Losses in a surface wave sensor in contact with electrolyte solutions

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Abstract— Surface acoustic wave (SAW) sensors are under development to characterize aqueous solutions for wellbore integrity monitoring. By using a 36° rotated Y-cut X-propagating LiTaO₃ substrate, emission of wave energy into the liquid is minimized. The surface wave velocity is influenced through the complex permittivity of the liquid. This permittivity is changed from that of pure water due to ionic conductivity and ion influence on the water dipole orientation. The surface wave also experiences attenuation which is dependent upon the chemistry of the aqueous phase in contact with the SAW device. In this paper, we calculate velocity change and attenuation for two aqueous solutions of interest: saline solutions and HCl solutions.

Keywords—surface acoustic wave, sensor, aqueous

I. INTRODUCTION

Wellbore fluids consist of a wide variety of chemicals including oil, gas, water, and dissolved hydrogen sulfide and carbon dioxide. Monitoring and characterizing the fluid composition may alarm potential failure of the integrity such as corrosion tendencies and leaks. We are exploring a range of sensor designs for the wellbore environment [1].

Both bulk and surface-wave sensors have been developed for sensing the properties of liquids in direct contact with the piezoelectric substrate. In order to obtain acceptable quality factor (in the case of bulk wave or resonant surface wave sensors) or acceptable attenuation (surface wave sensors) it is necessary to choose piezoelectric substrate orientations that do not significantly excite waves in the surrounding fluid. For example, a surface wave sensor for methanol-water solutions has been described [2]. In that work, a 36° rotated Y-cut Xpropagating LiTaO₃ substrate was used.

In this paper, we calculate surface wave attenuation for this substrate when exposed to aqueous solutions with a significant ionic concentration. While there is minimal acoustic wave emission into the liquid, the electric fields protruding into the liquid interact with the liquid causing dielectric losses. These losses depend in a complex way on the nature of the liquid. We will use experimental measurements of the dielectric properties of specific liquids to calculate the attenuation and surface wave velocity change.

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II. DIELECTRIC LOSSES IN AQUEOUS SOLUTIONS

Water is a dipolar molecule. Orientation of the dipoles in an electric field causes the relatively large relative static dielectric constant at room temperature ($\varepsilon_s = 80$). At high frequencies the dipoles cannot follow the electric field leading to a frequencydependent relative permittivity

$$\varepsilon(f) = \varepsilon_{\infty} + \frac{\varepsilon_s - \varepsilon_{\infty}}{1 + j2\pi f\tau} \tag{1}$$

where ε_{∞} is the relative permittivity at high frequencies and τ is the relaxation time for the orientation of dipoles in the applied field.

Introducing ions into the solution has two effects. Some water molecules create a shell around ions (referred to as hydration). This has the effect of reducing the static permittivity and may also change the relaxation time τ . The added ions also give rise to ionic conductivity, leading to an additional term in the complex permittivity

$$\varepsilon(f) = \varepsilon_{\infty} + \frac{\varepsilon_s - \varepsilon_{\infty}}{1 + j2\pi f\tau} - j\frac{\sigma}{2\pi f\varepsilon_0}$$
(2)

where σ is the ionic conductivity.

We will use published experimental results to model the permittivity in NaCl and HCl solutions. NaCl solutions with salt concentrations close to that of seawater have been extensively studied and empirical expressions with varying numbers of parameters have been developed. We will use the expressions from Klein and Swift [3], expressed in terms of the salinity (grams NaCl/ kg water). The fits reported in [3] are determined for a wide range of temperature and salinity; approximate fit parameters valid at 298 K are presented in Table I. Also shown in the table are fit parameters for HCl solutions developed from data in [4] (dipole contribution) and [5] (ionic contribution). Here *m* is the molality of the solution.

In both cases these parameters provide a good fit near room temperature in the concentrations of interest in this paper. Some terms in the more general expressions [3] have been omitted where the error incurred is not significant. In particular, temperature has a significant effect on the polar contributions to the permittivity.

TABLE I. PARAMETERS FOR THE COMPLEX PERMITTIVITY OF NACL AND HCL SOLUTIONS

	NaCl (298 K)	HCl (288 K)
ε _s	73.34·(1 – 0.0031·S)	82
8∞	4.9	5.0
τ	$8 \times 10^{-12} \text{ sec}$	$11 \times 10^{-12} \sec$
σ	$\begin{array}{c} 0.183{\cdot}\mathrm{S} - 1.5\times10^{\cdot3}{\cdot}\mathrm{S}^2 + 2.1\times10^{\cdot5}{\cdot}\mathrm{S}^3 \\ - 1.3\times10^{\cdot7}{\cdot}\mathrm{S}^4\left[1/\Omega{\cdot}\mathrm{m}\right] \end{array}$	0.31· <i>m</i> [1/Ω·cm]

III. SURFACE WAVES IN CONTACT WITH FLUID

Simulations were performed in the piezoelectric devices and pressure acoustics modes of COMSOL 5.4. Figure 1 (left) shows the geometry for simulations. The substrate domain is 2 $\mu m \times 8 \mu m \times 30 \mu m$ in size, and the substrate coordinate system is rotated to simulate surface waves corresponding to the 36° rotated YX direction. Periodic boundary conditions are imposed on the *xy* and *yz* surfaces in the figure. The water domain (colored in the figure) is coupled to the lithium tantalate domain at the interface (boundary load and boundary normal acceleration are continuous). A perfectly matched boundary condition is imposed on the bottom 5 micrometers of the domain. Eigenfrequency simulations were performed and modes corresponding to surface waves that have a periodicity of 8 μm in the *x* direction were identified, and the corresponding surface wave velocity was calculated.



