MULTI-PARAMETERS DECOUPLING METHOD WITH LAMB WAVE SENSOR FOR IMPROVING THE SELECTIVITY OF LABEL-FREE LIQUID DETECTION

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ABSTRACT
The anti-symmetric modes (A01 mode for low frequency, A03 mode for high frequency) and symmetric modes (S0 mode) produced by Lamb wave sensor are used to detect multi-parameters of a liquid, such as its density, sound velocity and viscosity. These modes are found to be very different to the parameters. For example, the A01 mode is very sensitive to the liquid’s density but the A03 mode is sensitive to its sound velocity. The measurements of the attenuation with S0 mode can give out liquid’s viscosity after its density been determined by A01 mode. That could be a way to distinguish an unknown liquid with high sensitivity or to solve the problem of the selectivity of label free detection on biosensors.

KEYWORDS
Lamb wave sensor, Density, Sound velocity, Viscosity, Determination of multi-parameters

1. INTRODUCTION
Acoustic sensors have been widely used in chemical/biological fields with label free detection method that is to detect the mass changing on the sensors’ surface [1]. But the selectivity of this method is often poor due to the non-target molecules absorption and it’s hard to be distinguished by one parameter detection sensor. Multi-parameters’ detection with a multi-mode acoustic sensor make it possible. There have been some successful investigations for high viscosity solutions by the combination detection of density and viscosity [2]. However, for low viscosity solutions, like aqueous electrolytic solutions or bio liquid, it is still a challenge [2, 3].

The micro Lamb wave sensor is one of the powerful tools on liquid detection because it is easier to get multi-modes vibration and it shows high sensitivity and low attenuation. The two basic modes, the antisymmetric mode (A4 mode) and the symmetric mode (S4 mode), have been well known, and already been used for solving the problem of temperature compensation on chip by authors [4, 5].

In this work, the characters of A03 mode is investigated for the first time, whose wavelength is about one third of A01 mode (low frequency of A4 mode). It shows high sensitivity to sound velocity difference for different liquids. Two kinds of experiments were set up. One is for measuring the concentration changes with the same species’ solution, such as NaCl solution. The other is for distinguishing the components of the liquid with the same or even with different concentrations, such as KCl, NaBr and KBr solutions. The physical characters (density, sound velocity, viscosity) corresponding the different responses of different Lamb wave sensor’s modes (A01 mode, A03 mode, S0 mode) are tried to be decoupled and be compared with the values from literatures. Then the components of several kinds of aqueous electrolytic solutions are identified to show the affectivity of the decoupling method. It should be a potential method to improve the performance of acoustic biosensors.

2. EXPERIMENTS FOR LIQUID DETECTION

The micro Lamb wave device (Fig. 1(a)), contains a silicon membrane with a ground layer (Ti/Mo, GND) and a piezoelectric layer (Aluminum Nitride, AlN). Lamb waves are excited and detected directly using inter-digital transducers (IDTs) [6], which are located on the surface of AlN layer. The Si membrane length is about 7.84 mm. More details can be found in reference [7].

![Fig. 1. The system for liquid detection. (a) Schematic diagram of the micro Lamb wave sensor interaction with liquid, ρL: density, cL: sound velocity, ηL: viscosity. (b) Micro Lamb wave sensor packaged with printed circuit board (PCB).](image)

The micro Lamb wave device is packaged directly with the printed circuit board (PCB), as shown in Fig. 1(b). The network analyzer (Agilent 4395A), connecting with...
the PCB, is used to excite and receive the acoustic signals. The device is protected with one cover on the top of the system. The tube is used to flow in/out the liquid to chamber, which is sealed up with the PMMA cover under the PCB.

When the device is loaded with air or water, multi-modes can be excited and detected effectively, including the A01 mode, the A03 mode and the S0 mode, as shown in Fig. 2. As the A01 mode and the A03 mode are harmonic ones, the wavelength of the A03 mode is about 130 μm which is about one third of the A01 mode (390 μm). The resonant frequency of the A01 mode and the A03 mode are 0.987 MHz and 9.233 MHz respectively which are measured by our system, and then the corresponding phase velocities are 385 m/s and 1200 m/s.

\[
\delta_E = \frac{2\lambda}{\pi \rho L c_L^2} \left[1 - \left(\frac{c_P}{c_L}\right)^2\right] \quad [8],
\]
in which \(\rho_L\) and \(c_L\) are the liquid’s density and sound velocity, \(\lambda\) denotes the Lamb wave wavelength, \(c_P\) is the Lamb wave phase velocity. It means that the penetration depth is related to the ratio of \(\frac{c_P}{c_L}\).

