A Layered SAW Device Based on ZnO/$LiTaO_3$ for Liquid Media Sensing Applications

David A. Powell*, Kourosh Kalantar-zadeh, Samuel Ippolito and Wojtek Wlodarski
Sensor Technology Group, School of Electrical and Computer Engineering
RMIT University, Melbourne, AUSTRALIA.
CRC for Microtechnology, Melbourne, AUSTRALIA.

Abstract – Surface Acoustic Wave (SAW) sensors comprising a zinc oxide guiding layer deposited on a 36°-YX lithium tantalate substrate were developed. They were found to have greater mass sensitivity than other $LiTaO_3$ based SAW sensors, such as the $SiO_2/LiTaO_3$ configuration. In this paper, the fabrication of the ZnO/$LiTaO_3$ sensor is described and micro-characterisation of the deposited films is presented. Sensitivity of these devices to surface mass and dielectric perturbations is then presented, followed by an analysis of temperature stability.

I. INTRODUCTION

90° rotated ST-quartz and 36°-YX $LitaO_3$ substrates are the most commonly utilised piezoelectric materials which allow the propagation of shear waves. As a result, they are suitable for use in liquid sensing applications. By depositing a wave guiding layer onto such substrates, mass sensitivity can be increased as the acoustic waves are more confined onto the surface.

Layered SAW sensors for liquid media applications using SiO_2 on 90° rotated ST-quartz have been intensively studied (eg. Kovacs et al [1] and Du et al [2]). SiO_2 on 36° YX-$LiTaO_3$ layered structure is another SAW structure that offers potential for liquid sensing applications [3, 4]. However, such devices have relatively lower sensitivity than devices based on ST cut quartz.

Adding a piezoelectric guiding layer generally increases the mass sensitivity. It has been shown that replacing the SiO_2 layer with ZnO, which is a piezoelectric material, results in considerable improvements in mass sensitivity [5]. In this paper, a ZnO/36° YX-$LiTaO_3$ structure is introduced for sensing applications.

Shoji et al. [6, 7] showed that deposition of ZnO layer on $LiNbO_3$ or $LiTaO_3$ results in an improved electromechanical coupling coefficient (K^2), whilst Lim and Shindo [8] studied the defects in ZnO deposited on $LiTaO_3$ by electron cyclotron resonance-assisted molecular-beam epitaxy (ECR-assisted MBE). In this paper, it will be shown that the deposition of ZnO on $LiTaO_3$ can be used in the fabrication of a liquid media sensor with improved mass sensitivity.

II. SENSOR FABRICATION

The sensor consisted of a two-port resonator with 38 input and output Inter Digital Transducer (IDT) finger pairs, 160 reflectors on each side, 700μm aperture width and a periodicity of 40μm. A 2-port resonator structure was chosen over a delay line as its higher phase slope increases oscillation stability. The IDTs and reflectors were formed by patterning a 300nm Au layer. The Au layer was deposited upon 300nm Ni/300nm Ti for improved adhesion to the substrate. Devices were fabricated with ZnO layer thickness of 1.5μm and 2.6μm, deposited by an RF magnetron sputterer. Sputtering gas was 40% O_2 in Ar at 1×10^{-2} Torr, substrate temperature was 260°C, which resulted in a deposition rate of ~0.4μm/hour. For comparison, transducers with 3μm SiO_2 were also fabricated.

III. MATERIAL ANALYSIS

ZnO films were micro-characterised using Scanning Electron Microscopy (SEM) and X-Ray Diffraction (XRD). Figure 1 shows the SEM micrograph of the film at the edge of an IDT finger. It can be seen that grain structure is quite different on the metallised and non-metallised regions.

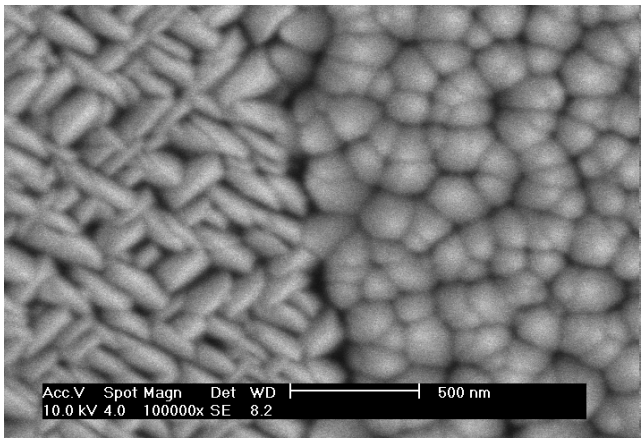

Figure 1: Micrograph ZnO on $LiTaO_3$ (left) and Au (right)

