

## Measurement of Liquid Viscosity and Density Using Single Piezoelectric Resonator with Two Vibration Modes

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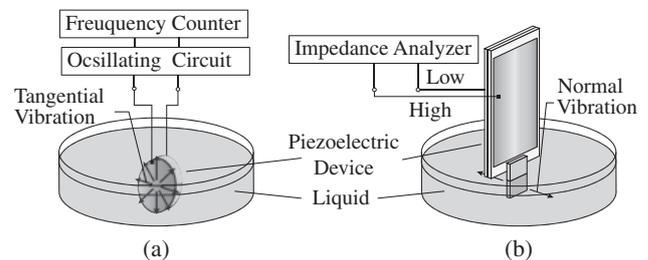
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We report the results of an experiment on measuring the viscosities and densities of several liquids using a single piezoelectric sensor, since only the numerical results obtained by finite element analysis were reported in our previous paper. The novelty of the sensor is that the viscosity and density can be inferred simply by measuring resonance frequencies in liquid for the vibration in the tangential and normal directions with respect to the contact surface between the sensor and the liquid, while the method suggested as reference requires measurements of resonance frequency and damping of a single vibration mode. By comparing the viscosities and densities measured by the proposed and conventional methods using food oil, the densities were found to correspond to the values measured using a weight meter with an error within 1% and the viscosity was evaluated to be higher than that measured using a viscometer with an error within 10%. The results suggest the possibility of measuring liquid density and viscosity by the proposed method. © 2012 The Japan Society of Applied Physics

### 1. Introduction

Many liquids are produced for foods, beauty goods, and industrial manufactures. Viscosity and density are the important criteria for quality control among the many physical parameters. Developments of a viscometer and a density meter are, therefore, a subject of considerable practical interest.<sup>1,2</sup> Many researchers have recently used a resonator in the measurement of physical properties in the three states of matter including liquid. These resonator sensors are based on time measurement, which is widely used for many applications, for example, those regarding gas density,<sup>3,4</sup> liquid density,<sup>2</sup> liquid viscosity,<sup>5</sup> fluid flow velocity, liquid level,<sup>6</sup> solid elasticity,<sup>7,8</sup> solid mass, and solid acceleration,<sup>9,10</sup> because the time measurement has a high accuracy. Plural resonators, however, are required to measure multiple parameters,<sup>11</sup> especially liquid viscosity and density.

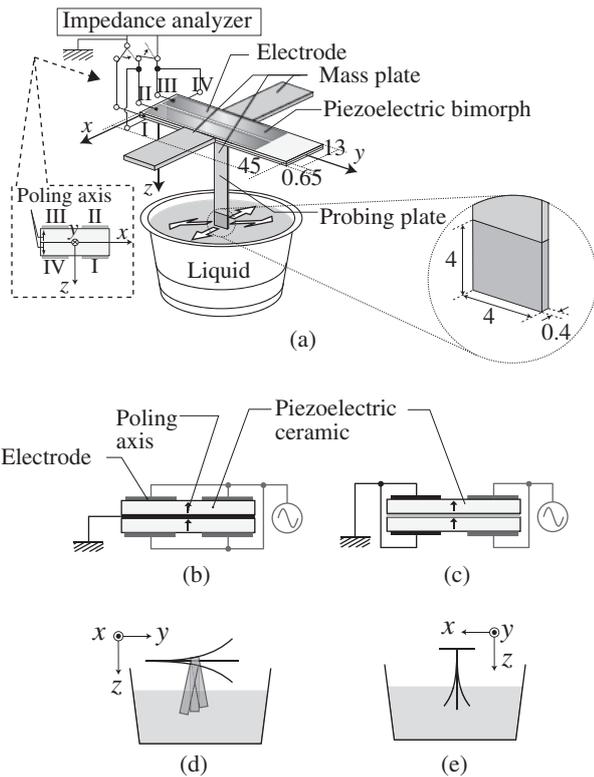
Thus, new devices that can measure two or more parameters at the same time have been developed. Among them, piezoelectric devices have attracted attention as they can be used to measure the density and viscosity. The devices are categorized as microscopic and macroscopic sensors. Examples of microscopic sensors are quartz crystal resonators<sup>12–14</sup> (QCRs) and surface acoustic wave (SAW) devices.<sup>15–17</sup> In these sensors, a small amount of liquid is placed on the device, and the liquid properties are measured. However, liquids tend to show elastic behavior at high frequencies, such as  $10^6$  through  $10^8$  Hz, owing to relaxation effects, which are mathematically represented by a Maxwellian model including an imaginary part in viscosity.<sup>15</sup> Therefore, even a standard liquid of low viscosity is treated as a non-Newtonian liquid, and the results cannot be compared with those obtained using conventional viscometers at low frequencies of  $10^1$  through  $10^2$  Hz. On the other hand, the macroscopic sensors are immersed in a liquid. In early studies, two types of macroscopic device were developed. In one type, the contact surface between the device and the liquid vibrates in the tangential direction.<sup>5,18,19</sup> In the other type, the contact surface vibrates in the normal direction.<sup>20–22</sup> For instance, Momozawa and



