

# Improvement of estimation method for physical properties of liquid using shear horizontal surface acoustic wave sensor response

メタデータ	言語: eng 出版者: 公開日: 2019-06-04 キーワード (Ja): キーワード (En): 作成者: Takayanagi, Masakatsu, Kondoh, Jun メールアドレス: 所属:
URL	<a href="http://hdl.handle.net/10297/00026651">http://hdl.handle.net/10297/00026651</a>

# **Improvement of estimation method for physical properties of liquid using shear horizontal surface acoustic wave sensor response**

Kazuya Takayanagi<sup>1</sup> and Jun Kondoh<sup>1, 2\*</sup>

<sup>1</sup> *Graduate School of Integrated Science and Technology, Shizuoka University, Hamamatsu 432-8561, Japan*

<sup>2</sup> *Graduate School of Science and Technology, Shizuoka University, Hamamatsu 432-8561, Japan*

\*E-mail: kondoh.jun@shizuoka.ac.jp

An estimation method for liquid properties, such as density and viscosity, using a shear horizontal surface acoustic wave sensor is proposed. The method is effective when the bulk modulus of a sample liquid is almost equal to that of water. When the bulk modulus of the sample liquid is different from that of water, the estimated results do not agree with literature values. In this paper, an estimation method for density, viscosity, and bulk modulus was proposed. When the liquid properties are estimated using the numerical simulation results, they agree with literature values. However, the results estimated using the measured results are significantly different from literature values. The reasons for this difference are discussed on the basis of liquid properties. It is necessary to select the optimum reference liquid for the measurements.

## 1. Introduction

Online measurements of liquid properties are required in many fields, such as those of automobiles, chemicals, pharmaceuticals, and petrochemicals. If continuous online measurements of physical properties of liquids are realized, quality control and productivity will be improved. Recently, acoustic wave sensors, such as Love wave sensors and quartz crystal microbalance sensors, have been studied to estimate the physical properties of liquids.<sup>1-4)</sup> A sensor based on an acoustic wave device can measure the viscosity and density of liquids. Acoustic wave sensors are also applied to various sensors, such as immunosensors and heavy metal sensors.<sup>5-19)</sup>

In this study, a shear horizontal surface acoustic wave (SH-SAW) sensor was used to measure the physical properties of liquids. The advantages of the SH-SAW sensor are the simultaneous detection of the electrical and mechanical properties of liquids.<sup>16)</sup> In addition, the SH-SAW sensor has high sensitivity, is small, can be mass produced and is inexpensive. We evaluated the physical properties of liquids by numerical analysis and using approximate equations derived by the perturbation method.<sup>20)</sup> Using the approximate equations, the physical properties of liquids can be obtained. However, as the accuracy is not sufficient, it is not suitable for practical use. Therefore, we focused on the inverse problem analysis to estimate the viscosity and density of liquids.<sup>21)</sup> In a previously reported method<sup>21)</sup>, the bulk modulus was assumed to have a constant value, which was the same as that of water. However, the bulk modulus depends on the kind and concentration of a liquid. Therefore, the bulk modulus cannot be assumed as constant and it is estimated with viscosity and density, simultaneously.

In this paper, the simultaneous estimation method for the bulk modulus, density, and viscosity is described. We improved a previously reported inverse problem analysis method<sup>21)</sup> to estimate the bulk modulus. We confirmed the superiority of the proposed estimation method by simulation. In addition, glycerol or ethanol aqueous solutions were measured using the SH-SAW sensor. The measured results were used for the proposed estimation method. The estimated results were compared with literature values.<sup>22, 23)</sup> However, no good agreements were obtained. We performed examinations using the material constant and found that the bulk modulus of the water was larger than those of other liquids. This means that water is not suitable as the reference liquid.

## 2. SH-SAW sensor

Figure 1 shows the SH-SAW sensor, which was fabricated on 36° YX-LiTaO<sub>3</sub>. It consists of input and output interdigital transducers (IDTs) and a propagation surface. When a high-frequency signal is applied to the input IDT, the SH-SAW is excited. The SH-SAW propagates on the surface and is reconverted to a high-frequency signal at the output IDT. When the liquid is loaded on the propagation surface, the phase and amplitude of the SH-SAW changes on the basis of the physical properties of the liquids. Therefore, the physical properties of liquids can be determined from output signals. In this study, the propagation surface was metallized with gold and titanium films for detection.

## 3. Measurement system

The measurement system is shown in Fig. 2. The sinusoidal signal from the signal generator was divided into two signals. One was input to the SH-SAW sensor and the other was used as the reference signal of the vector voltmeter. The phase difference and the amplitude ratio of the output signal from the SH-SAW sensor to the reference signal were obtained with a vector voltmeter. For the measurements, water was used as the reference liquid. The phase difference and amplitude ratio of the reference liquid to the sample liquid were measured using the network analyzer. The velocity change  $\Delta V/V$  and the attenuation change  $\Delta\alpha/k$  normalized by wave number were calculated from the phase difference and amplitude ratio, respectively. In the measurements, distilled water was used as the reference.

## 4. Direct problem analysis method

Moriizumi et al. proposed a numerical analysis method for SAW for a liquid/piezoelectric substrate structure.<sup>24)</sup> In this paper, it is called the direct problem analysis method. The method is based on the SAW analysis methods proposed by Campbell and Jones<sup>25)</sup> and Yamanouchi and Shibayama.<sup>26)</sup> The details of the method are reported written in Ref. 21).

## 5. Inverse problem analysis method

In the inverse problem analysis method, the physical properties of the liquid are obtained from  $\Delta V/V$  and  $\Delta\alpha/k$ , which are measured by the SH-SAW sensor. In previous study, it was impossible to simultaneously determine three parameters, namely, the density  $\rho$ , the viscosity  $\eta$ , and the bulk modulus  $K$ , from the two parameters  $\Delta V/V$  and  $\Delta\alpha/k$ . Therefore, the bulk modulus was assumed to be constant. However, the bulk modulus also depends on the kind and concentration of a liquid. The bulk modulus affects the propagation characteristics of SH-SAW. Figure 3 shows the results calculated using the direct problem analysis method. The frequency was 51.5 MHz. The physical properties, viscosity and density, of the glycerol aqueous solution were obtained from the literature.<sup>22)</sup> Only the bulk modulus was fixed at that of water or glycerol. The effect of the bulk modulus appears as an attenuation change. The results indicate that the bulk modulus cannot be assumed as the constant value and is estimated with viscosity and density.

Figure 4 shows a flow chart of the inverse problem method proposed in this study. The initial value of the bulk modulus  $K$  was set as that of water. Then,  $\Delta V/V$  and  $\Delta\alpha/k$  were calculated by changing  $K$  as functions of  $\rho$  and  $\eta$ . The calculation was based on the direct problem analysis method. Figure 5 shows the examples of  $\Delta V/V$  and  $\Delta\alpha/k$  as functions of  $\rho$  and  $\eta$ . The same figures were obtained for each  $K$ . From the results, the approximate expressions of  $\Delta V/V$  and  $\Delta\alpha/k$  are calculated using the least squares method.<sup>27)</sup> The measured values of  $\Delta V/V$  and  $\Delta\alpha/k$  were substituted into the obtained equations to derive  $\rho$  and  $\eta$ . The process of determining  $\rho$  and  $\eta$  is the same as that in a previous method.<sup>21)</sup>

The bulk modulus normally increases with increasing  $\rho$  and  $\eta$ . In the proposed inverse problem analysis method, we used this relationship. The details of the method to determine the liquid properties are as follow.

