Novel Micro-Laboratory on Piezoelectric Crystal

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With the continuing development of biotechnology and nanotechnology, a miniaturized and integrated device that is capable of controlling and measuring a small droplet has become desirable. Therefore, we fabricated an integrated device by adopting surface acoustic wave (SAW) streaming to control a droplet and an interdigitated electrode (IDE) sensor to measure the impedance of the droplet. The integrated device is called a “micro-laboratory.” In this work, a micro-laboratory was realized by fabricating an interdigital transducer (IDT) for generating SAWs and an IDE for measuring a droplet on a single piezoelectric crystal. In the experiment, water and glycerol were mixed by SAWs and water/glycerol mixtures were measured by the IDE sensor. From the result, we found that the mixtures were uniformly mixed by the SAWs. Moreover, as a new application, we fabricated a novel micro-laboratory using an isotropic material/matching layer/128YX-LiNbO₃ structure (three-layer structure). By using the three-layer structure, a disposable and low-cost SAW device was realized.

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1. Introduction

With the continuing development of biotechnology and nanotechnology, a miniaturized and integrated device that is capable of controlling and measuring a small droplet has been requested. For example, a pressure-type microchip¹⁾ and a microreactor using electrostatic force²⁾ have been proposed. We propose a novel microchip fabricated on a surface acoustic wave (SAW) piezoelectric crystal. It is called a “micro-laboratory.” When a liquid is placed on a SAW propagating surface, the SAW generated from an interdigital transducer (IDT) radiates a longitudinal wave into the liquid. The radiated longitudinal wave induces liquid dynamics, such as vibrating, streaming, and jetting, with increasing SAW amplitude. These phenomena are known as SAW streaming.³⁾ We have used these phenomena to control a droplet. In addition, an interdigitated electrode (IDE) sensor was used to measure an impedance of a droplet. The impedance depends on liquid concentration and the type of liquid used. The IDE sensor, which has the same structure as the IDT, has high sensitivity and rapid response.⁴⁾ An integrated device comprising a sensor and an actuator is realized on the SAW piezoelectric crystal (128YX-LiNbO₃). In this study, we first compared IDE sensors fabricated on 128YX-LiNbO₃ with those fabricated on SiO₂, because IDE sensors are normally fabricated on SiO₂. Second, we manipulated droplets and mixed them on 128YX-LiNbO₃. Then, the mixed droplets were measured using the IDE sensor. Moreover, as a new application, we propose a novel micro-laboratory using a sensor plate/matching layer/128YX-LiNbO₃ structure.⁵⁾ Using this structure, a disposable and low-cost micro-laboratory was realized.

2. Experimental

A 128YX-LiNbO₃ single crystal (Yamaju Ceramics) was used as a SAW substrate in this study. An IDT with 32 finger pairs, apertures of 2 mm, and a center frequency of 48.17 MHz was designed and fabricated on 128YX-LiNbO₃. The IDT was fabricated using chromium and gold evaporated films with thickness of 20 nm and 200 nm, respectively. The experimental setup is shown in Fig. 1. An RF signal from a standard signal generator (Leader 3220) and a pulse signal from a multifunction synthesizer (NF 1940) were mixed. Then, the signal was amplified by an RF power amplifier (R&K A1000-510). The amplified

signal was fed to the IDT. The amplitude of the input signal was measured using an oscilloscope (Agilent 54615B). Between the amplifier and the IDT, an antenna tuner (Daiwa CNW-319II) was inserted to reduce the reflection from the IDT.

The IDE sensors on 128YX-LiNbO₃ and SiO₂ were connected to a LCR meter (HP 4285A). The number of finger pairs and the aperture of the IDE are the same as those of the IDT. The IDE and IDT were fabricated at the same time. For all impedance measurements, a sine-modulated ac potential with an amplitude of 10 mV was applied to the IDE sensor. Before measurements were taken, the calibration of the IDE was carried out. Impedance spectroscopy was performed at fixed frequencies. All measurements were performed at room temperature.

3. SAW Streaming

When a liquid is placed on a SAW-propagating surface, a longitudinal wave radiates into the liquid, as shown in Fig. 2. The liquid vibrates, streams and jets with increasing SAW amplitude. These phenomena are known as SAW streaming. Several SAW streaming phenomena have been investigated.^{3,6(8)} In this study, we used SAW streaming to manipulate and mix droplets.

