A lithium-tantalate based acoustic reflective delay line for chemical sensing in water

J.-M Friedt, D. Rabus, L. Arapan, V. Luzet, F. Chérioux
FEMTO-ST
Besançon, France
jmfriedt@femto-st.fr

N. Nief, S. Dehez, J.R Ordonez-Varela
TotalEnergies
Pau, France
nathalie.nief@totalenergies.com

***Abstract*—Surface acoustic delay lines patterned on lithium tantalate piezoelectric substrate propagating a guided shear wave is demonstrated for direct detection chemical sensing in aqueous solutions with no need for additional microfluidic handling. The high dielectric permittivity substrate prevents excessive losses from capacitive coupling with water when dipping the whole chip, including interdigitated electrodes, in water, while the shear wave only couples with the first few hundred nanometers of the adsorbed layer. A measurement of non-specific protein detection demonstrates the principle.**

***Index Terms*—lithium tantalate, reflective delay line**

I. INTRODUCTION

Thin film-based direct chemical sensing using Surface Acoustic Wave (SAW) transducers has announced promising improved sensitivity over the bulk acoustic wave quartz crystal resonator. Nevertheless, detection limits remain improved in the latter over the former measurement in liquid phase: indeed, the quartz crystal resonator confines energy at the center of the electrodes, allowing an O-ring seal to confine liquid over the sensing area where acoustic energy is maximum and hardly affected by fluidic handling elements, as opposed to the surface acoustic wave geometry that requires confining the liquid over the acoustic cavity between interdigitated transducers (IDTs) or transducer and mirrors, with a significant impact of the fluidic structures over the acoustic path. The tradeoff between efficient sealing requiring a strong bonding of the fluidic handling structure and the weak interaction with the propagating acoustic wave leads to unstable and hardly reproducible ad-hoc solutions [1]–[3].

As long as low permittivity substrates such as quartz are considered, the capacitive short circuit of the electric field in the high permittivity water with a high capacitance favoring currents in the liquid over the piezoelectric substrate will necessarily induce strong insertion losses as long as water is not prevented from reaching the IDTs. In this abstract, we consider the use of lithium tantalate (LTO) for propagating a pure shear wave with low coupling with the surrounding liquid, generated on a high permittivity substrate inducing little losses when the sensor surface is coated with water. We demonstrate sensing capability in an ionic aqueous medium without the need for fluidic confinement.

II. LITHIUM TANTALATE SHEAR WAVE GRAVIMETRIC SENSITIVITY

Quartz is most commonly used for SAW sensors for its ability to propagate a pure shear wave confined to the substrate surface by a guiding layer in a Love mode configuration. However the low electromechanical coupling of quartz makes it poorly suited for transmission or reflective delay line due to the high insertion losses or low received power degrading the signal to noise ratio and hence the detection limit, despite a turnover at room temperature when appropriate crystalline orientations (ST, AT cuts) are selected depending on the temperature coefficient of the guiding layer. Maximum sensitivity is reached when the gradient of the acoustic velocity is maximum as a function of guiding layer thickness, under the condition of limited insertion losses, as assessed experimentally in Fig. 1.

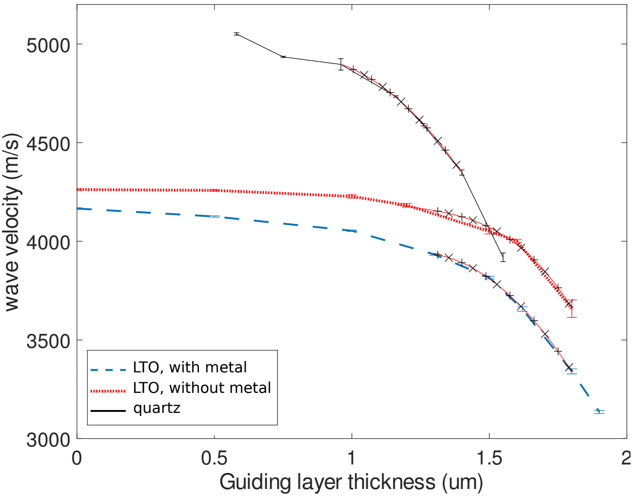

Fig. 1. Acoustic velocity for Love mode propagating on LTO and quartz as a function of polymer Shipley photoresist S18XX guiding layer thickness, with or without metallization on the acoustic path.

Improving the signal to noise ratio with the selection of a strongly coupled substrate is beneficial, especially in a reflective delay line configuration for wireless measurement. Furthermore, a high permittivity substrate will prevent capacitive coupling of the electric field with the high permittivity ($\varepsilon_r = 80$) water by keeping the electric field in the piezoelectric substrate. While lithium niobate would be a candidate with its high permittivity ($\varepsilon_r = 20$ to 80 depending on

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the direction), it is not known to propagate a pure shear wave other than the longitudinal wave with poor gravimetric sensitivity. The selected substrate is hence LTO, with a relative permittivity of $\varepsilon_r = 40$ to 50 depending on the direction but a pseudo-shear wave readily converted to a Love mode with a guiding layer or maintained at close proximity of the substrate surface with a metalization layer slowing the wave velocity. Indeed capacitive coupling, even in the presence of insulating guiding layer preventing resistive coupling to the conducting liquid, between the ε_1 substrate and ε_2 liquid leads to a voltage divider bridge between the capacitors of common area and distance between electrodes so that the division ratio is $\varepsilon_1/(\varepsilon_1+\varepsilon_2)$. When IDTs patterned on quartz with $\varepsilon_1 \simeq 5$ are exposed from air ($\varepsilon_2 \simeq 1$) to water, the coupling to the piezoelectric substrate drops from $5/6$ to $5/85$ or 14-fold, i.e. 23 dB. On the opposite when IDTs on LTO with $\varepsilon_1 \simeq 40$, the same procedure leads to a voltage drop of a factor $(40/41)/(40/120) \simeq 3$ or 9 dB. These quantities match experimental observations.