**Fig. 1.** Conventional sensor of liquid viscosity and density: (a) tangential vibration type for measuring product of viscosity and density observed from resonance frequency and (b) normal vibration type for independently measuring viscosity and density observed from resonance frequency and damping.

Imano developed a piezoelectric disk sensor using the tangential vibration of the contact surface, as shown in Fig. 1(a),<sup>5</sup> and Riesch *et al.* developed a sensor using the normal vibration, as shown in Fig. 1(b).<sup>22</sup> Both macroscopic devices can be compared with conventional viscometers, because these macroscopic devices measure the viscosity at low frequencies, such as  $10^2$  through  $10^4$  Hz. These methods, however, have technical issues. The tangential vibration device cannot independently measure viscosity and density, but can measure only the product of the liquid density and viscosity. The normal vibration device requires the measurement of resonance frequency and damping to independently infer the density and viscosity; thus, the measurement equipment, such as the impedance analyzer, is complex, because the impedance must be measured at various frequencies to induce damping. Recently, we devised a measurement technique in which a resonator is oscillated in two vibration modes by a single device, and independently measured the viscosity and density by observing only the resonance frequencies.<sup>23,24</sup> The proposed technique may make measurement systems simpler than the previous equipment because the proposed technique only requires the observation of two resonance frequencies corresponding to the two vibration modes to be measured. We have confirmed that the viscosity and density are independently measurable by this measurement technique

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**Fig. 2.** Proposed sensor for independently measuring liquid viscosity and density observed from resonance frequencies in tangential and normal vibrations: (a) measurement setup, (b) vibration mode of contact surface between sensor and liquid displaced in tangential direction, and (c) vibration mode of contact surface between sensor and liquid displaced in normal direction.

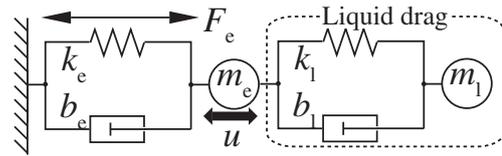
with the help of a finite element method, which is effective for the analysis of a complex-shaped piezoelectric device.

In this paper, we report the results of an experiment on measuring the liquid viscosity and density of several liquids using the proposed sensor, since only the numerical results obtained by finite element analysis were reported in our previous paper.<sup>23)</sup>

## 2. Structure of Proposed Sensor and Principle of Liquid Viscosity and Density Measurement

The sensor consists of a piezoelectric bimorph and copper plates, as shown in Fig. 2(a). One end of the bimorph is fixed, and the bimorph works as a resonating cantilever. The tip of the probing plate attached to the bimorph is immersed in a liquid. The thickness and width of the plate are 0.4 and 4 mm, respectively, and the immersion depth in the liquid is set to 4 mm. The other two plates attached to the two sides of the bimorph work as extra masses to obtain a noteworthy resonance characteristic of a vibration mode in the normal direction with respect to the contact surface between the probing plate and the liquid. Two electrodes are located on each surface of the bimorph. By selectively applying voltage to the electrodes, as shown in Figs. 2(b) and 2(c), the probing plate vibrates in the tangential and the normal direction with respect to the contact surface, as shown in Figs. 2(d) and 2(e), respectively.

