lateral modes for the outer elements is due to the decrease in the width-to-height ratio of the rings. This ratio is over 20 for the inner element (disk shape and denotes as element-1), while this value is only about 2 for the outer ring (element-7). Consequently, resonant frequencies of the two modes begin to overlap for a value of this ratio below 10. The electromechanical properties in thickness mode of the element-1 were obtained through a fitting of an equivalent theoretical electrical model with the measured electrical impedance results. The theoretical behavior of

the electrical impedance was computed as a function of frequency for the thickness-mode using a one-dimensional model based on the KLM equivalent electrical circuit [19-21]. A fitting process was then applied to fit the KLM electrical model with the experimental measured impedance results. During this fitting process the thickness mode parameters of the element can be deduced [22]. Five parameters of the thick film were deduced: the longitudinal wave velocity C_L , the dielectric constant at constant strain $\epsilon_{33}^S/\epsilon_0$, the effective thickness coupling factor k_t , the mechanical loss δ_m and the electrical loss δ_e . According to these parameters, elastic constant c_{33}^E and piezoelectric coefficient e_{33} can be calculated. These parameters were summarized in **Table 2**. It was observed in Figure 4 that the coupled modes existed in the other elements. Consequently standard electromechanical characterization for the thickness mode cannot be applied and the one-dimensional KLM model cannot be used for the electromechanical characterization. Therefore, only the electromechanical properties of the element-1 were performed and obtained.

Figure 4. Complex electrical impedance curves ((a) real part, (b) imaginary part) of the annular-array transducer measured in air (number 1: inner element; number 7 : outer ring).

According to the obtained values in Table 2 the thickness-mode coupling coefficient k_t was 0.25 for the element-1. This value was lower than that of PZT bulk material (0.47) because of the lower density of the PZT thick film as deduced previously. Similar behavior was also seen in a kerfless structure indicating that it is the dimensions of the active area as defined by the electrode [23], as opposed to the physical dimension as defined by a structuring process (such as etching), that define the behavior. The highest coupling coefficient measured on the kerfless structures was 0.47 which was the same to that of the bulk material [23], indicating that the etch process may degrade the functional properties of the PZT film – possibly through opening up the porosity of the structure through selective etching of the grain boundary material.

The impulse responses of the rings were measured using a needle hydrophone. Distilled water was used as the propagation medium, showing that this device is able to operate in water. The electroacoustic response of each element was measured in water very closed to the element surface along the central axis. **Figure 5a** and **5b** show that lateral modes degrade the time-domain and frequency-domain responses of the inner element (element-1)

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and outer element (element-6). For the inner element, the spurious vibrations appear towards the end of the main oscillation, and the thickness mode remains predominant. For the outer element (element-6) whose width-to-height ratio is close to 3, the strong lateral modes interrupt the main vibration and the maximum energy is not radiated at the intended frequency.

Figure 5. Impulse responses of element-1 and ring-6 of the annular-array transducer measured in water (a) and corresponding spectra (b).

Figure 6 shows the theoretical evolution of pressure along the central axis of the transducer and in the transverse direction. The pressure was simulated using the FD-PSTD method. This modeling method was described in our previous work [24-26]. The input data for the simulations were taken from **Table 2** and assumed to be the same for the seven elements. This simulation was carried out for two types of annular arrays.

Table 2 Electromechanical properties of the inner element (element-1)

A (mm ²)	f ₀ (MHz)	k _t (%)	$\epsilon_{33}^S / \epsilon_0$	C ₃₃ ^E (GPa)	e ₃₃ (C/m ²)	δ_e (%)	δ_m (%)
0.212	67	25	990	113.4	6	2	4.3

One had no physical separation of the elements and the design was only defined by the electrode shape for each element. The other device had a 90 μ m kerf between each element. All the elements were excited with an impulse centered at 67 MHz and time delays between elements were chosen to deliver a focal distance at 4.2 mm which correspond to an f-number of 2. This value is the ration between the focal distance and the diameter of the outer element. For medical imaging this ration is often between 2 and 3. The results showed that the kerfed annular array provided higher sensitivity at the focus compared to the kerfless structure, as lateral clamping reduces the amplitude of vibration of the elements in the case of the kerfless array. Despite the higher cross-talk between elements, the kerfless array provided similar depth-of-field (0.8 mm) and lateral resolution (70 μ m) compared to the kerfed structure. However, in the case of the kerfed array, the rings were only separated by water and better performances could be expected by adding an

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attenuating material between the elements and at the back of the array. Moreover, according to these theoretical results for high frequency annular arrays, if the acoustical performances are preferred, the kerfled configuration should be retained. However, taking into account the technological aspect for the fabrication of such multi-element structure, the kerfless configuration may be an alternative to define a good trade-off between performance and fabrication.

Figure 6. Theoretical evolution of pressure along the central axis (a) and in the transverse direction (b) in water of the annular-array transducer simulated using FD-PSTD method.

4. Conclusions

The fabrication of a 7-ring, annular-array, high-frequency ultrasonic transducer using a composite sol-gel thick film processing, in combination with silicon micromachining, was demonstrated. The active material was 27 μm thick, PZT thick film, which was fabricated using an optimized composite sol-gel ceramic deposition technique. A wet etching method was used to mechanically separate the elements. The deposited PZT film presented a homogeneous microstructure and uniform thickness feature. A non-uniform etching of the PZT thick film was observed, which was mainly due to the difficulty in refreshing the etchant at the base of the kerf structures. The sub-micrometer surface cracking was also observed on the PZT film. Further optimization of the deposition and etching process were suggested to improve the uniformity and density of patterned PZT thick film. The array showed a resonant frequency of about 67 MHz in air. A FD-PSTD model showed the effect of mechanically separating the different elements of the array was to increase sensitivity while lateral resolution remained comparable to a kerfless structure.

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