4. Measurements of Liquid Properties Using IDE Sensors

4.1 Impedance characteristic of IDE sensor

Sensors with microscaled IDEs have been investigated.^{9,10)} The advantages of IDEs are high sensitivity and rapid response. Generally, the IDE is fabricated on a glass (SiO₂) plate. The electric behavior of the IDE sensor is represented by the equivalent circuit¹¹⁾ shown in Fig. 3. In the circuit, two identical double-layer capacitances (C_{dl}) are connected to the resistance of the solution (R_{sol}) in series, and the dielectric capacitance of the solution (C_{de}) is introduced in parallel with these series elements. Figures 4(a) and 4(b) show the impedance spectra of the IDE sensor for a 0.002 mol/l KCl solution and a 40 wt% glycerol/water mixture, respectively. Impedance spectra were measured at fixed frequencies (75 k, 100 k, 200 k, 300 k, 450 k, 600 k, 1 M, 1.5 M, 3 M, 5 M, 10 M, 15 M, and 19 MHz). As shown in Fig. 4(a), there are three distinct regions in the impedance spectrum, which correspond to the three types of element in the equivalent

circuit. The low-frequency range below 100 kHz is the double-layer region, wherein the double-layer capacitance dominates the signal. In the frequency range from 100 kHz to 10 MHz (the resistive region), the conduction of ions in the solution determines the signal. At even higher frequencies (>10 MHz; the dielectric region), the signal is dominated by the dielectric behavior of the solution. However, as shown in Fig. 4(b), the electric behavior of the IDE sensor is represented only in the resistive and the dielectric regions. Because a glycerol/water mixture is not an electrolyte, it is not necessary to consider the influence of the double-layer capacitance.

The frequency-dependent properties of these three regions are interpreted using the equivalent circuit mentioned above. In this paper, however, the double-layer capacitance can be ignored because it has a negligible effect on measurement frequencies. Therefore, the equivalent circuit of the IDE sensor is simplified, as shown in Fig. 5. The resistance of the solution, R_{sol} , and the dielectric capacitance, C_{de} , are represented by¹²⁾

$$R_{sol} = \frac{1}{nl} \frac{1}{\sigma} \frac{2K}{K'} \frac{\sin \frac{\omega w}{2L}}{\cos \frac{\omega w}{2L}}; \quad (4:1)$$

$$C_{de} = nl \epsilon'' \frac{K \cos \frac{\omega w}{2L}}{2K' \sin \frac{\omega w}{2L}}; \quad (4:2)$$

Here, n is the number of fingers, l is the length of the fingers, σ and ϵ'' are the conductivity and permittivity of the medium, respectively, w is the spacing between electrodes, $L = w + (\text{electrode width})$, and K is the complete elliptic integral of the first kind. In our case, w and the electrode width are equal; thus, $L = 2w$. To evaluate the results, eqs. (4.1) and (4.2) were used to obtain theoretical values.

4.2 Impedance spectra of KCl solutions and glycerol/water mixtures

IDE sensors are normally fabricated on SiO_2 . In this study, we fabricated IDEs on SiO_2 and on 128YX- LiNbO_3 and compared the results with theoretical values. Figures 6(a) and 6(b) show the impedance spectra of the IDE sensors fabricated on SiO_2 and 128YX- LiNbO_3 for six different concentrations (0.0002, 0.0003, 0.0005, 0.002, 0.003, and 0.005 mol/l) of KCl solutions and five different concentrations (20, 40, 60, 80, and 100 wt%) of

glycerol/water mixtures, respectively. Using eqs. (4.1) and (4.2), theoretical values were also calculated. Conductivity was determined from a conventional conductivity meter. Dielectric constants for glycerol/water solutions were obtained from the literature.¹³⁾ For the KCl solution, the dielectric constant was assumed to be equal to that of water and was thus fixed at 78.3.¹³⁾ The measured results agree with the theoretical values. Similar results were obtained at different frequencies (75 k , 200 k , 300 k , 450 k , 600 k, and 1 MHz). Therefore, the IDE sensor fabricated on 128YX-LiNbO₃ is applicable as a liquid sensor.

5. Development of Novel Micro-Laboratory

5.1 Micro-laboratory on piezoelectric crystal

An IDT and an IDE were integrated on 128YX-LiNbO₃. The IDT was used to generate a SAW and the IDE was used as a detector. The voltage applied to the IDT was 20 V_{PAP}. Figure 7 shows the measurement method. A glycerol droplet was placed on the IDE and a distilled water (DW) droplet was placed on the propagation surface by a micropipette. The water droplet was transported and mixed with the glycerol droplet. Because droplet temperature increases during mixing,^{6,8)} it was measured using a thermocouple thermometer (Fluke 51K/J). The thermometer was inserted into the glycerol droplet on the IDE. The results obtained are shown in Fig. 8. At time = 10 s, a SAW was generated and the water was transported. Within 1 s, the water droplet arrived at the IDE and mixing started. The mixing time was 30 s. Then, the signal applied to the IDT was stopped. The impedance was measured after the droplet temperature became constant. In this study, four different concentrations (20, 40, 60, and 80 wt%) of glycerol/water mixtures were produced. The volume of DW and glycerol were varied. The total volume of the glycerol/water mixtures was 20 μ l. For comparison, samples with the same concentrations were prepared in beakers. The results obtained are shown in Fig. 9. The two sets of values agree well. From the results, we concluded that a uniform mixture was achieved by SAW streaming, as expected.

