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Measurements of Standard-Viscosity Liquids Using Shear Horizontal Surface Acoustic Wave Sensors

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A shear horizontal surface acoustic wave (SH-SAW) sensor can be used to detect the properties of liquids. We propose one application of the SH-SAW sensor as a viscosity sensor. The Japanese Industrial Standards Committee (JISC) has determined the standard liquids used to calibrate a viscometer. If the SH-SAW sensor is to be commercialized, it is necessary that the viscosities of these liquid can be measured. In this study, the standard-viscosity liquids are measured using the SH-SAW sensor operating at different frequencies. The sensor responses to the standard-viscosity liquids are less than those for a glycerol/water mixture. This is due to the viscoelasticity of the standard-viscosity liquids. A simple viscoelastic model is applied to explain the experimental results.

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1. Introduction

Surface acoustic wave (SAW) devices are being applied in a variety of fields. Sensors are one application of SAW devices.[1–3] The Rayleigh-SAW is used in a gas sensor, and the shear horizontal SAW (SH-SAW) is used in a liquid-phase sensor.[1] The SH-SAW sensor is used to detect liquid properties, such as conductivity, dielectric constant, viscosity, and density. A feature of the SH-SAW sensor is the simultaneous detection of the mechanical and electrical properties of a liquid with high sensitivity.

We have mainly investigated SH-SAW sensors, which are capable of measuring the dielectric constant and conductivity.[3,4] Studies of the measurement of density and viscosity products using SH-SAW sensors, however, have not been enough. In the fundamental experiments on density and viscosity products, a glycerol/water mixture was used as a calibrating liquid.[5] However, because the properties of glycerol change depending on the amount of absorbed water and because its viscosity is a function of temperature, a glycerol/water mixture is not applicable as a standard liquid for viscosity measurements. The Japanese Industrial Standards Committee (JISC) has determined the standard liquids used for calibrating viscometers (JIS Z8809). Standard-viscosity liquids are hydrocarbon oils. We have not previously measured standard-viscosity liquids using an SH-SAW sensor. If an SH-SAW sensor is to be developed and commercialized as a viscosity sensor, the viscosity measurements of standard-viscosity liquids is required. In this study, standard-viscosity liquids are measured and compared with a glycerol/water binary mixture. The results for standard-viscosity liquids cannot be explained by the theory of Newtonian fluids. Theory based on the Maxwell model is utilized to discuss the results.

2. Experimental

A 36YX-$LiTaO_3$ single crystal was utilized for the SH-SAW substrate. The propagation surface was electrically shorted by evaporated films of chromium and gold, as shown in Fig. 1. The design parameters of the SH-SAW sensor are summarized in Table I. A liquid cell was fabricated on the propagation surface, as shown in Fig. 1.

The measurement system is shown in Fig. 2. A 50 MHz signal from a signal generator (Anritsu MG3601A) was fed to the SH-SAW sensor. The outputs from the sensor were monitored with a vector voltmeter (HP 8508A). The phase difference between the sensor output and the reference signal was measured using a vector voltmeter. The velocity shift, $\Delta V/V$, was derived from the phase shift. SH-SAW sensors were used at the frequencies of 30, 50, and 100 MHz. Distilled water was used as the reference liquid. The sample liquids were glycerol/water binary mixtures with different concentrations (20, 40, 60, and 80 wt%) and standard-viscosity liquids (4.7, 9.5, 49, 98, 500, and 990 mPa·s). All samples were injected into the liquid cell using a micropipette.

3. Results and Discussion

Using the SH-SAW sensors, we measured the glycerol/water binary mixtures and standard-viscosity liquids. The experimental results are shown in Figs. 3 and 4, respectively. The abscissa is the square root of density and viscosity products. The ordinate is the velocity shift. For the glycerol/water binary mixtures, the velocity shift is proportional to the square root of density and viscosity products. Glycerol/water binary mixtures are known to be Newtonian fluids. For this reason, they were previously used as standard-viscosity liquids for the SH-SAW sensor. For standard-viscosity liquids, the velocity shift saturates as the square root of density and viscosity products increases. The currently used standard-viscosity liquids are composed of silicone oil. Because the intermolecular force between siloxane molecules in silicone oil is weak, the interaction between molecules in silicon oil is weak and the temperature dependence of its viscosity is also weak. Standard-viscosity liquids, which are known to be Newtonian fluids, have been used for calibrating viscometers. The frequency of conventional viscometers is low. However, because the frequencies of SH-SAW sensors are higher than those of viscometers, the standard-viscosity liquids do not behave as Newtonian fluids. Therefore, the liquids must be treated as non-Newtonian fluids. As the standard-viscosity liquids are high-polymer solutions, a viscoelastic fluid model is used.