- (1)  $\rho'$  was determined by taking the average of  $\rho$ , which was determined using the bulk modulus of  $K$ , and the density of the water  $\rho_w$ .  $\eta'$  was also determined from the average of  $\eta$  and  $\eta_w$ . Here,  $\eta_w$  is the viscosity of water.
- (2) To obtain  $\Delta V/V$  and  $\Delta\alpha/k$ ,  $\rho$  and  $\eta$  were fixed and only  $K$  was varied (see Fig. 6).
- (3) A new  $K$  was determined, which minimized the difference between the measured value and the plotted point using the following equation (see Fig. 7)

$$D = \left| \left[ \left( \frac{\Delta V}{V} \right) - \left( \frac{\Delta V}{V} \right)_m \right] / \left( \frac{\Delta V}{V} \right)_m \right| + \left| \left[ \left( \frac{\Delta \alpha}{k} \right) - \left( \frac{\Delta \alpha}{k} \right)_m \right] / \left( \frac{\Delta \alpha}{k} \right)_m \right|. \quad (1)$$

Approximate equations were formulated for deriving  $\rho$  and  $\eta$ .<sup>21)</sup> For obtaining  $K$ , however,

an approximate equation is not necessary. After obtaining the plotted points on Fig. 6 with the required step size, we compared  $D$  at each point. If  $\rho$ ,  $\eta$ ,  $\Delta V/V$ , and  $\Delta\alpha/k$  are known,  $K$  can be accurately determined by using the required step size. The bulk modulus determined by fixing  $\rho'$  and  $\eta$  is called  $K1$ , and that obtained by fixing  $\rho$  and  $\eta'$  is called  $K2$ . Then, the average of  $K1$  and  $K2$  is defined as the new  $K$  and used in the next loop. Also,  $K1$  obtained in the first loop, when  $K$  is the same as that of water, is called  $K1_1$ . In the second and subsequent loops,  $\rho'$  and  $\eta'$  in the preceding loop are used instead of density and viscosity of water on average to calculate new  $\rho'$  and  $\eta'$ . The loop was repeated until the optimum values of  $\rho$ ,  $\eta$ , and  $K$  were estimated.

## 6. Results and discussion

### 6.1 Estimation of physical properties of glycerol aqueous solutions

The physical properties of 5, 10, 30, and 50 wt% glycerol aqueous solutions were estimated. The temperature of the liquids was fixed at 20 °C. The physical properties were estimated using  $\Delta V/V$  and  $\Delta\alpha/k$  obtained by the direct problem analysis method using literature values or measurement values. The bulk modulus of literature values was calculated as

$$K = \rho V^2, \quad (2)$$

where  $V$  is the sound velocity of a liquid. The bulk modulus was also compared with that obtained the previous estimation method. Table I shows the literature values and the results estimated using  $\Delta V/V$  and  $\Delta\alpha/k$  derived by the direct problem analysis method. Table II shows the literature values and the estimated results using  $\Delta V/V$  and  $\Delta\alpha/k$  obtained from the measurements.

The literature values and estimated results using the previous method and the proposed method using  $K$  or  $K1_1$  are summarized in Table I. The estimated results using the previous estimation method do not agree with the literature values. For the improved method, the bulk modulus approaches the actual value and the accuracy of the estimated results is improved. Especially when the bulk modulus is large, it has improved significantly. It is important to consider the bulk modulus and the improved method is effective. The precisions of the estimated results using  $K1_1$  are higher than those estimated using  $K$ , which is the final estimated result in the proposed method. In the estimation, the averages of the viscosity and density were used to obtain  $K$ . However, the difference between the viscosity

of the water and that of the sample is large. We considered that averaging was not applied for these cases.

For the estimated results using the measured values, their differences from the literature value are large. The estimated results using the proposed method do not agree with the literature values. We prepared the sample liquids for measurements ourselves. However, their concentrations were not confirmed by other methods, such as measurements using a density meter or an optical sensor. It is necessary to measure the concentration of the samples used.

## 6.2 Estimation of physical properties of ethanol aqueous solutions

The physical properties of 5, 10, 30, and 50 wt% ethanol aqueous solution were estimated. The liquid temperature was 20 °C. The physical properties were estimated using  $\Delta V/V$  and  $\Delta\alpha/k$  obtained by the direct problem analysis method. The bulk modulus of literature values was obtained from Eq. (2). The estimated results and the literature values are summarized in Table III. Also, the relationship between the ethanol concentration and the bulk modulus is plotted in Fig. 8.

From the table and figure, the bulk modulus estimated  $K$  better than  $K_{11}$ . Table IV and Fig. 9 show the relationship between the bulk modulus and viscosity or density for various solutions. For the ethanol aqueous solutions, their densities are lower than that of water. On the other hand, their viscosities are higher than that of water between 0 and 50 wt%. In these cases, the estimation of the bulk modulus is more difficult than that of glycerol aqueous solutions. The estimated  $K$  reflects the change of the bulk modulus. However, since  $K_{11}$  is obtained by considering only density, it is different from the actual value. Therefore, for the ethanol aqueous solutions, the estimated  $K$  must be used to determine their physical properties.

## 6.3 Bulk modulus of liquids

When a liquid is measured using an acoustic wave-based sensor, water is normally used as the reference solution. Here, we compared physical properties of liquids. The sound velocity, density, viscosity, and bulk modulus of liquids are summarized in Table V.<sup>22, 28-32)</sup> Figures 10(a) and 10(b) show the relationship between the density and the bulk modulus, or the viscosity and the bulk modulus from Table V. In the estimation method in this paper, the bulk modulus is increased or decreased by comparing the density and viscosity of the

reference liquid. However, the bulk modulus of water is larger than those of other liquids. This means that water is not an optimum reference liquid for estimating the physical properties of liquids. It is necessary to find the optimum reference liquid for such estimations.

## 7. Conclusions

In this study, the estimation method for three physical properties of liquids was proposed. For the glycerol aqueous solutions, compared with the results estimated using  $K$ , those estimated using  $K_{11}$  show good agreements with literature values. On the other hand, for the ethanol aqueous solutions, the results estimated using  $K$  agree with literature values. This finding indicates that it is necessary to choose  $K_{11}$  or  $K$ . The results estimated based on the basis of both the proposed and previous methods do not agree with literature values. As good reproducibility of the experimental results was confirmed, we concluded that the concentrations of the liquid samples prepared were not correct. Measurements of the concentrations by other methods are required.

In this study, water was used as the reference liquid. The bulk modulus of water, however, is larger than that of the other solutions. This large difference in the bulk modulus negatively affects the accuracy of estimating the physical properties of liquid. Therefore, the optimum reference liquid must be selected in conformity with the target liquid.



## References

- 1) P. Kielczyński, M. Szalewski, and A. Balcerzak, *Int. J. Solids Struct.* **49**, 2314 (2012).
- 2) P. Kielczyński, M. Szalewski, and A. Balcerzak, *J. Appl. Phys.* **116**, 044902 (2014).
- 3) C. Riesch, E. K. Reichel, F. Keplinger, and B. Jakoby, *J. Sens.* **2008**, 697062 (2008).
- 4) N. Doy, G. McHale, M. I. Newton, C. Hardacre, R. Ge, R. W. Allen, and J. M. MacInnes, *IEEE Sensors Conf.*, 2009, p.287.
- 5) A. Tretjakov, V. Syritski J. Reut R. Boroznjak, and A. Öpik, *Anal. Chim. Acta* **902**, 182 (2016).
- 6) Z. Ramshani, A. S. G. Reddy, B. B. Narakathu, J. T. Wabeke, S. O. Obare, and M. Z. Atashbar, *Sens. Actuators B* **217**, 72 (2015).
- 7) K. Kano, T. Kogai, N. Yoshimura, H. Yatsuda, J. Kondoh, and S. Shiokawa, *Jpn. J. Appl. Phys.* **51**, 07GC20 (2012).
- 8) S. Hohmann, S. Kögel, Y. Brunner, B. Schmieg, C. Ewald, F. Kirschhöfer, G. Brenner-Weiß, and K. Länge, *Sensors* **15**, 11873 (2015).
- 9) C. Viespe and D. Miu, *Sensors* **17**, 1529 (2017).
- 10) X. Zhang Y. Zou, C. An, K. Ying, X. Chen, and P. Wang, *Sens. Actuators B* **217**, 100 (2015).
- 11) K. Tada, T. Nozawa, and J. Kondoh, *Jpn. J. Appl. Phys.* **56**, 07JD15 (2017).
- 12) S. Endo, J. Kondoh, K. Sato, and N. Sawada, *Jpn. J. Appl. Phys.* **51**, 07GC19 (2012).
- 13) T. Kogai, H. Yatsuda, and J. Kondoh, *Jpn. J. Appl. Phys.* **56**, 07JD09 (2017).
- 14) K. Länge, B. Rapp, and M. Rapp, *Anal. Bioanal. Chem.* **391**, 1509 (2008).
- 15) H. Wu, X. Xiong, H. Zu, J. Wang, and Q. Wang, *J. Appl. Phys.* **121**, 054501 (2017).
- 16) J. Kondoh, *Electron. Commun. Jpn.* **131**, 1094 (2011).
- 17) J. Kondoh, Y. Okiyama, S. Mikuni, Y. Matsui, M. Nara, T. Mori, and H. Yatsuda, *Jpn. J. Appl. Phys* **47**, 5, 4065 (2008).
- 18) G. Sehra, M. Cole, and J.W. Gardner, *Sens. Actuators B* **103**, 233 (2004).
- 19) J. Vivancos, Z. Rácz, M. Cole, and J. W. Gardner, *Sens. Actuators B.* **171–172**, 469 (2012).
- 20) J. Kondoh and S. Shiokawa, *Electron. Commun. Jpn. Part 2* **76**, 69 (1993).
- 21) K. Ueda and J. Kondoh, *Jpn. J. Appl. Phys.* **56**, 07JD08 (2017).