The sensor vibrating in each direction with respect to the contact surface in a liquid is expressed by a simple harmonic



**Fig. 3.** Equivalent spring-mass-damper model of proposed sensor immersed in liquid.

motion model, as shown in Fig. 3.<sup>25)</sup> Here,  $F_e$ ,  $k_e$ ,  $m_e$ , and  $b_e$  represent the external force, the equivalent spring, the mass, and the damper, respectively. When the simple harmonic motion model vibrates with the displacement  $u$  induced by the external force  $F_e$ , the reaction force from the liquid is caused by  $u$ .<sup>26)</sup> The additional liquid spring  $k_1$ , mass  $m_1$ , and damper  $b_1$  are expressed in terms of the reaction force. The following is the motion equation expressing the simple harmonic motion model:<sup>27)</sup>

$$F_e = [(k_e + k_1) - \omega^2(m_e + m_1)]u + j\omega(b_e + b_1)u. \quad (1)$$

Here,  $\omega$  and  $j$  are the angular frequency and an imaginary unit, respectively.

Firstly, the compressibility of liquid in this measurement is estimated from the change ratio of the liquid density  $\rho$ . When the change ratio  $d\rho/\rho$  becomes larger than 5%, we must consider the compressibility.<sup>28)</sup> Thus, we focus on the additional liquid spring  $k_1$  involved with the compressibility and consider  $k_1$  as the bulk modulus of liquid. The change ratio is given by

$$\frac{d\rho}{\rho} = \frac{1}{2} M_k^2. \quad (2)$$

Here,  $M_k$  is the kinetic Mach number indicated by the ratio of flow velocity to sound velocity given by

$$M_k = \frac{\omega D}{\sqrt{k_1/\rho}}. \quad (3)$$

Here,  $D$  shows the characteristic length of the contact surface. When the values of  $\omega$ ,  $D$ ,  $k_1$ , and  $\rho$  assumed in this measurement are on the order of  $10^3$  rad/s,  $10^{-3}$  m,  $10^9$  Pa, and  $10^3$  kg/m<sup>3</sup>, respectively, the values of  $M_k$  and  $d\rho/\rho$  are on the order of  $10^{-3}$  and  $10^{-6}$ . We leave the additional liquid spring out in eq. (1), because the change ratio in this measurement becomes sufficiently smaller than 5%.

When the sensor immersed in the liquid vibrates in the tangential or normal direction, the additional liquid masses  $m_{lt}$  and  $m_{ln}$  are expressed by the viscosity  $\eta$  and density  $\rho$ .<sup>28,29)</sup>

$$m_{lt} = \frac{A}{\sqrt{2}} \sqrt{\frac{\eta\rho}{\omega}}, \quad (4)$$

$$m_{ln} = 3\sqrt{2}\pi R^2 \sqrt{\frac{\eta\rho}{\omega}} + \frac{2\pi R^3}{3} \rho. \quad (5)$$

Here,  $A$  and  $R$  show the area of the contact surface and the radius of a sphere in the liquid, respectively. In the normal direction, the vibration of the liquid around the contact surface is approximately substituted by that of the sphere.<sup>22)</sup> The radius  $R$  becomes almost one-half the width of the plate, if the width and depth of the plate in the liquid are the same.

When the sensor vibrates in the liquid, the resonance frequency shift is caused by the additional liquid masses expressed in eqs. (4) and (5). The resonance frequency in air,  $f_{rAir}$ , and that in liquid,  $f_{rLiq}$ , are given by

$$f_{rAir} = \frac{1}{2\pi} \sqrt{\frac{k_e}{m_e}}, \tag{6}$$

$$f_{rLiq} = \frac{1}{2\pi} \sqrt{\frac{k_e}{m_e + m_l}}, \tag{7}$$

from eq. (1), respectively. In the experiment, both the additional masses  $m_{lt}$  and  $m_{ln}$  can be obtained from  $f_{rAir}$  and  $f_{rLiq}$  of the tangential and normal vibrations expressed in eqs. (6) and (7), as

$$m_{lt} = m_{et} \left[ \left( \frac{f_{rAir}}{f_{rLiq}} \right)^2 - 1 \right], \tag{8}$$

$$m_{ln} = m_{en} \left[ \left( \frac{f_{rnAir}}{f_{rnLiq}} \right)^2 - 1 \right]. \tag{9}$$