A viscoelastic fluid has viscous and elastic properties. The liquid is modeled by the Maxwell model,[6,7] as shown in Fig. 5. The Maxwell model is expressed as a series of springs and dashpots, and its behavior depends on frequency: at low frequency, it acts as a viscous liquid and the spring can be ignored. When the frequency increases, the influence of the spring becomes apparent. In this paper, the theoretical equation for the SH-SAW sensor is derived on the basis of the Maxwell model. The relationship between stress, T, and strain, S, for the Maxwell model is expressed as

$$\frac{\partial S}{\partial t} = \frac{1}{\eta}T + \frac{1}{G}\frac{\partial T}{\partial t}. \tag{3.1}$$

Here, η and G are the viscosity and shear modulus, respectively. When the time variation of strain is assumed to be $\exp(j\omega t)$, the relationship between stress and strain becomes

$$T = \frac{j\omega\eta}{1 + j\omega\tau}S. \tag{3.2}$$

Here, ω is the angular frequency and $\tau(= \eta/G)$ is the relaxation time. When $\omega\tau << 1$, eq. (3.2) becomes

$$T = j\omega\eta S.$$

This equation represents the Newtonian fluid.

The surface acoustic impedance, Z, for the Maxwell model is derived from eq. (3.2) as[8–10]

$$Z = \begin{bmatrix} -j\frac{\mu}{V}\sqrt{1 - \frac{\rho V^2}{\mu}} & 0 & \frac{\mu}{V} \\ 0 & -j\frac{\mu}{V}\sqrt{1 - \frac{\rho V^2}{\mu}} & 0 \\ \frac{\lambda}{V} & 0 & -j\frac{\lambda+2\mu}{V}\sqrt{1 - \frac{\rho V^2}{\lambda+2\mu}} \end{bmatrix};$$

where V is the phase velocity, ρ is the density of the sample liquid, and λ and μ are

$$\lambda = \kappa - \frac{j\frac{2}{3}\omega\eta}{1 + j\omega\tau},$$

$$\mu = \frac{j\omega\eta}{1 + j\omega\tau}.$$

Here, κ is the bulk modulus of the fluid. Substituting the imaginary part of Z into eq. (3.3), the velocity shift, $\Delta V/V$, for the Maxwell model is obtained.

$$\frac{\Delta V}{V} = -A\,(\mathbf{v}_n \cdot \mathbf{Z}_i^{*} \cdot \mathbf{v}_n + \mathbf{v}_n \cdot \mathbf{Z}_i^{*} \cdot \mathbf{v}_n)\,; \tag{3.3}$$

$$\frac{\Delta V}{V} = -A\left[\frac{1}{V}\left(M_1^2 + M_2^2\right)^{1/4}\sin(M_3) - \sqrt{\frac{\omega\rho'\eta'}{2}}\right]. \qquad (3.4)$$

Here,

$$M_1 = \frac{(\omega\eta)^2 - (\omega^2\eta\tau)^2}{[1-(\omega\tau)^2]^2 + (2\omega\tau)^2} + \frac{\omega^2\rho\eta\tau V^2}{1+(\omega\tau)^2}$$

$$M_2 = \omega\eta\left\{\frac{\rho V^2}{1+(\omega\tau)^2} - \frac{2\omega^2\eta\tau}{[1-(\omega\tau)^2]^2 + (2\omega\tau)^2}\right\}$$

$$M_3 = \frac{1}{2}\tan^{-1}\left(\frac{M_2}{M_1}\right);$$

where ρ' and η' are density and viscosity of a reference liquid, respectively, and A is the material constant of the substrate.