- 22) D. R. Lide, *CRC Handbook of Chemistry and Physics* (CRC Press, Boca Raton, FL, 2005) 86th ed., Sect. 8.
- 23) S. G. Bruun, P. G. Sørensen, and A. Hvidt, *Acta Chem. Scand. A* **28**, 1047 (1974).
- 24) T. Moriizumi, Y. Unno, and S. Shiokawa, *Proc. IEEE Ultrasonics Symp.*, 1987, p. 579.
- 25) J. J. Campbell and W. R. Jones, *IEEE Trans. Sonics Ultrason.* **15**, 209 (1968).
- 26) K. Yamanouchi and M. Shibayama, *J. Appl. Phys.* **43**, 856 (1972).
- 27) K. Madsen, H. B. Nielsen, and O. Tingleff, *Methods for Non-Linear Least Squares Problems* (Informatics and Mathematical Modelling, Kongens Lyngby, 2004) 2nd ed., p. 24.
- 28) J. Saneyosi, Y. Kikuchi, and O. Nomoto, *Choonpa Gijyutsu Binran* (Ultrasound Technology Handbook), (Nikkan Kogyo Shinbun, Tokyo, 1985) 5<sup>th</sup> ed., p. 1205 [in Japanese].
- 29) [http://www.fujielectric.co.jp/products/instruments/QA/box/pdf/portaflowX\\_3.pdf](http://www.fujielectric.co.jp/products/instruments/QA/box/pdf/portaflowX_3.pdf)
- 30) <http://www.chiyoda-electric.co.jp/%E9%9F%B3%E3%81%AE%E7%89%B9%E6%80%A7%E3%81%A8%E6%B3%A2%E9%95%B7>
- 31) <http://www.geocities.jp/jr2bvb/jr2bvb/kagaku/nendo.htm>
- 32) <http://www.siyaku.com/uh/Myp.do?now=1509632748417>

## Figure Captions

**Fig. 1.** (Color online) Schematic illustration of the SH-SAW sensor used.

**Fig. 2.** (Color online) Measurement system in this study.

**Fig. 3.** (Color online) Numerical calculated results of the effect of the bulk modulus. (a)  $\Delta V/V$  and (b)  $\Delta\alpha/k$ .

**Fig. 4.** Flow chart of the inverse problem analysis method.

**Fig. 5.** (Color online) Scatter diagram obtained by changing  $\rho$  and  $\eta$ . (a)  $\Delta V/V$  and (b)  $\Delta\alpha/k$ .

**Fig. 6.** (Color online) Scatter diagram obtained by changing  $K$ . (a)  $\Delta V/V$  and (b)  $\Delta\alpha/k$ .

**Fig. 7.** (Color online) Determination of  $K$  that minimizes the difference between measured and plotted values.

**Fig. 8.** (Color online) Relationship between the mass concentration and the bulk modulus of ethanol aqueous solutions.

**Fig. 9.** (Color online) Relationship between the bulk modulus and the density or viscosity of the ethanol aqueous solutions. (a) Density and (b) viscosity.

**Fig. 10.** (Color online) Relationship between the bulk modulus and the density or viscosity of various solutions shown in Table V. (a) Density and (b) viscosity.

**Table I.** Results estimated using previous method and proposed methods in this paper for glycerol aqueous solutions. (a) Density, (b) viscosity, and (c) bulk modulus.

(a)

	5 wt%	10 wt%	30 wt%	50 wt%
(i) Literature value	$1.010 \times 10^3$	$1.022 \times 10^3$	$1.072 \times 10^3$	$1.125 \times 10^3$
(ii) Estimated using previous method	$1.066 \times 10^3$	$1.138 \times 10^3$	$1.467 \times 10^3$	$1.861 \times 10^3$
Difference between (i) and (ii)	5.57%	11.44%	36.93%	65.40%
(iii) Estimated value using $K$	$1.015 \times 10^3$	$1.033 \times 10^3$	$1.085 \times 10^3$	$1.053 \times 10^3$
Difference between (i) and (iii)	0.56%	1.10%	1.21%	-6.46%
(iv) Estimated value using $K1_1$	$1.011 \times 10^3$	$1.026 \times 10^3$	$1.075 \times 10^3$	$1.070 \times 10^3$
Difference between (i) and (iv)	0.14%	0.39%	0.31%	-4.93%

(b)

	5 wt%	10 wt%	30 wt%	50 wt%
(i) Literature value	$1.127 \times 10^{-3}$	$1.291 \times 10^{-3}$	$2.458 \times 10^{-3}$	$6.040 \times 10^{-3}$
(ii) Fixed bulk modulus in previous method	$1.066 \times 10^{-3}$	$1.157 \times 10^{-3}$	$1.786 \times 10^{-3}$	$3.630 \times 10^{-3}$
Difference between (i) and (ii)	-5.40%	-10.41%	-27.35%	-39.90%
(iii) Estimated value using $K$	$1.120 \times 10^{-3}$	$1.276 \times 10^{-3}$	$2.439 \times 10^{-3}$	$6.470 \times 10^{-3}$
Difference between (i) and (iii)	-0.63%	-1.13%	-0.75%	7.13%
(iv) Estimated value using $K1_1$	$1.125 \times 10^{-3}$	$1.286 \times 10^{-3}$	$2.462 \times 10^{-3}$	$6.364 \times 10^{-3}$
Difference between (i) and (iv)	-0.20%	-0.42%	0.17%	5.38%

(c)

	5 wt%	10 wt%	30 wt%	50 wt%
(i) Literature value	$2.286 \times 10^9$	$2.380 \times 10^9$	$2.791 \times 10^9$	$3.258 \times 10^9$
(ii) Fixed bulk modulus in previous method	$2.196 \times 10^9$	$2.196 \times 10^9$	$2.196 \times 10^9$	$2.196 \times 10^9$
Difference between (i) and (ii)	-3.92%	-7.73%	-21.33%	-32.59%
(iii) Estimated value using $K$	$2.276 \times 10^9$	$2.361 \times 10^9$	$2.768 \times 10^9$	$3.397 \times 10^9$
Difference between (i) and (iii)	-0.42%	-0.80%	-0.84%	4.26%
(iv) Estimated value using $K1_1$	$2.283 \times 10^9$	$2.373 \times 10^9$	$2.785 \times 10^9$	$3.363 \times 10^9$
Difference between (i) and (iv)	-0.12%	-0.30%	-0.23%	3.24%

**Table II.** Results estimated using the measured results for glycerol aqueous solutions. (a) Density, (b) viscosity, and (c) bulk modulus.