Here,  $m_{et}$  and  $m_{en}$  show the equivalent masses in each vibration mode. Firstly, in the tangential vibration, the root of the product of the viscosity and density can be derived by observing  $f_{rtAir}$  and  $f_{rtLiq}$ , from eqs. (4) and (8), as

$$\sqrt{\eta\rho} = m_{et} \frac{\sqrt{2\omega}}{A} \left[ \left( \frac{f_{rtAir}}{f_{rtLiq}} \right)^2 - 1 \right]. \tag{10}$$

Next, in the normal vibration,  $f_{rnAir}$  and  $f_{rnLiq}$  are observed to determinate the density  $\rho$  of the liquid, which is described by eqs. (5), (9), and (10), as

$$\rho = \frac{3}{2\pi R^3} \left\{ m_{en} \left[ \left( \frac{f_{rnAir}}{f_{rnLiq}} \right)^2 - 1 \right] - m_{et} \frac{6\pi R^2}{A} \sqrt{\frac{\omega_{rtLiq}}{\omega_{rnLiq}}} \left[ \left( \frac{f_{rtAir}}{f_{rtLiq}} \right)^2 - 1 \right] \right\}. \tag{11}$$

Finally, the viscosity of the liquid is determined from eqs. (10) and (11), as

$$\eta = \frac{\left\{ m_{et} \frac{\sqrt{2\omega}}{A} \left[ \left( \frac{f_{rtAir}}{f_{rtLiq}} \right)^2 - 1 \right] \right\}^2}{\frac{3}{2\pi R^3} \left\{ m_{en} \left[ \left( \frac{f_{rnAir}}{f_{rnLiq}} \right)^2 - 1 \right] - m_{et} \frac{6\pi R^2}{A} \sqrt{\frac{\omega_{rtLiq}}{\omega_{rnLiq}}} \left[ \left( \frac{f_{rtAir}}{f_{rtLiq}} \right)^2 - 1 \right] \right\}}. \tag{12}$$

Therefore,  $\eta$  and  $\rho$  are inferred by observing the resonance frequencies of the tangential and normal vibrations in air and liquid. Similar to the case of the conventional sensor in the tangential vibration, only the product of the liquid  $\eta$  and  $\rho$  can be inferred by observing the resonance frequency. On the other hand,  $\eta$  and  $\rho$  can be independently inferred by observing those of both the tangential and normal vibrations in the proposed method. Moreover, it is not necessary to measure the damping of the device immersed in the liquid, whereas the measurement of both resonance frequency and damping is required in the previous sensor to independently infer  $\eta$  and  $\rho$ . Therefore, the measurement equipment can be simplified because an oscillating circuit for observing resonance frequency can be used instead of the impedance analyzer for observing a resonance characteristic. In this method, however, we must experimentally determine the equivalent mass of the device in the tangential vibration,  $m_{et}$ , and that in the normal vibration,  $m_{en}$ , because the structure of the sensor device is so complex that  $m_{et}$  and  $m_{en}$  are difficult to derive theoretically.

### 3. Experimental Procedure

We must obtain quantitative values of equivalent masses  $m_{et}$  and  $m_{en}$  in an experiment before measuring the liquid viscosity and density. The values can be obtained by measuring each resonance frequency when pairs of copper sheets, whose length, width, and thickness are 4.0, 4.0, and 0.035 mm, respectively, are attached to the tip of the sensor assumed to be the contact surface between the sensor and the liquid, as shown in Fig. 4. The mass of the sheets is obtained using a commercial weight meter (Shimadzu EL-600). Figure 5 shows the mass of the sheets with changing volume. The density of the sheets ( $8170 \text{ kg/m}^3$ ) is obtained

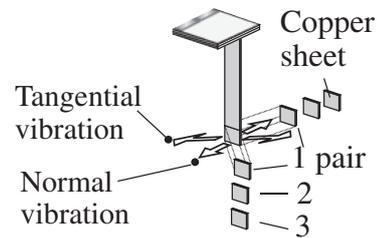


Fig. 4. Experimental setup for measuring quantitative values of equivalent masses of sensor in tangential and normal vibration modes under copper sheet attached to tip of sensor assuming contact surface between sensor and liquid.