When the phase velocity is far less than the liquid sound velocity, the effective loading mass \(m_E\) equals \(\frac{\rho_L \lambda}{2 \pi \left[1 - \left(\frac{c_P}{c_L}\right)^2\right]}\) \([\delta_E]\), in which \(\rho_L\) and \(c_L\) are the liquid’s density and sound velocity, \(\lambda\) denotes the Lamb wave wavelength, \(c_P\) is the Lamb wave phase velocity. It means that the penetration depth is related to the ratio of \(\frac{c_P}{c_L}\).

Experiments for different unknown species solutions

The second experiment is to measure these modes’ responses to three different species aqueous electrolytic solutions with different concentrations (Fig. 4). The S0 mode is still insensitive to the solutions changes compared with the measurement in water. The values of \(\Delta f/\text{f}\) of the A01 mode are not always negative so as the values of \(\Delta f/\text{f}\) in the A03 mode (Fig. 4). Especially for No. 6, No. 8 and No. 10 solutions, the values of \(\Delta f/\text{f}\) of the A01 mode are almost the same, but the values of \(\Delta f/\text{f}\) of the A03 mode are apparently different.

Fig. 3: Relative frequency shifts \(\Delta f/f\) for the A01 mode, the A03 mode and the S0 mode in the measurements of the NaCl solutions.

Experiments for one species solutions (NaCl solutions) with different concentrations

The first measurement is to measure one known species solution with different concentrations (Fig. 3), like NaCl solutions. When the water is taken into account as the reference liquid, the relative frequency shifts \(\Delta f/\text{f} = (f_{\text{solution}} - f_{\text{water}})/f_{\text{water}}\) with concentrations are different for these three modes (A01 mode, A03 mode and S0 mode), where \(f_{\text{solution}}\) and \(f_{\text{water}}\) are the measured frequencies for the solution and the water. The frequency of A01 mode decreases with the concentrations and the A03 mode has opposite behavior. The frequency of the S0 mode does not show apparently shifts with different concentrations as the real part of the phase velocity of the longitudinal waves (S0 mode) will not be influenced by the liquid [9].

Experiments for some different unknown species solutions

The second experiment is to measure these modes’ responses to three different species aqueous electrolytic solutions with different concentrations (Fig. 4). The S0 mode is still insensitive to the solutions changes compared with the measurement in water. The values of \(\Delta f/\text{f}\) of the A03 mode are not always negative so as the values of \(\Delta f/\text{f}\) in the A01 mode (Fig. 4). Especially for No. 6, No. 8 and No. 10 solutions, the values of \(\Delta f/\text{f}\) of the A01 mode are almost the same, but the values of \(\Delta f/\text{f}\) of the A03 mode are apparently different.

Fig. 4: Relative frequency shifts \(\Delta f/f\) for the A01 mode, the A03 mode in the measurements of some unknown solutions.

Viscosity measurements with the S0 mode

The central frequency of the S0 mode does not show
apparently shifts with different density and acoustic velocity (Fig. 3), which indicates that the S0 mode is suitable for determining the viscosity. As it was proposed in reference [8], the attenuation coefficient $\alpha$ is proportional to $\rho \eta^2$. The amplitude ($A_L$) response one of an unknown solution can be expressed by

$$A_L = A_0 e^{-\alpha_{\text{water}} x}$$  \hspace{1cm} (1)

Similarly, the amplitude response ($A_W$) of water is given by

$$A_W = A_0 e^{-\alpha_{\text{water}} x}$$  \hspace{1cm} (2)

in which, $\alpha_{\text{water}}$ is the attenuation coefficient in water.

In engineering, insertion loss (dB) is used widely. The values of $A_L$ and $A_W$ can be transformed into the values in dB scale which are denoted $A_{L\text{dB}}$ and $A_{W\text{dB}}$ respectively. Therefore, the attenuation difference ($\Delta A_{L\text{dB}}$) between an unknown solution and water can be expressed in dB scale

$$\Delta A_{L\text{dB}} = A_{L\text{dB}} - A_{W\text{dB}} = \gamma_1 (\rho \eta)^{1/2} + \gamma_2$$  \hspace{1cm} (3)

in which, the slope $\gamma_1$ is constant, and the constant $\gamma_2$ is decided by the reference liquid (water). The amplitude of the S0 mode in NaBr solution (Fig. 5) shows that the energy losses increase with the concentration.