Using eq. (3.4), the experimental results for the standard-viscosity liquids are evaluated. The density and viscosity for the standard-viscosity liquids are known. Because the shear modulus is unknown, it is varied. The calculated results at 17 MPa and experimental results at 50 MHz are shown in Fig. 6. For a small density and viscosity product, the experimental values agree with those of the theory. Using the Maxwell model, the fluid can be divided into two categories, Newtonian and viscoelastic fluids, on the basis of the product of angular frequency and relaxation time, $\omega\tau$, as follows: when $\omega\tau \ll 1$, the liquid is a Newtonian fluid, and when $\omega\tau \gg 1$, the liquid is a viscoelastic fluid. The region with a small density and viscosity product corresponds to Newtonian fluids, for which there is no influence of the shear modulus, G. The shear modulus was then varied and the results were compared with the experimental results. Figure 6 indicates that the calculated results do not agree with the experimental results above a certain shear modulus. Next, the experimental results for each liquid are substituted into eq. (3.4) and the shear modulus is estimated. Figure 7 shows the results. The figure indicates that the shear modulus depends on the sensor frequency and the density and viscosity product. The relationship between the shear modulus and the square root of the density and viscosity product is linear. The relationship between these and frequency is also linear.

Poly(ethylene glycol) (PEG) is selected as another example of a high polymer solution. As it is assumed to be a viscoelastic liquid, we compared PEG with the standard-viscosity liquids. PEG samples with different molecular weights (200, 300, and 400) were measured. The experimental results are shown in Fig. 8. All other results in this study are also shown in Fig. 8. Since the liquid properties of PEG are unknown, the abscissa and ordinate of the figure are the velocity shift and attenuation change, $\Delta\alpha/k$, respectively. The attenuation change is derived from the amplitude change. For PEG, the amplitude change increases with increasing molecular weight. However, the velocity shift does not change. In Fig. 8, the solid line corresponds to Newtonian fluids. The results for low-concentration glycerol/water mixtures fall on this line. Therefore, they behave as Newtonian fluids. For high-concentration glycerol/water mixtures, however, the results do not agree with those for the Newtonian fluids. Therefore, it is necessary to consider the influence of the viscoelastic properties. The figure also indicates that the standard-viscosity liquids and PEG samples show similar tendencies. Therefore, it may be possible to discuss the viscoelastic properties using the velocity shift - attenuation change plane.

4. Conclusions

In this paper, standard-viscosity liquids, which have been used for calibrating viscometers, were measured using an SH-SAW sensor. It is well known that a standard-viscosity liquid is a Newtonian fluid. As the frequency of the SH-SAW sensor is higher than that of a conventional viscometer, the velocity shifts for standard-viscosity liquids with high density and viscosity products were saturated. Our results were too complicated to explain with the theory for Newtonian fluids. Therefore, theory based on the Maxwell model for viscoelastic fluids was utilized. For the case of $\omega\tau \ll 1$, the Maxwell model agrees with Newtonian fluids. The results for the standard-viscosity liquids with low density and viscosity products agreed with the theory. This means that a standard-viscosity liquid with a small density and viscosity product can be used for calibrating SH-SAW sensor. For the case of $\omega\tau \gg 1$, the influence of the shear modulus cannot be ignored. Using the experimental results and the theory, the shear modulus was estimated.

As an other example of a viscoelastic fluid, PEG was measured. The velocity shift - attenuation plane was used to evaluate the results. For a Newtonian fluid, the absolute value of velocity shift agrees with the attenuation change. For a viscoelastic fluid, however, it is less than the attenuation change. Using the velocity shift - attenuation change plane, it is possible to decide whether a liquid is a Newtonian or a viscoelastic fluid. In future work, we aim to establish a method of simultaneously detecting density, viscosity, and shear modulus.

Acknowledgement

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Figure captions

Fig. 1. Schematic illustration of SH-SAW sensor.

Fig. 2. Measurement system and samples.

Fig. 3. Experimental results for glycerol/water binary mixtures. The parameter is SH-SAW sensor frequency.

Fig. 4. Experimental results for standard-viscosity liquids. The parameter is SH-SAW sensor frequency.

Fig. 5. Maxwell model. G and η are shear modulus and viscosity, respectively.

Fig. 6. Experimental results for 50 MHz SH-SAW sensor and solution of eq. (3.4). The shear modulus was fixed at 17 MPa.

Fig. 7. Estimated results of shear modulus from experimental results.

Fig. 8. Experimental results for glycerol/water binary mixture, standard-viscosity liquid, and PEG. The solid line shows the theoretical result for a Newtonian fluid.

Table I. Design parameters of SH-SAW sensors.

Frequency (MHz)	ï (ñm)	W (mm)	N	L (mm)
30	140	3.0	34	11
50	80	2.0	32	13
100	40	2.0	25	11

ï: wavelength, W: IDT aperture, N: number of pairs, L: IDT center-center separation.