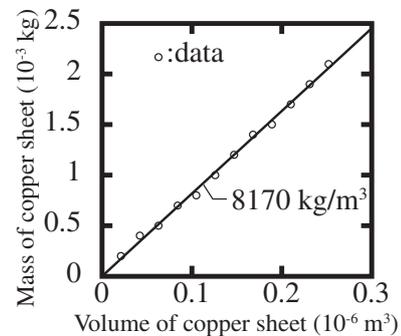
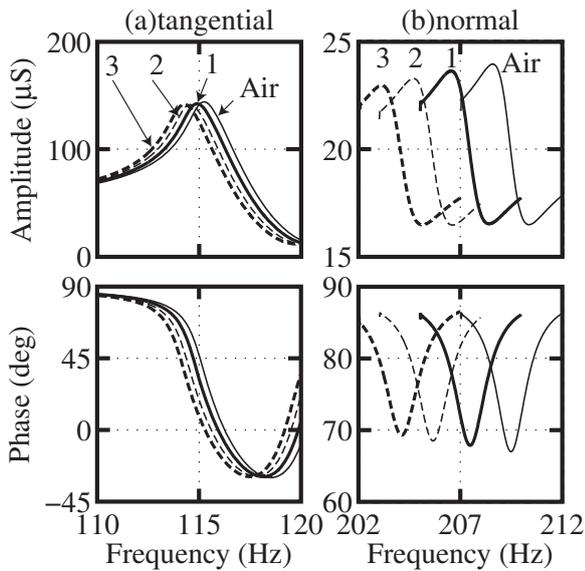


Fig. 5. Mass of copper sheet with changing volume of copper sheet. Density is slope of fitted line calculated by least-squares method using observed data.

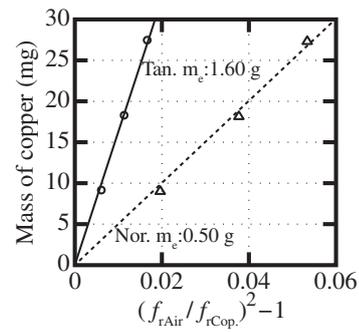
from the slope calculated by the least-squares method. Resonance characteristics of the sensor are observed using an impedance analyzer (Hewlett Packard 4194A) when one



**Fig. 6.** Admittance amplitude and phase characteristics around the resonance frequency without and with attachment of single, double, and triple pairs of sheets: (a) tangential and (b) normal vibration modes.

to three pairs of copper sheets, whose mass is  $4.6 \times 10^{-6}$  kg, are attached to both surfaces of the tip. Figure 6 shows the admittance amplitude and phase characteristics of each vibration mode around the resonance frequency without and with the attachment of single, double, or triple pairs of weights. As a result, the resonance frequency shift is caused by the mass loading of the sheet, while the damping of the sensor is almost unchanged. The resonance frequencies, at which the real part becomes the maximum, are obtained from the observed admittance. The equivalent mass in each vibration mode of the sensor is calculated from eqs. (8) and (9). We substitute the resonance frequencies,  $f_{rCOP}$  and  $f_{rLiq}$ , at which the copper sheet is attached, for the resonance frequency in the liquid of the tangential vibration,  $f_{rLiq}$ , expressed in eq. (8) and that in the liquid of normal vibration,  $f_{rLiq}$ , expressed in eq. (9). The equivalent masses  $m_{et}$  and  $m_{en}$  are obtained from the slope of each fitted line calculated by the least-squares method using the value of the additional copper mass and that of  $(f_{rAir}/f_{rCOP})^2 - 1$ . Figure 7 shows the experimental value obtained from eq. (8) or (9) and the fitted line. As a result, we found that both equivalent masses remain constant in the mass loading range in this experiment, and the vibration mode is not greatly changed because the fitted line corresponds to the experimental data. The values of equivalent masses in the tangential and normal vibrations  $m_{et}$  and  $m_{en}$  become  $1.60 \times 10^{-3}$  and  $0.50 \times 10^{-3}$  kg, respectively. These values are device-specific and are required to be measured only once.