(a)

	5 wt%	10 wt%	30 wt%	50 wt%
(i) Literature value	$1.010 \times 10^3$	$1.022 \times 10^3$	$1.072 \times 10^3$	$1.125 \times 10^3$
(ii) Estimated using previous method	$9.219 \times 10^2$	$1.022 \times 10^3$	$1.199 \times 10^3$	$1.612 \times 10^3$
Difference between (i) and (ii)	-8.691%	0.093%	11.868%	43.277%
(iii) Estimated value using $K$	$9.022 \times 10^2$	$9.304 \times 10^2$	$8.609 \times 10^2$	$8.965 \times 10^2$
Difference between (i) and (iii)	-10.65%	-8.92%	-19.67%	-20.34%
(iv) Estimated value using $K1_1$	$1.011 \times 10^3$	$1.002 \times 10^3$	$9.972 \times 10^2$	$9.877 \times 10^2$
Difference between (i) and (iv)	0.14%	-1.93%	-6.95%	-12.24%

(b)

	5 wt%	10 wt%	30 wt%	50 wt%
(i) Literature value	$1.127 \times 10^{-3}$	$1.291 \times 10^{-3}$	$2.458 \times 10^{-3}$	$6.040 \times 10^{-3}$
(ii) Estimated using previous method	$1.389 \times 10^{-3}$	$1.508 \times 10^{-3}$	$2.952 \times 10^{-3}$	$5.050 \times 10^{-3}$
Difference between (i) and (ii)	23.24%	16.78%	20.10%	-16.38%
(iii) Estimated value using $K$	$1.420 \times 10^{-3}$	$1.659 \times 10^{-3}$	$4.155 \times 10^{-3}$	$9.113 \times 10^{-3}$
Difference between (i) and (iii)	25.98%	28.52%	69.05%	50.88%
(iv) Estimated value using $K1_1$	$1.296 \times 10^{-3}$	$1.539 \times 10^{-3}$	$3.568 \times 10^{-3}$	$8.260 \times 10^{-3}$
Difference between (i) and (iv)	14.99%	19.22%	45.14%	36.77%

(c)

	5 wt%	10 wt%	30 wt%	50 wt%
(i) Literature value	$2.286 \times 10^9$	$2.380 \times 10^9$	$2.791 \times 10^9$	$3.258 \times 10^9$
(ii) Fixed bulk modulus in previous method	$2.196 \times 10^9$	$2.196 \times 10^9$	$2.196 \times 10^9$	$2.196 \times 10^9$
Difference between (i) and (ii)	-3.92%	-7.73%	-21.33%	-32.59%
(iii) Estimated value using $K$	$2.230 \times 10^9$	$2.350 \times 10^9$	$2.770 \times 10^9$	$3.334 \times 10^9$
Difference between (i) and (iii)	-2.44%	-1.26%	-0.78%	2.33%
(iv) Estimated value using $K1_1$	$2.089 \times 10^9$	$2.229 \times 10^9$	$2.514 \times 10^9$	$3.150 \times 10^9$
Difference between (i) and (iv)	-8.60%	-6.35%	-9.94%	-3.30%

**Table III.** Estimation using the measured value of ethanol aqueous solution. (a) Density,

(b) viscosity, and (c) bulk modulus.

(a)

	5 wt%	10 wt%	30 wt%	50 wt%
(i) Literature value	$9.893 \times 10^2$	$9.819 \times 10^2$	$9.539 \times 10^2$	$9.139 \times 10^2$
(ii) Estimated using previous method	$9.693 \times 10^2$	$9.820 \times 10^2$	$9.24 \times 10^2$	$9.013 \times 10^2$
Difference between (i) and (ii)	-2.02%	0.01%	-3.14%	-1.38%
(iii) Estimated value using $K$	$9.456 \times 10^2$	$9.217 \times 10^2$	$7.600 \times 10^2$	$8.890 \times 10^2$
Difference between (i) and (iii)	-4.42%	-6.13%	-20.32%	-2.73%
(iv) Estimated value using $K1_1$	$9.961 \times 10^2$	$9.991 \times 10^2$	$1.003 \times 10^3$	$1.006 \times 10^3$
Difference between (i) and (iv)	0.69%	1.75%	5.11%	10.11%

(b)

	5 wt%	10 wt%	30 wt%	50 wt%
(i) Literature value	$1.228 \times 10^{-3}$	$1.501 \times 10^{-3}$	$2.667 \times 10^{-3}$	$2.813 \times 10^{-3}$
(ii) Estimated using previous method	$1.304 \times 10^{-3}$	$1.503 \times 10^{-3}$	$3.183 \times 10^{-3}$	$3.324 \times 10^{-3}$
Difference between (i) and (ii)	6.16%	0.15%	19.36%	18.16%
(iii) Estimated value using $K$	$1.337 \times 10^{-3}$	$1.603 \times 10^{-3}$	$3.884 \times 10^{-3}$	$3.371 \times 10^{-3}$
Difference between (i) and (iii)	8.86%	6.81%	45.64%	19.82%
(iv) Estimated value using $K1_1$	$1.268 \times 10^{-3}$	$1.477 \times 10^{-3}$	$2.930 \times 10^{-3}$	$2.972 \times 10^{-3}$
Difference between (i) and (iv)	3.25%	2.44%	9.86%	5.67%

(c)

	5 wt%	10 wt%	30 wt%	50 wt%
(i) Literature value	$2.325 \times 10^9$	$2.408 \times 10^9$	$2.454 \times 10^9$	$2.005 \times 10^9$
(ii) Fixed bulk modulus in previous method	$2.196 \times 10^9$	$2.196 \times 10^9$	$2.196 \times 10^9$	$2.196 \times 10^9$
Difference between (i) and (ii)	-5.55%	-8.81%	-10.53%	9.55%
(iii) Estimated value using $K$	$2.234 \times 10^9$	$2.295 \times 10^9$	$2.494 \times 10^9$	$2.238 \times 10^9$
Difference between (i) and (iii)	-3.93%	-4.69%	1.62%	11.65%
(iv) Estimated value using $K1_1$	$2.152 \times 10^9$	$2.168 \times 10^9$	$2.069 \times 10^9$	$2.027 \times 10^9$
Difference between (i) and (iv)	-7.44%	-9.97%	-15.70%	1.12%

**Table IV.** Physical properties of ethanol aqueous solution.

Mole fraction	Concentration (wt%)	Velocity (m/s)	Density (kg/m <sup>3</sup> )	Viscosity (Pa s)	Bulk modulus (Pa)
1	100	1144	$7.852 \times 10^2$	$1.030 \times 10^{-3}$	$1.028 \times 10^9$
0.78	90	1221	$8.140 \times 10^2$		$1.214 \times 10^9$
0.61	80	1284	$8.392 \times 10^2$		$1.383 \times 10^9$
0.48	70	1345	$8.634 \times 10^2$		$1.562 \times 10^9$
0.37	60	1416	$8.901 \times 10^2$	$2.140 \times 10^{-3}$	$1.785 \times 10^9$
0.28	50	1481	$9.098 \times 10^2$	$2.250 \times 10^{-3}$	$1.996 \times 10^9$
0.21	40	1549	$9.325 \times 10^2$		$2.237 \times 10^9$
0.14	29	1604	$9.509 \times 10^2$	$2.100 \times 10^{-3}$	$2.446 \times 10^9$
0.12	26	1619	$9.597 \times 10^2$		$2.515 \times 10^9$
0.09	20	1615	$9.666 \times 10^2$		$2.521 \times 10^9$
0.06	14	1596	$9.735 \times 10^2$		$2.480 \times 10^9$
0.04	10	1566	$9.805 \times 10^2$	$1.270 \times 10^{-3}$	$2.405 \times 10^9$
0.02	5	1533	$9.881 \times 10^2$		$2.322 \times 10^9$
0	0	1497	$9.971 \times 10^2$	$8.900 \times 10^{-4}$	$2.234 \times 10^9$