![Fig. 5. Amplitude response to the different concentration of NaBr solutions in the S0 mode.](image)

3. RESULTS AND DISCUSSIONS

To decouple the density and the acoustic sound velocity with $A_{01}$ mode

When a low viscosity fluid contacts one side of a thin Lamb wave device, the mass loading caused by the fluid within the evanescent wave field can be described by simply adding an additional term of the mass per unit area of the membrane ($M$). Then, taking into account the membrane bending stiffness and the in-plane tension, the mass loading effect to the phase velocity of $A_{01}$ mode or $A_{03}$ mode can be expressed as [9].

$$\rho_L \lambda_i / 2 \pi c_i \left[1 - (c_i / c_L)^2\right] = B / c_{L_0} - M$$  \hspace{1cm} (4)

where $i$ denotes 1 or 3 for the mode $A_{0i}$. $B$ reflects the influences of the bending stiffness and the in-plane tension of the plate for the mode $A_{0i}$. $\lambda_i$ denotes the Lamb wave wavelength in each mode. In this formula, the constants $B$, $\lambda_i$, and $M$ don't depend on the liquid type. $\lambda_i$ is determined by the structure of the device. $M$ is about 0.0355 kg/m$^2$. $c_{L_0}$ is determined by the frequency ($f$) via $c_{L_0} = f \lambda_i$.

In order to decouple the density and the sound velocity of one liquid, the process can be distinguished into two steps:

Step 1. to determine the constant $B$ by measuring the frequency response of the reference liquid.

Step 2. to get the physical parameters ($\rho_L$, $c_L$) for one unknown liquid by measuring $c_{L_0}$. All other parameters ($B$, $\lambda_i$, $M$) in Equation (4) has all ready been calculated or measured.

The first decoupled result using the Equation (4) is the NaCl solutions with different concentrations, as it was measured in Fig. 3. Comparing the measured density and sound velocity with the values from the literature [10], the two results are not far, as shown in Fig. 6.

![Fig. 6: The measured density and sound velocity of NaCl solutions and other solutions, using measurements in $A_{01}$ mode and $A_{03}$ mode.](image)

Based on the same process, the density and the sound velocity for different species solutions with different concentrations are also decoupled and shown in Fig. 6. Comparing with the values from the literature [10], No. 1-No. 6 solutions are KCl solutions; No. 7 and No. 8 solutions are NaBr solutions; No. 9 and No. 10 solutions are KBr solutions. Beside of decoupled density and sound velocity, the species of the solutions can be identified with comparing the measured values with the already known values [10].

For No. 6, 8 and 10 solutions, the decoupled densities (Fig. 6) are almost the same as the relative frequency shifts are very close (Fig. 4). For these solutions with
adjacent density, sound velocities become the main factors affecting the frequency shifts in the A₃₁ mode (Fig. 4). With the relations of the absolute frequency shifts: Δ/fₚ₆ > Δ/fₚ₈ > Δ/fₚ₁₀ (Fig. 4), the decoupled sound velocity of these three solutions have such relation: cₚ₆ > cₚ₈ > cₚ₁₀ (Fig. 6).

The linear response of (density×viscosity)⁰.⁵ changing with the item of amplitude shifts (ΔA dB) in S₀ mode.

The item (density×viscosity)⁰.⁵ changes almost linearly with the item of amplitude shifts (ΔA dB) and the linear fitting coefficient is about 0.32 (dB/kg m⁻² s⁻⁰.⁵), like NaBr and NaCl solutions (Fig. 7). By analyzing the amplitude shifts (ΔA dB) in S₀ mode, the viscosity of the solution can be got. With the determined density measuring the frequency response in A₃₁ mode and the A₃₃ mode, the viscosity will be known by checking the amplitude response in S₀ mode.

Fig. 7: Amplitude shifts vs. (Density×Viscosity)⁰.⁵ for the NaCl and NaBr solutions in the S₀ mode.

4. CONCLUSION

In this work, for the first time with our knowledge, the density, sound velocity and viscosity of the liquid are obtained simultaneously based on the measurements of the same volume solution with a Lamb wave sensor. Combination of the frequency shifts of A₃₁ mode and A₃₃ mode, the density and sound velocity are decoupled. It is because that the phase velocity of A₃₁ mode is far from the sound velocity of the liquid and the phase velocity of A₃₃ mode is close to the sound velocity of the liquid. The viscosity is obtained with the amplitude measurement of S₀ mode. With this multi-parameters detection method, the unknown solutions, like aqueous electrolytic solutions, have been distinguished successfully. The present results clearly indicate that the multi-modes of micro Lamb wave sensor could have great promise applications in the investigation of the molecular thermodynamics, adiabatic compressibility, molecular label free detection, etc.

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