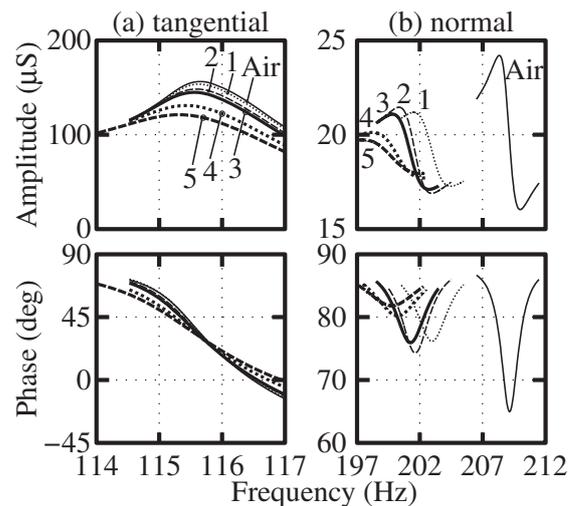
We compare the additional liquid masses  $m_{lt}$  and  $m_{ln}$  experimentally obtained using eqs. (8) and (9) with theoretical ones derived using eqs. (4) and (5) to ensure the validity of the measurement method. In this comparison, we use five standard liquids shown in Table I. The resonance characteristics in each vibration mode are observed using an impedance analyzer, as shown in Fig. 2. Figure 8 shows the admittance amplitude and phase characteristics of each



**Fig. 7.** Resonance frequency shift by changing mass of copper sheet attached to tip of the sensor. Equivalent masses of tangential and normal vibration modes are slopes of each fitted line calculated by least-squares method using observed data.

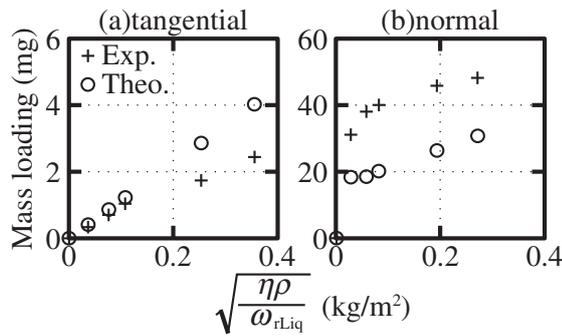
**Table I.** Viscosity and density of standard liquids.

	Liquid				
	1	2	3	4	5
Viscosity (mPa·s)	1.0	4.2	8.5	47	92
Density (kg/m <sup>3</sup> )	1000	920	940	960	968

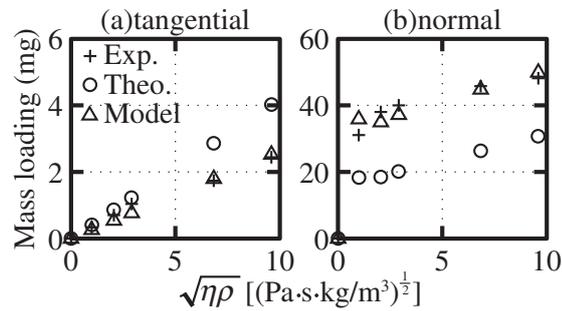


**Fig. 8.** Admittance amplitude and phase characteristics around the resonance frequency in air and five standard liquids shown in Table I: (a) tangential and (b) normal vibration modes.

vibration mode around the resonance frequencies in air and standard liquids. A resonance frequency shift is caused by the effect of additional liquid mass, and the damping of the sensor is increased by the effect of additional liquid damping. The mass loadings  $m_{lt}$  and  $m_{ln}$  are derived by substituting the resonance frequencies for eqs. (8) and (9). Figure 9 shows  $m_{lt}$  and  $m_{ln}$  versus the root of the product of viscosity and density divided by resonance angular frequencies. These experimental values are indicated by crosses, and the theoretical ones calculated using eqs. (4) and (5) by circles. In the tangential mode shown in Fig. 9(a), the experimental mass  $m_{lt}$  becomes smaller than the theoretical one. On the other hand, the experimental mass  $m_{ln}$  becomes larger than the theoretical one in the normal mode shown in Fig. 9(b). The theory concerning the tangential direction



**Fig. 9.** Additional liquid masses versus square root of product of viscosity and density divided by angular frequency in (a) tangential and (b) normal vibration modes.