It has previously been reported that ZnO deposited by ECR-assisted MBE grows on $LiTaO_3$ with c-axis alignment parallel to the surface [6]. To confirm that the same relative orientation of thin film and substrate had been achieved with deposition conditions described in section II, a sample with no metal layer was subjected to XRD analysis. The results are presented in Figure 2. The strong (110) peak indicates that the c-axis lies in the plane of the substrate. A smaller (102) peak is also present, but is dominated by the (110) peak. Diffraction angles for crystal orientations have been taken from [6], [10], [11].

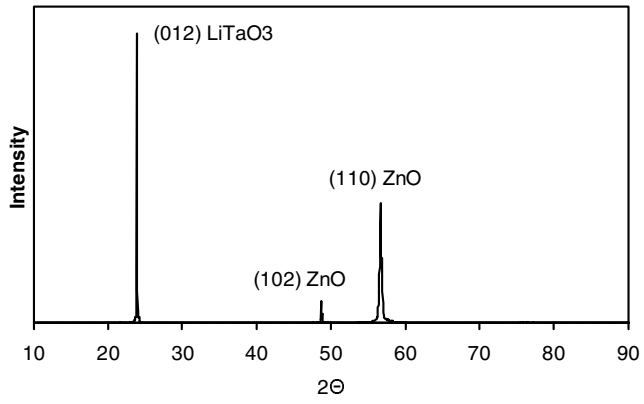

Figure 2: XRD pattern of ZnO on LiTaO3

The orientation of ZnO on Au under similar sputtering conditions has previously been reported by the authors [2] as showing a strong (002) peak, and a grain formation similar to that on the right side of Figure 1. This is indicative of strong surface-normal c-axis alignment.

IV. Mass Sensitivity Analysis

To compare the mass sensitivity of the devices with SiO_2 and ZnO layers, a 0.4μm layer of the polymer APC (Poly(Bisphenol A carbonate)) was spin coated onto the surface of each SAW device. The frequency shift was measured on a network analyser, and taken as the change at the point of maximum phase slope. From [12], the change in velocity due to mass loading is known to be

$$\frac{\Delta V}{V} = \frac{Vhv_2^2}{4P}\left(\rho - \frac{c_{44}}{V^2}\right), \qquad (1)$$

where V is the SAW phase velocity, h is the thickness of the perturbing layer, ρ and c_{44} are density and shear stiffness of the film respectively, P is the power flow per unit width, and v_2 is the particle velocity of the horizontally polarised shear wave. This equation was derived from perturbation theory, using the weak coupling assumption. This is invalid for a strong piezoelectric material such as $LiTaO_3$. However, it is assumed here that the qualitative results are still valid.

Table I: Frequency Response to Polymer Layer

Device	Frequency Shift
1.5μm ZnO	250kHz
2.6μm ZnO	500kHz
3μm SiO_2	300kHz

Table I shows that a 2.6um ZnO device is approximately 1.6 times as sensitive as a 3μm SiO_2 device. For comparison, the results of Hermann et al. [3] indicate a sensitivity of $0.5\times10^{-8}m^2s/kg/Hz$ for a 36° YX-$LiTaO_3$ device with periodicity of 40μm and a 3μm SiO_2 layer. It was also noted that further sensitivity improvement could be gained by increasing the thickness of the SiO_2 layer, however further research needs to be undertaken to determine if this is also the case for a ZnO layer.

V. Liquid Sensitivity Analysis

SAW devices show frequency shift when exposed to different liquids. This is usually due to a combination of viscosity, conductivity and permittivity effects upon the wave propagation.

To assess the response of the SAW devices in liquid media, they were placed within a 6µl flow cell. To obtain maximum sensitivity, and to minimise the effects of variable gasket pressure or leakage, the flow cell covered the entire structure, including IDTs and reflectors. This has the disadvantage that input and output impedance will be affected, which has the potential to prevent oscillation [13].

A pump with a flow rate of 250µL/min supplied DI-water to the surface of the sensor. The sample was injected into a 500µL sample loop and switched into the flow path. Figure 3 shows the response of the three devices when exposed to methanol.