Figure 1. Geometry for the simulation of eigenmodes of the SAW substrate in contact with a liquid (left) a mode corresponding to a surface wave (right). In the right figure color corresponds to the transverse (y) displacement in the substrate and the pressure in the liquid.

As noted in the literature [6] either SH-SAW waves or surface skimming bulk waves (SSBW) can propagate on this surface depending on the electrical boundary condition. Figure 1 (right) shows an eigenmode found when the liquid domain has the properties of deionized water ($\varepsilon_r = 80$, $\sigma = 5.5 \times 10^{-6}$ $1/\Omega$ ·cm). For values of the relative permittivity near unity a surface-skimming bulk wave is observed (not shown). The eigenmode shown in Figure 1 (right) has a complex frequency $5.156 \times 10^8 + j22000$ Hz. The imaginary part of the eigenfrequency captures energy loss due to wave emission into the liquid and dielectric losses in the liquid. The attenuation due to these losses is very small (~0.29 dB/mm).

In the following sections we repeat these simulations including the dielectric properties for salt water and HCl solutions. These calculations yield the surface wave velocity and attenuation as a function of NaCl and HCl concentration, respectively.

IV. SURFACE WAVE PROPAGATION IN CONTACT WITH SALT WATER

Figure 2 and Figure 3 show the surface wave velocity and attenuation, respectively, for a substrate in contact with salt water.



Figure 2. Simulated surface wave velocity for a 36° rot YX LiTaO3 substrate in contact with salt water.



Figure 3. Simulated surface wave attenuation for a 36° rot YX LiTaO₃ substrate in contact with salt water.

Attenuation is very high at the salinity of ocean water (35 ppt). For lower values of the salinity the attenuation is less and there is a significant velocity change. A practical range of sensor operation is anticipated to lie between 2 and 20 ppt but

even in this range, the interaction length will need to be short, probably less than 1 mm.

V. SURFACE WAVE PROPAGATION IN CONTACT WITH HCL SOLUTION

Figure 4 and Figure 5 show the surface wave velocity and attenuation for a substrate in contact with an HCl solution. A pH value of 2 represents the highest concentration of interest in this study, and pH less than 6 corresponds to an additional ion content only an order of magnitude greater than the H^+ and OH⁻ concentrations in deionized water.



Figure 4. Simulated surface wave velocity for a 36° rot YX LiTaO₃ substrate in contact with an HCl solution.



Figure 5. Simulated surface wave attenuation for a 36° rot YX LiTaO₃ substrate in contact with an HCl solution.

For this range of pH, the attenuation is small and independent of pH. The small constant attenuation is due to the dissipation associated with reorientation of the water dipoles (eq. 1). The contribution to the loss of the ionic conductivity is negligible for this pH range. The velocity (Figure 5) is almost constant, with a very slight increase observed for the smallest value of pH. These results indicate that it is not practical to make a pH sensor by direct sensing of the dielectric properties. Instead one should seek a sensing overlayer that undergoes a mass change due to changes in pH.

VI. INITIAL EXPERIMENTAL RESULTS

SAW devices were fabricated on 36° rotated Y-cut Xpropagating LiTaO₃ substrates with 0.12 µm thick aluminum electrodes. The finger width and spacing was 2 µm and the aperture was 100 λ . The emitting IDT had 30 electrode pairs and there were two open (IDT-type) reflectors with 30 and 50 electrode pairs.

Initial testing was performed using a vector network analyzer (Rohde and Swartz ZVB4). Figure 6 shows the magnitude of the reflection coefficient S_{11} after Fourier transformation. In air reflections are observed at 1.46 and 1.14 μ s, consistent with the IDT path lengths of 2.38 and 3.08 mm.



Figure 6. Measured reflection coefficient S₁₁ after Fourier transformation to the time domain. Reflections remain observable for deionized water and pH 4 HCl solutions.

Initial experiments were performed by immersing the entire SAW device in aqueous solutions. This is highly non-optimal, as it introduces parasitic capacitive loading at the emitting and reflecting IDTs. In a practical sensor the emitting and reflecting IDTs would be covered with an insulating layer. Nevertheless, reflections were observed with reduced magnitude in deionized water solutions. Reflections with similar magnitude were observed in pH 4 HCl solution. However, reflections were completely absent in 10 ppt NaCl solution. These results are consistent with the simulations discussed above, which predict high attenuation in saline solutions.

Based on the results reported here, changes to the complex permittivity are too small over the pH range of interest to make a practical pH sensor. A better design will consist of a thin conducting layer on the interaction path (to favor the propagation of SH-surface waves) with a chemical sensing layer on top.

VII. SUMMARY

We have simulated the propagation of surface waves on a 36° rotated Y-cut X-propagating LiTaO₃ substrate in contact with ionic solutions. Experimental data from the literature has been used to obtain fits for the complex permittivity. The results show strong surface wave attenuation in salt water solutions and provide a direction for practical sensor development targeting pH sensing in aqueous conditions.

ACKNOWLEDGEMENTS

This work was performed in support of the US Department of Energy's Fossil Energy Crosscutting Technology Research Program. The Research was executed through the NETL Research and Innovation Center's Natural Gas Transmission and Delivery FWP. Research performed by Leidos Research Support Team staff was conducted under the RSS contract 89243318CFE000003.

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