A continuous flow is normally utilized in a microfluidic device. A Y-shaped flow channel is normally utilized for liquid mixing. For a Y-shaped microfluidic device, however, the

mixing of different liquids is difficult because of the presence of laminar flow. Also, the sample amount increases as a result of continuous flow, even if the device is small. In the SAW device, a liquid droplet is transported. The minimum volume of liquid in a 50 MHz SAW device is less than 0.2 μL . When the frequency is increased, the minimum volume decreases. In other words, the required amount of a sample solution is small. This is an advantage in the use of the SAW device as a micro-laboratory substrate. The other advantage is that the droplet is digitally transported. Therefore, the position control of the droplet is performed using an electrical signal.

5.2 Micro-laboratory with three-layer structure

Using a SAW device, a micro-laboratory was realized. Compared with other methods, the use of the proposed device has many advantages. However, because a piezoelectric substrate is expensive, the realization of a disposable micro-laboratory using a piezoelectric substrate is difficult. If a droplet can be moved on a glass plate, a low-cost micro-laboratory can be realized. Therefore, we propose the use of SAW streaming phenomena in a sensor plate/matching layer/128YX-LiNbO₃ structure (three-layer structure). The three-layer structure is shown in Fig. 10. The IDT is fabricated on a piezoelectric substrate. The IDE is fabricated on a glass plate. Using a matching liquid layer, the two plates are bonded. The piezoelectric 128YX-LiNbO₃ substrate and a glass sensor plate (Matsunami; thickness, 200 μm) are used. Distilled water is used as the matching liquid. The thickness of the distilled water layer is 5 μm . The voltage applied to the IDT was fixed at 35 V_{PAP} . The SAW generated from the IDT radiates a longitudinal wave into the matching liquid. The radiated wave generates an acoustic wave in the glass plate. Therefore, droplet streaming on the glass plate can be induced. In the measurement, one liquid was placed on the IDE sensor and the other was transported. The measurement method is shown in Fig. 11. In this paper, several different concentrations of KCl solution were mixed and six different concentrations (0.0002, 0.0003, 0.0005, 0.002, 0.003, and 0.005 mol/l) of KCl solution were produced. Concentrations of transported and placed droplets are summarized in Table I. For comparison, samples with the same concentrations were prepared in beakers. The results obtained are shown in Fig. 12. The two sets of values

agree well. We obtained highly stable and reproducible results owing to the fact that KCl solutions are electrolytes. Using the proposed structure and by changing the sensor plate, the piezoelectric substrate can be reused. In other words, a disposable micro-laboratory was realized by adopting a three-layer structure.

6. Conclusions

A “ micro-laboratory ” fabricated on SAW piezoelectric crystal is presented as a new application. Moreover, as a new application, we fabricated a novel micro-laboratory using an isotropic material/matching layer/128YX-LiNbO₃ structure. A disposable and low-cost micro-laboratory with a SAW device was realized by adopting this structure. Using the micro-laboratory, a small amount of sample liquid was transported, mixed and measured. In the future, we will apply the micro-laboratory to the measurement of immunoreactions. The theoretical calculation of characteristics of the propagating wave is necessary for the optimization of the structure.

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Figure captions

Fig. 1. Experimental setup used in this study.

Fig. 2. Schematic illustration of SAW streaming. A longitudinal wave radiates from a leaky-SAW.

Fig. 3. Equivalent circuit of IDE sensor with double-layer capacitances, resistance of the solution, and dielectric capacitance.

Fig. 4. Impedance spectra of IDE sensor in (a) 0.002 mol/l KCl solutions and (b) 40 wt% glycerol/water mixture.

Fig. 5. Simplified equivalent circuit of IDE sensor with the resistance of the solution and the dielectric capacitance.

Fig. 6. Impedance spectra of IDE sensor fabricated on SiO_2 and 128YX-LiNbO₃ in (a) KCl solutions and (b) glycerol/water mixtures.

Fig. 7. Schematic illustration of micro-laboratory fabricated on 128YX-LiNbO₃.

Fig. 8. Time response of droplet temperature.

Fig. 9. Experimental results for the micro-laboratory.

Fig. 10. Schematic illustration of three-layer structure.

Fig. 11. Schematic illustration of novel micro-laboratory with three-layer structure.

Fig. 12. Experimental results for novel micro-laboratory with three-layer structure.

Table I. Concentrations of transported and placed droplets of KCl solutions.

Transported (mol/l)	Placed (mol/l)	After mixture (mol/l)
0.0001	0.0003	0.0002
0.0001	0.0005	0.0003
0.0001	0.0009	0.0005
0.001	0.003	0.002
0.001	0.005	0.003
0.001	0.009	0.005

Figure 1
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Figure 2
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Figure 3
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Figure 4(a)
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Figure 4(b)
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Figure 5
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Figure 6(a)
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Figure 6(b)
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Figure 7
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Figure 11
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Figure 12
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