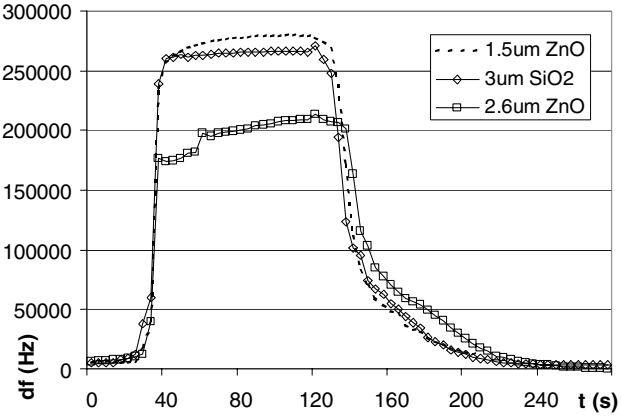

Figure 3: Responses to Methanol

The response is due to the lower dielectric constant and viscosity of methanol compared to water (see Table II). The perturbation relations [12] for velocity shift due to the electrical and mechanical properties of a liquid are respectively

$$\frac{\Delta V}{V} = \frac{K^2}{2}\frac{(\sigma'/\omega)^2 + \varepsilon_0(\varepsilon_r' - \varepsilon_r)(\varepsilon_r'\varepsilon_0 + \varepsilon_P^T)}{(\sigma'/\omega)^2 + (\varepsilon_r'\varepsilon_0 + \varepsilon_P^T)^2} \quad (2)$$

$$\frac{\Delta V}{V} = \frac{V{v_2}^2}{4\omega P}\left(\sqrt{\frac{\omega\eta'\rho_l'}{2}} + \sqrt{\frac{\omega\eta\rho_l}{2}}\right), \quad (3)$$

where σ is the conductivity, ε_r is the relative permittivity, η is the viscosity and ρ_l is the density of the liquid. Quantities denoted by $'$ indicate the perturbing liquid (methanol), while others indicate the water. It is most likely that the dominant effect is the change in dielectric constant, since this sensitivity is proportional to K^2. It has been shown that K^2 is higher for the 1.5µm ZnO device than the 2.6µm ZnO device [7].

Table II: Properties of Methanol and Water

	ε_r	η (mPa s)
Water	80	1
Methanol	33	0.58

This shows that with an appropriate choice of ZnO layer thickness, it is possible to increase the mass sensitivity, whilst reducing cross-sensitivity due to dielectric constant and viscosity change.

VI. TEMPERATURE COEFFICIENT

For dispersive SAW devices, including any with layered structure or reflectors, the magnitude of the temperature coefficient of delay (TCD) and temperature coefficient of frequency (TCF) are not identical. Instead, the relationship is [8]

$$TCF = -\frac{v_g}{V}TCD, \quad (4)$$

where v_g is the group velocity of the acoustic wave.

To measure TCF, the sensors were placed within an environmental chamber, with the remainder of the oscillator system being kept outside. Figure 4 shows a typical plot of fractional frequency deviation versus temperature for a device with 2.6µm ZnO layer. It can be seen that it is highly linear over a broad range of frequencies, and gives a TCF of –40ppm/K. It was observed that a change in cable lengths between the SAW and amplifier, or an adjustment of the phase shift network, which alters the oscillation frequency, could have a strong impact upon the TCF. Variation was observed over repeated measurement. Since the SAW devices were not hermetically sealed, it is possible that humidity variation caused some additional error.

Table III: TCF of SAW devices

Device	TCF (ppm/K)	Variation (ppm/K)
1.5µm ZnO	-22	±6
2.6µm ZnO	-40	±4
3µm SiO_2	-25	±4

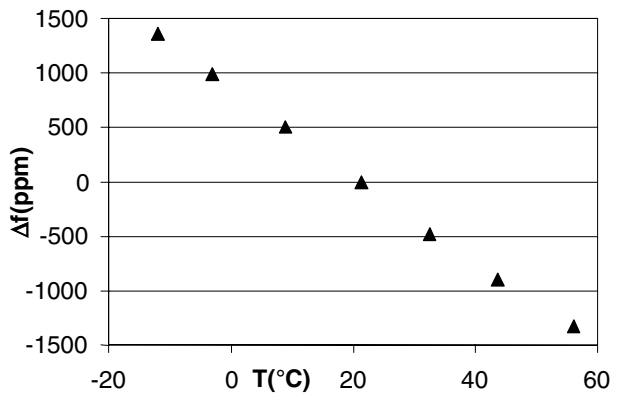

Figure 4: Frequency deviation vs.temperature for 2.6μm ZnO device.