**Table V.** Physical properties of various liquids<sup>22, 28-32</sup>.

	Sound velocity (m/s)	Density (kg/m <sup>3</sup> )	Viscosity (Pa s)	Bulk modulus (Pa)
Carbon tetrachloride	$9.380 \times 10^2$	$1.594 \times 10^3$	$9.690 \times 10^{-4}$	$1.403 \times 10^9$
Chloroform	$1.001 \times 10^3$	$1.487 \times 10^3$	$5.700 \times 10^{-4}$	$1.490 \times 10^9$
Ether	$1.006 \times 10^3$	$7.135 \times 10^2$	$2.400 \times 10^{-4}$	$7.221 \times 10^8$
N-pentane	$1.032 \times 10^3$	$6.260 \times 10^2$	$2.340 \times 10^{-4}$	$6.667 \times 10^8$
N-hexane	$1.083 \times 10^3$	$6.540 \times 10^2$	$3.260 \times 10^{-4}$	$7.671 \times 10^8$
Methanol	$1.120 \times 10^3$	$7.910 \times 10^2$	$5.970 \times 10^{-4}$	$9.922 \times 10^8$
Acetic acid	$1.159 \times 10^3$	$1.050 \times 10^3$	$1.220 \times 10^{-3}$	$1.410 \times 10^9$
Ethyl acetate	$1.164 \times 10^3$	$9.000 \times 10^2$	$4.550 \times 10^{-4}$	$1.219 \times 10^9$
Ethanol	$1.168 \times 10^3$	$7.892 \times 10^2$	$1.200 \times 10^{-3}$	$1.077 \times 10^9$
IPA	$1.170 \times 10^3$	$7.809 \times 10^2$	$2.000 \times 10^{-3}$	$1.069 \times 10^9$
Methyl acetate	$1.181 \times 10^3$	$9.280 \times 10^2$	$3.810 \times 10^{-4}$	$1.294 \times 10^9$
Acetone	$1.190 \times 10^3$	$7.905 \times 10^2$	$3.220 \times 10^{-4}$	$1.119 \times 10^9$
Toluene	$1.327 \times 10^3$	$8.640 \times 10^2$	$5.860 \times 10^{-4}$	$1.522 \times 10^9$
Carbon disulfide	$1.451 \times 10^3$	$1.263 \times 10^3$	$3.660 \times 10^{-4}$	$2.660 \times 10^9$
Nitrobenzene	$1.473 \times 10^3$	$1.207 \times 10^3$	$2.010 \times 10^{-3}$	$2.619 \times 10^9$
Water	$1.496 \times 10^3$	$9.980 \times 10^2$	$1.010 \times 10^{-3}$	$2.234 \times 10^9$
Aniline	$1.659 \times 10^3$	$1.022 \times 10^3$	$4.400 \times 10^{-3}$	$2.812 \times 10^9$
Ethylene glycol	$1.666 \times 10^3$	$1.113 \times 10^3$	$1.610 \times 10^{-2}$	$3.089 \times 10^9$
Glycerol	$1.923 \times 10^3$	$1.261 \times 10^3$	$7.805 \times 10^{-1}$	$4.664 \times 10^9$

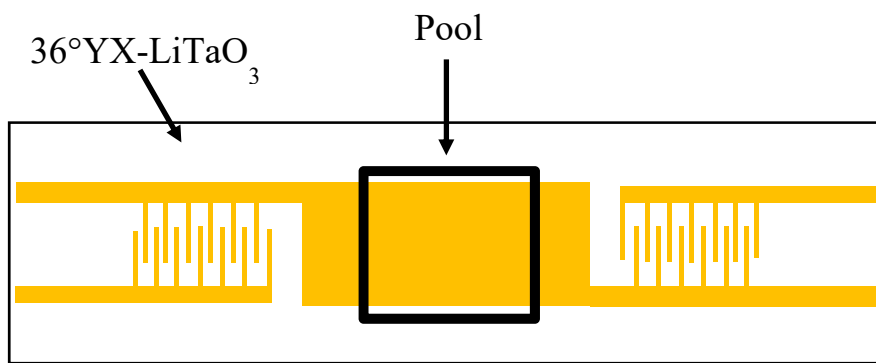


Fig. 1. (Color Online)

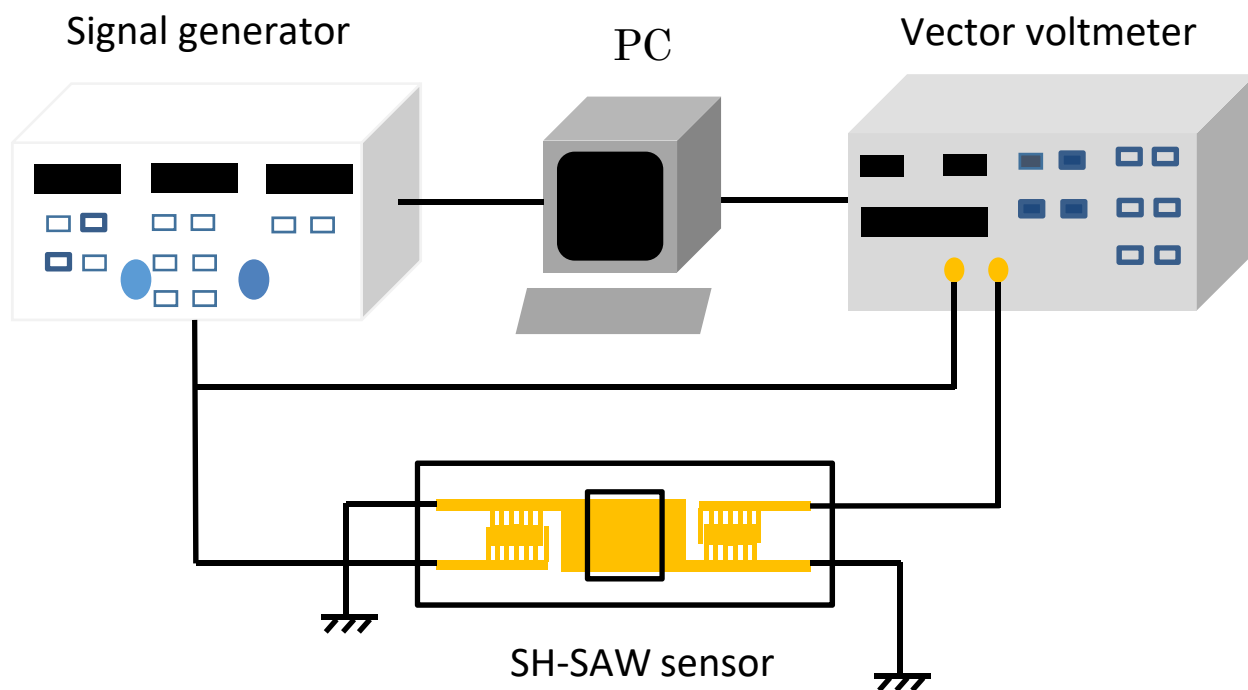
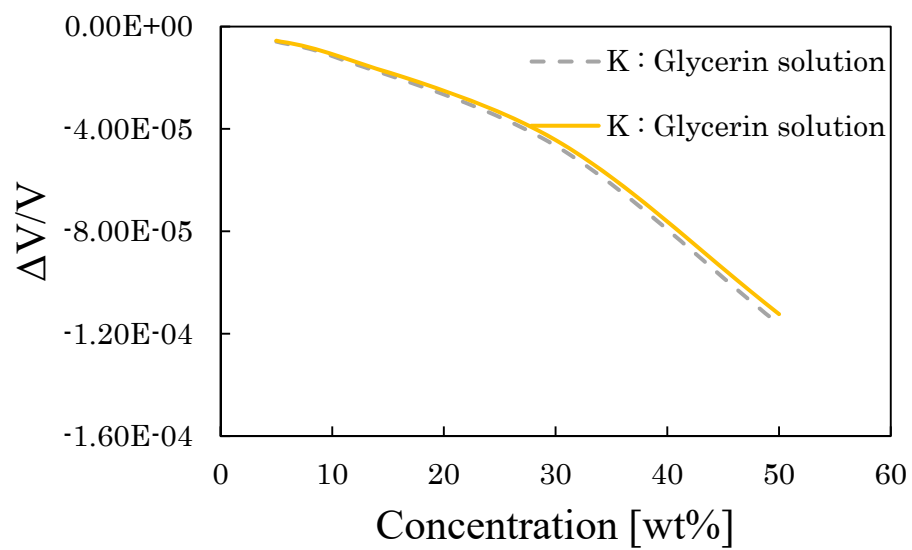
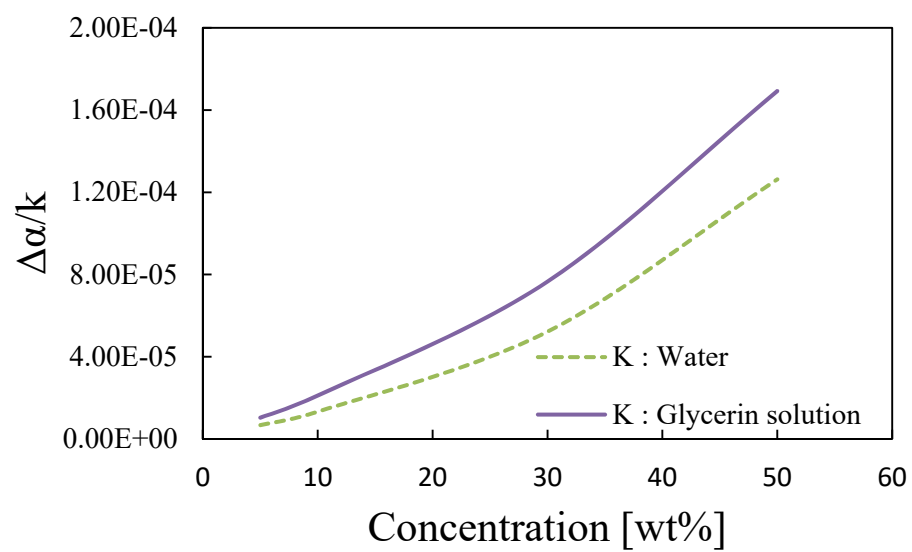