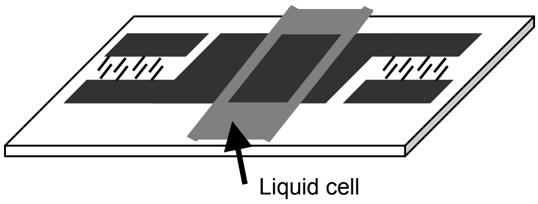

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Fig. 1

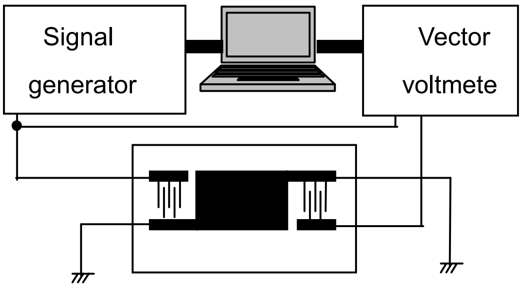

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Fig. 2

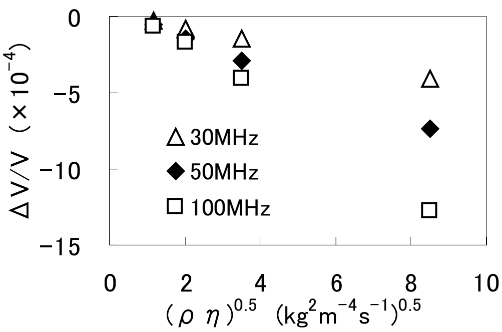

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Fig. 3

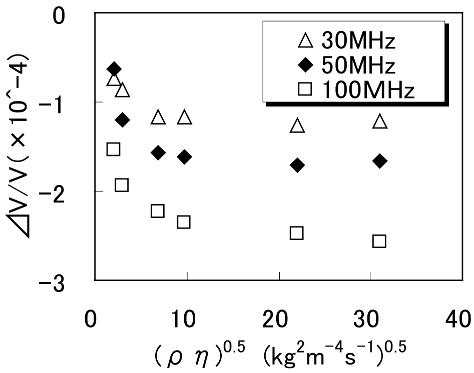

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Fig. 4

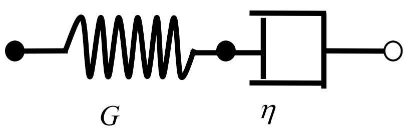

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Fig. 5

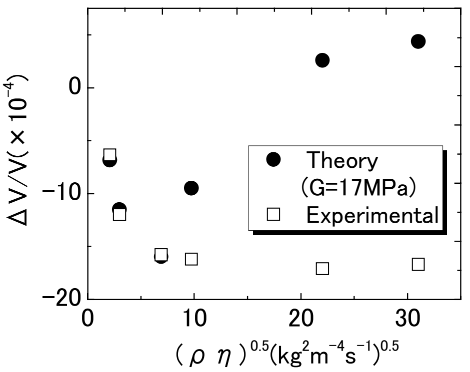

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Fig. 6

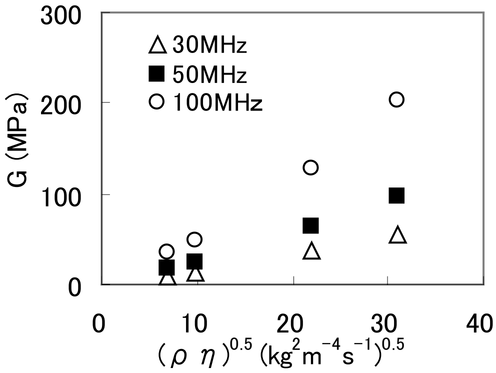

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Fig. 7

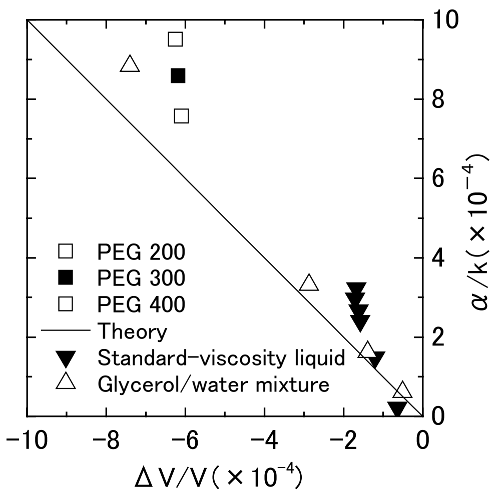

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Fig. 8