It can be seen that the TCF of the devices with ZnO layer is inferior to that of the SiO_2 devices. This is due to the opposite signs of the TCD of SiO_2 and 36° YX-$LiTaO_3$ having a cancelling effect.

VII. Conclusion and Future Work

Mass sensitivity of the 36° YX-$LiTaO_3$ devices with 1.5μm ZnO, 2.6μm ZnO and 3μm SiO_2 layers were compared. It was shown that mass sensitivity of the 2.6μm ZnO device is 1.6 times that of the 3μm SiO_2 device, however it suffers from a higher temperature sensitivity. The response to a change in liquid on the surface was measured, and it was found that the 1.5μm ZnO and 3μm SiO_2 devices had a similar response, which was about 1.3 times that of the 2.4μm ZnO device.

For liquid sensing application, it is proposed to use an SiO_2 or polymer protective layer to ensure that ZnO does not react with any acidic materials within the liquid. Work will be carried out by the authors to model the behaviour of sensors based on ZnO/$LiTaO_3$, and to apply them to chemical and biochemical sensing applications.

VIII. References

[1] G. Kovacs, et al., "Love Waves for (Bio)chemical Sensing in Liquids", in *Proc. IEEE Ultrason. Symp.*, 1992, pp. 281-285.

[2] J. Du, G.L. Harding, A. F. Collings, P.R. Dencher, "An Experimental Study of Love-wave Acoustic Sensors Operating in Liquids", *Sens. Actuators A*, vol. 60, pp. 54-61, 1997.

[3] F. Herrmann, M. Weihnacht, "Properties of Sensors Based on Shear-Horizontal Surface Acoustic Waves in $LiTaO_3/SiO_2$ and Quartz/SiO_2 Structures", *IEEE Trans. on Ultrason., Ferroelec., and Freq. Contr.*, vol. 48, no. 1, pp. 268-273, 2001.

[4] J. Freudenberg, M. von Schickfus, S. Hunklinger, "A SAW Immunosensor for Operation in Liquid Using a SiO_2 Protective Layer", *Sens. Actuators B*, vol. 76, pp. 147-151, 2001.

[5] K. Kalantar-zadeh, A. Trinchi, W. Wlodarski and A. Holland, "A Novel Love-mode Device Based on a ZnO/ST-cut Quartz Crystal Structure for Sensing Applications", *Sens. Actuators A*, vol. 100, pp. 135-143, 2002.

[6] K. Nakamura, T. Shoji and H.B. Kang, "Growth of ZnO Films on (012)$LitaO_3$ by ECR-MBE and Determination of Their Polarity" in *Proc. Int. Symp. App. Ferroelec.*, pp. 467-470, 2000.

[7] T. Shoji, K. Nakamura and D. Yamazaki, "Propagation Characteristics of the SH-SAW on (110)ZnO/(012)$LiTaO_3$" in *Proc. IEEE Ultrason. Symp.*, 2001, pp. 215-219.

[8] S.-H. Lim and A. Shindo, "Defects and Interfaces in an Epitaxial ZnO/$LiTaO_3$ Heterostructure", *Journ. of App. Phys.*, vol. 88, no 9, pp. 5107-5112, 2000.

[9] B. Jakoby and M. J. Vellekoop, "Reducing the Temperature Sensitivity of Love-Wave Sensors" in *Proc. IEEE Ultrason. Symp.*, 1998, pp. 447-450.

[10] Y. Shibata, N. Kuze and M. Matsui, "Piezoelectric $LiNbO_3$ and $LiTaO_3$ Films for SAW Device Applications" in *Proc. IEEE Ultrason. Symp.*, 1996, pp. 247-254.

[11] X. T. Zhang et al, "Temperature Dependence of Excitonic Luminescence from Nanocrystalline ZnO films", *Journal of Luminescence*, vol. 99, pp. 149-154, 2002.

[12] J. Kondoh, K. Saito, S. Shiokawa and H. Suzuki, "Simultaneous Measurements of Liquid Properties Using Multichannel Shear Horizontal Surface Acoustic Wave Microsensor", Jpn. J. Appl. Phys., vol. 35, Part 1, No 5B, pp. 3093-3096, 1996.

[13] B. Jakoby and M. J. Vellekoop, "Analysis and Optimization of Love Wave Liquid Sensors", *IEEE Trans. on Ultrason., Ferroelec., and Freq. Contr.*, vol. 45, no. 5, pp. 1293-1302, 1998.

* Corresponding author: david.powell@ieee.org