Fig. 2. (Color Online)



(a)



(b)

Fig. 3. (Color Online)

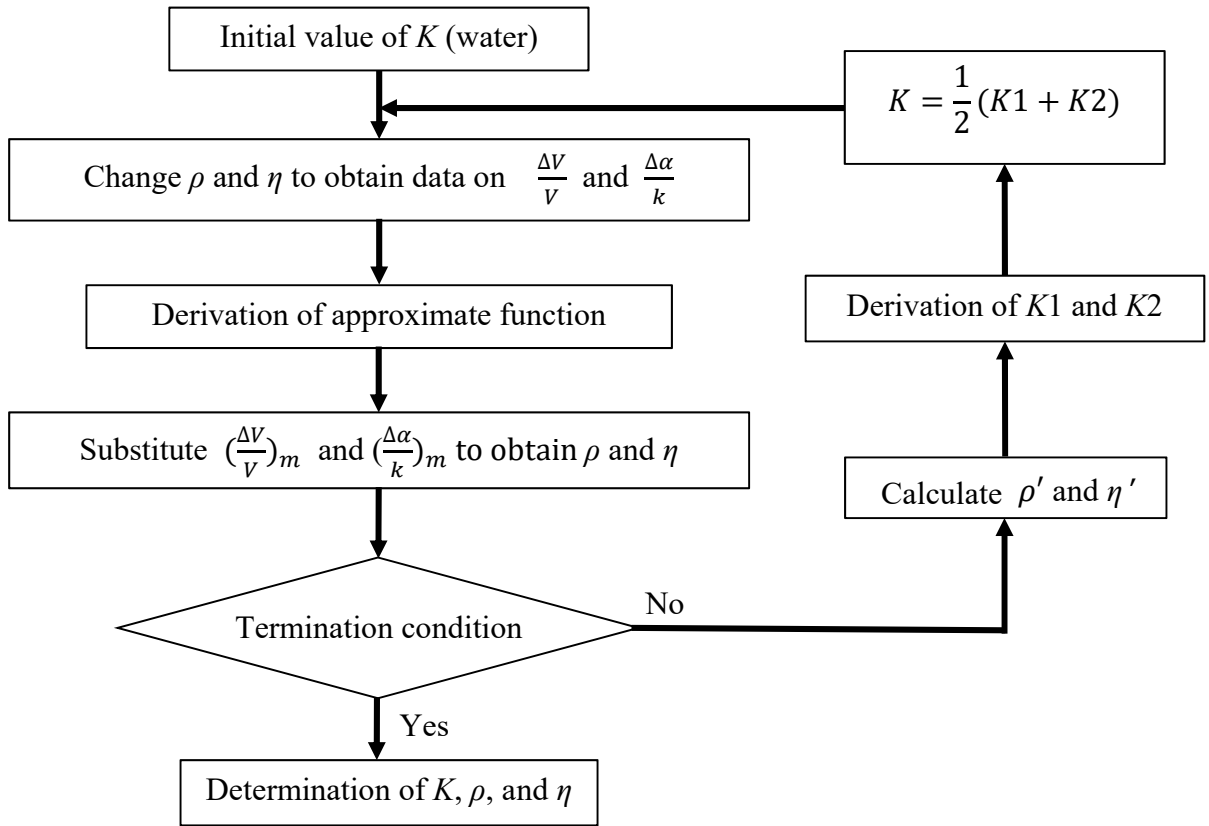
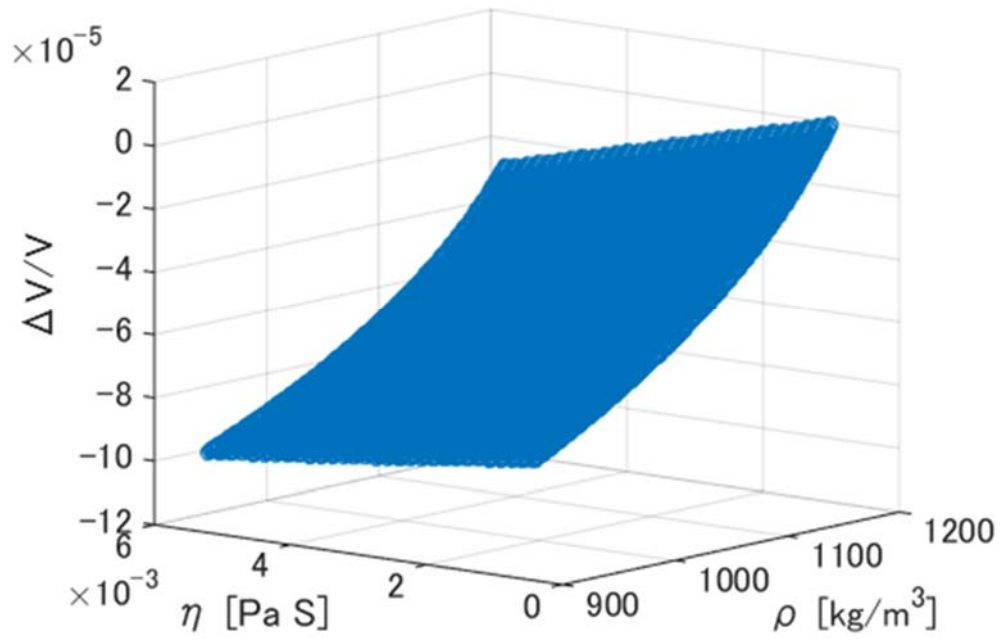
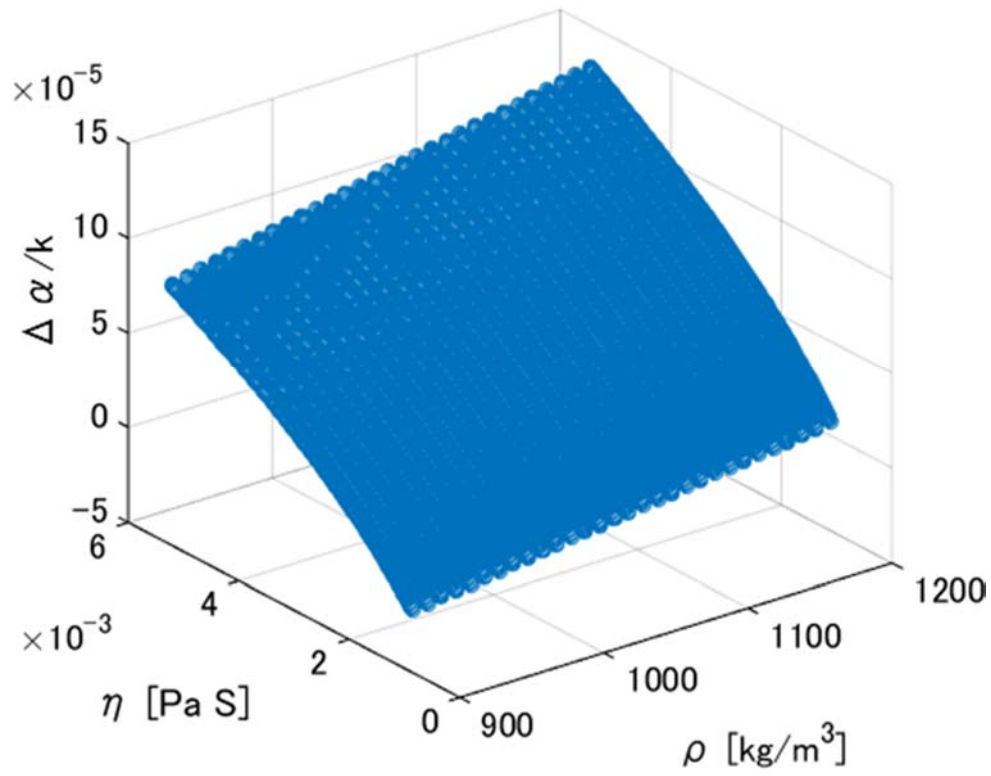


Fig. 4.