**Fig. 10.** Additional liquid masses in devised model expressed by eqs. (12) and (13) versus square root of product of viscosity and density in (a) tangential and (b) normal vibration modes.

assumes the vibration of a plate with an infinite contact surface,<sup>27)</sup> and the theory concerning vibration in the normal direction assumes the vibration of a sphere. On the other hand, a rectangular plate with a finite contact surface actually vibrates in two directions. The difference between the experimental and theoretical  $m_{lt}$  and  $m_{ln}$  values arises owing to the rectangular edge and thickness of the plate. Thus, we devised a more generalized model. On the basis of eqs. (4) and (5), we introduce three independent coefficients  $C_1$ ,  $C_2$ , and  $C_3$ , as

$$m_{lt} = C_1 \sqrt{\frac{\eta\rho}{\omega}}, \quad (13)$$

$$m_{ln} = C_2 \sqrt{\frac{\eta\rho}{\omega}} + C_3\rho. \quad (14)$$

The values of the coefficients  $C_1$ ,  $C_2$ , and  $C_3$  are determined by the least-squares method using the experimental  $m_{lt}$  and  $m_{ln}$  values of five standard liquids indicated by crosses. The results obtained using model eqs. (13) and (14) are indicated by triangles in Fig. 10. As a result, we confirm the validity of the devised model because the masses indicated by triangles approximately correspond to the experimental ones indicated by crosses.

Next, as a practical example, the parameters  $\eta$  and  $\rho$  of food oil, whose viscosity and density are unknown, are measured using the determined coefficients. We observe the resonance frequencies by immersing the sensor in the oil and derive the values using eqs. (8), (9), (13), and (14). The results are shown in Table II. As a reference, the values are measured using a weight meter (Shimadzu EL-600) and a

**Table II.** Viscosities and densities of food oil derived using proposed and reference sensors.

	Viscosity (mPa·s)	Density (kg/m <sup>3</sup> )
Conventional	62.2	916
Proposed	69.2	926

viscometer (A&D SV-10). The density almost corresponds to the reference within a 1% error and the viscosity experimentally obtained is higher than that obtained using the viscometer within a 10% error. The reason considered is that the change in resonance frequency in the tangential vibration, as shown in Fig. 8(a), is very small considering the frequency resolution of the impedance analyzer. The derived product of  $\eta$  and  $\rho$  in the tangential vibration becomes larger than the reference value. In the normal vibration, however,  $\rho$  is more accurately derived because the body force from the liquid acting as the second term of the mass loading in eq. (14) significantly decreases the resonance frequency, as shown in Fig. 8(b). The value of  $\eta$  derived is larger than the actual one because the product of  $\eta$  and  $\rho$  derived is larger than the actual value.

#### 4. Conclusions

We developed a single sensor that can selectively oscillate at resonance frequency in two vibration modes to simultaneously measure the viscosity and density of liquids. We actually measured the viscosity and density of several samples using the proposed sensor.

We firstly obtained quantitative values of equivalent masses of the sensor in an experiment before measuring the liquid viscosity and density. As a result, we found that both equivalent masses remain constant in the mass loading range in this experiment, and the vibration mode is not greatly changed.

Next, we experimentally compared additional liquid masses with theoretical ones using five standard liquids to ensure the validity of the measurement method. From the results obtained, in the tangential mode, the experimental masses were evaluated to be smaller than the theoretical ones. On the other hand, the experimental masses were evaluated to be larger than the theoretical ones in the normal mode. The theory concerning the tangential direction assumes the vibration of a plate with an infinite contact surface, and the theory concerning the vibration in the normal direction assumes the vibration of a sphere. On the other hand, a rectangular plate with a finite contact surface vibrates actually in the two directions.

Thus, we devised a more generalized model of additional liquid mass. The model was calibrated using experimental data of the standard liquids. As a result, we confirmed the validity of the devised model.

Finally, we measured the viscosity and density of the food oil. From the value derived using the resonance frequencies of each mode in air and food oil, the density corresponds to the value measured using a weight meter with an error within 1% and the viscosity was evaluated to be higher than that obtained using a viscometer with an error within 10%. The errors are considered to be due to the change in resonance frequency in the tangential vibration being very

small considering the frequency resolution of the impedance analyzer. In the future, we plan to increase the area force from the liquid only in the tangential direction by changing the structure of the sensor. A more precise model of the liquid reaction around the contact surface should also be considered. Moreover, we will clarify the reason for the difference observed between theoretical and experimental additional liquid masses. Such a difference may be due to the thickness and edge of the plate. We will confirm the reason in detailed experiments.

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