III. DELAY LINE DESIGN AND REALIZATION

A transmission delay line has been fabricated on LTO exhibiting an insertion loss of 11 dB with the optimized guiding layer thickness confining a Love mode on the subtrate surface, a favorable outcome with respect to a quartz delay line typically exhibiting insertion losses in the 18 dB range (Fig. 2) but dramatically increasing when exposed to water. Selecting an arbitrary threshold on insertion losses of 25 dB for high signal to noise ratio measurement, a quartz substrate is unable to meet the requirement when exposed to water whereas LTO remains at a level of $11 + 9 = 20$ dB insertion losses when dipped in water.

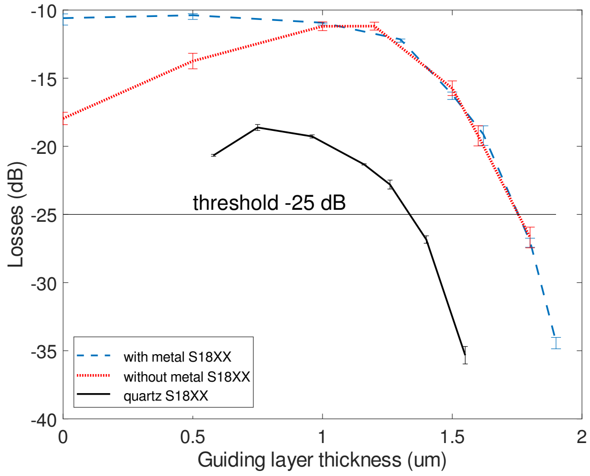

Fig. 2. Acoustic losses of quartz and LTO devices, the latter coated with a Shipley S18XX polymer guiding layer propagating a Love mode either on metalized areas between IDT and mirror or without metalization.

Most interestingly for wireless sensing applications, the transmission delay line has been extended to a reflective delay line architecture, replacing the second IDT with Bragg mirrors with lengths tuned to allow for two mirrors on both sides of the transducer, hence leading to a differential measurement on a single device when propagation delays introduced by the mirrors are different. Indeed with local chemical functionalization between each mirror, the contribution of sensor range to the interrogation (RADAR) unit, temperature and interfering chemical compound can be subtracted by differential analysis.

IV. EXPERIMENTAL RESULTS

As a demonstration of the usability of the proposed device, a LTO reflective delay line with no specific surface functionalizion has been exposed to proteins from dehydrated milk in deionized water. The objective of the experiment is to assess the ability to recover the phase from multiple echoes of the reflective delay line under non-specific physisorption of immunoglobins as would be done classically for saturating the free areas of a functionalized biosensor surface. Fig. 3

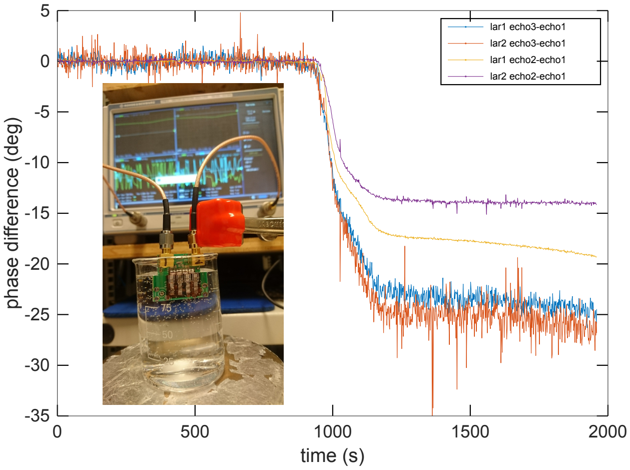

Fig. 3. Response of the four echoes patterned on a reflective SAW delay line upon exposure to a high concentration of immunoglobins from dehydrated milk in deionized water injected a 1000 s. Inset: picture of the experiment, emphasizing the lack of dedicated microfluidic handling elements on the acoustic path.

The various echoes i at delays τ_i ranging from 0.8 to 3.2 μs exhibit a phase shift φ_i proportional to the adsorbed mass and their respective absolute delay since $\varphi_i = 2\pi f \tau_i$ with $f = 200$ MHz the center frequency of the IDTs, and $\tau_i = d_i/c$ with d_i the two-way trip from IDT to echo length and c the acoustic velocity, varying upon exposure to the analyte to be detected.

V. CONCLUSION

Transmission and reflective delay lines on lithium tantalate have been designed to overcome the challenges of capacitive coupling of interdigitated electrodes exposed to high permittivity water.

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