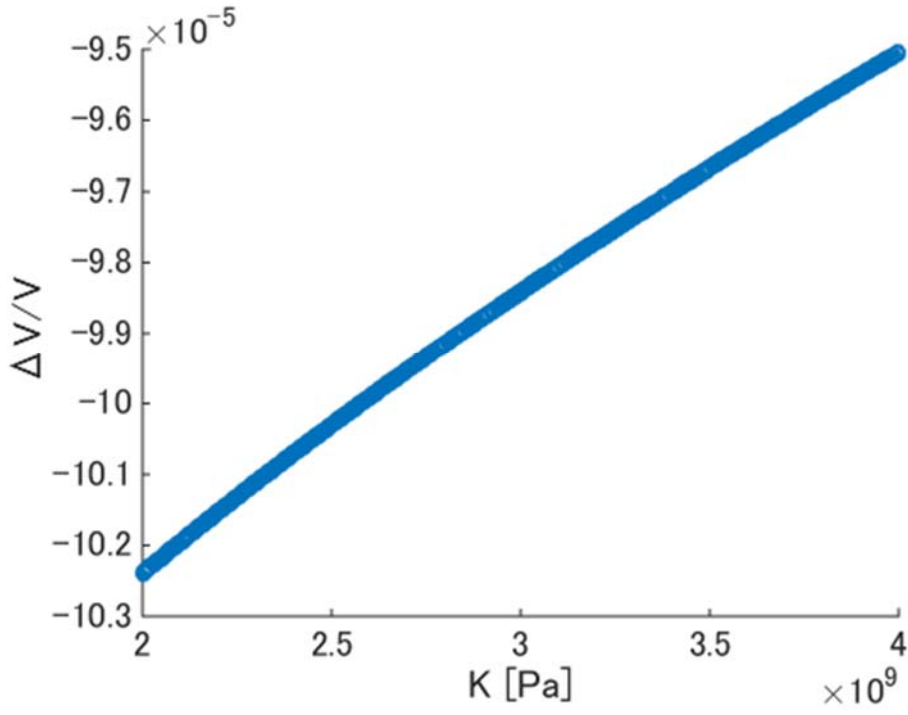


(a)

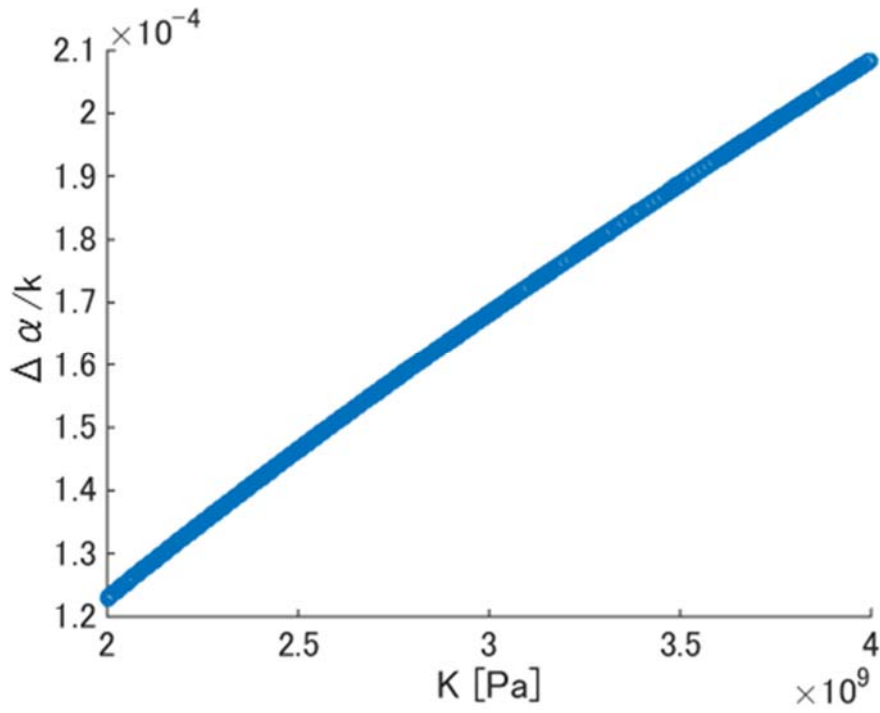


(b)

Fig. 5. (Color Online)



(a)



(b)

Fig. 6. (Color Online)

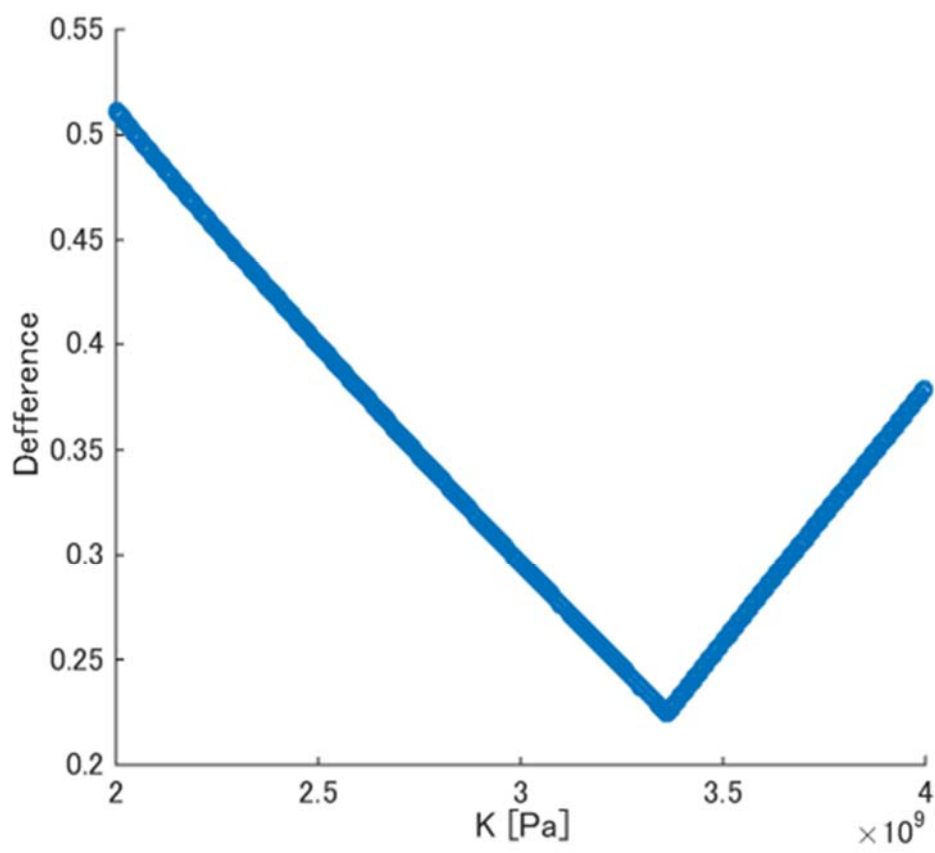


Fig. 7. (Color Online)



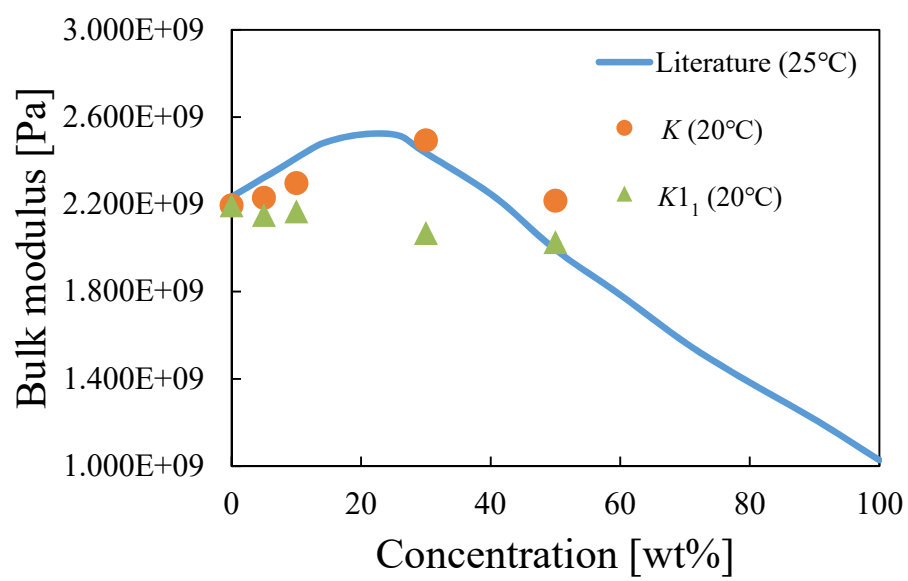
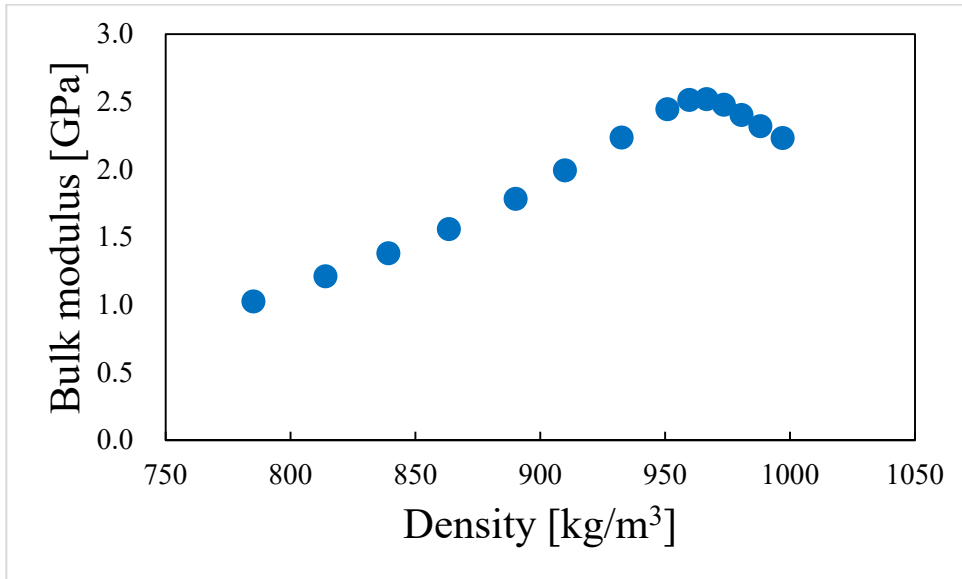
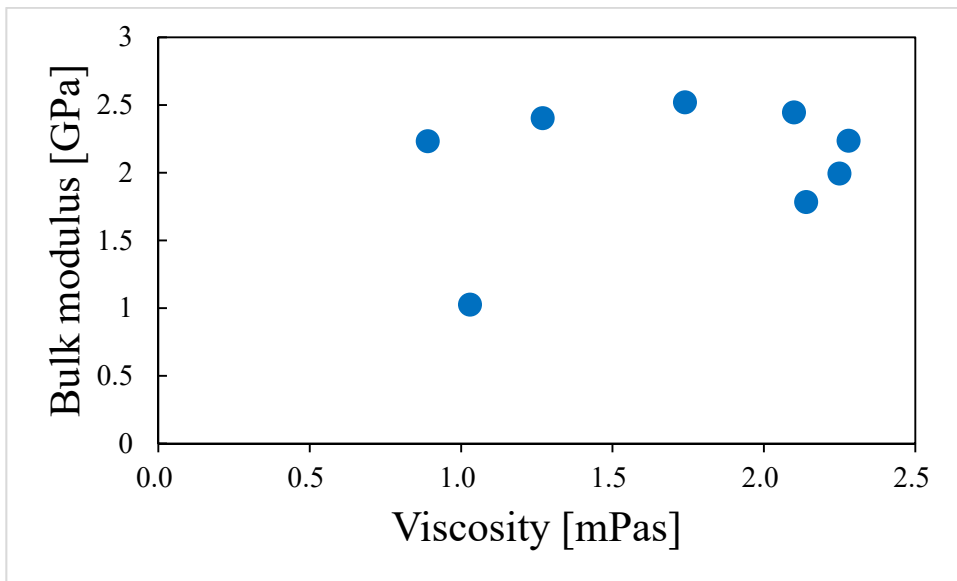


Fig. 8. (Color Online)

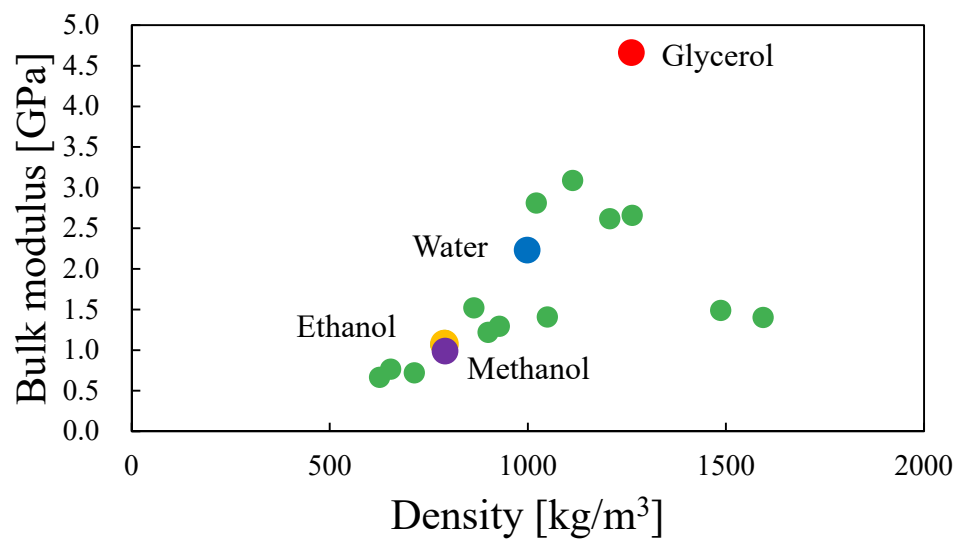


(a)

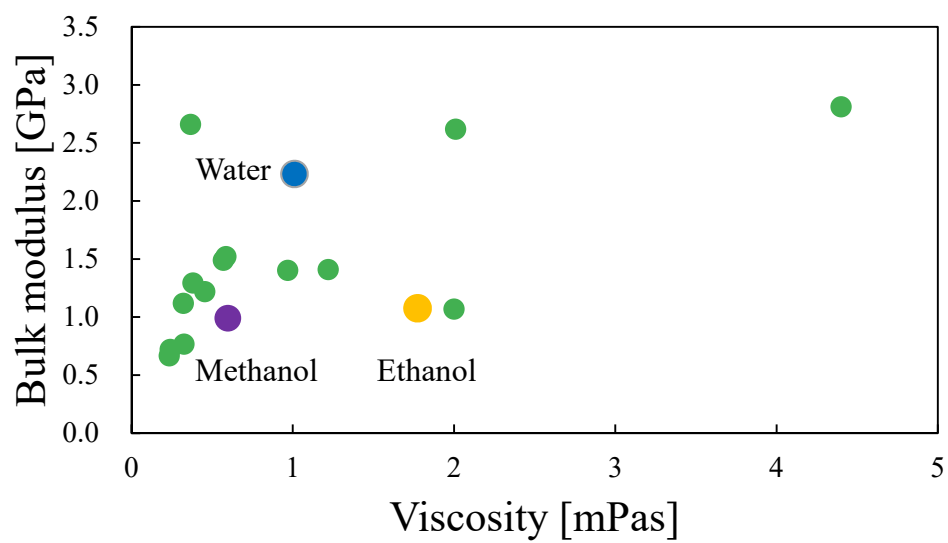


(b)

Fig. 9. (Color Online)



(a)



(b)

Fig. 10. (Color Online)