Behavior of Surface Acoustic Wave Resonators in Supercritical CO₂

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1. Introduction

A supercritical fluid is a fluid at a temperature and pressure above its critical point (31.05°C and 7.285 MPa for CO₂) in which a gas phase and a liquid phase coexist. Since supercritical fluids have properties between those of a gas and a liquid, they have been used in many fields. It has been noticed that the solubility of a substance rapidly decreases with a sudden reduction in the pressure, leading to the precipitation of small particles. Moreover, supercritical fluids have high permeability in fine structures because there is no surface tension. Therefore, research and development on the formation and processes of nano- and microscale substances have been carried out actively.¹

A problem of applying supercritical fluids is the heterogeneity of their molecular distribution, i.e., their density fluctuations. This induces unusual effects such as the acceleration of which reaction velocity and a rapid increase in solubility. By utilizing a surface acoustic wave (SAW), which has superior performance for sensing physical properties on a surface, it may be possible to realize a sensor for measuring the density fluctuation of supercritical fluids. With the aim of investigating the behavior of acoustic waves in high-pressure and supercritical CO₂, the resonance frequency change of an AT-cut quartz crystal microbalance (QCM) has been previously reported.²

In this study, the behaviors of the resonance frequency and impedance of SAW resonators were measured in high-pressure CO₂, and the frequency changes were compared with the reported changes using a QCM.

2. Fabrication of SAW Resonators

To produce a Rayleigh wave on 128° Y-X LiNbO₃ and a Love wave with only pure shear-horizontal (SH) particle motion on ST-90° X quartz, SAW resonators consisting of a single-electrode interdigital transducer (IDT) with the wavelength λ of 20 μm, 30.5 (LiNbO₃) or 100.5 (quartz) finger pairs, and an overlap length of 100 λ, and shorted grating reflectors with 20 metal strips were fabricated using an aluminum film with a thickness of 0.03 λ. The sample was fixed to an aluminum holder so that the whole surface of the SAW resonator came in contact with high-pressure CO₂ and its electrodes were bonded to an SMA receptacle by an aluminum wire.

3. Measurement of Resonance Properties

The experimental setup is shown in Fig. 1. A flow of liquid CO₂ into the chamber in which the sample holder was set was induced using a liquid feed pump. The resonance properties were measured using a network analyzer as a function of the pressure in the chamber, which was controlled by a back-pressure regulator. The temperature in the

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chamber was monitored by a thermocouple. Figure 2 shows the impedance properties (amplitude) of the SAW resonator when the pressure was reduced from 10 to 0.1 MPa. For both LiNbO$_3$ and quartz samples, a marked change in the impedance ratio was observed at approximately 5.5 MPa. The rate of change of the resonance frequency $f_r$ and antiresonance frequency $f_a$ and the corresponding impedances ($Z_r$ and $Z_a$) are shown in Figs. 3 and 4, respectively. $f_r$ and $f_a$ rapidly decreased between 5 and 6 MPa except for $f_a$ of LiNbO$_3$. This behavior is similar to that reported for an AT-cut QCM. However, the rate of change of $f_a$ for the LiNbO$_3$ sample reached 1,300 ppm and was 3.5 times larger than that of the AT-cut QCM. When the pressure was applied in the case of the LiNbO$_3$ sample, the behaviors of $f_r$ and $f_a$ were different from those at a reduced pressure. This is caused by the temperature change due to the inflow of liquid CO$_2$.

As can be seen from Fig. 4, it was found that the impedance, particularly $Z_a$, can be used to sense the difference between gas and liquid phases without affecting the temperature. In the future, a theoretical verification of the above results and an investigation of the resonance properties in supercritical CO$_2$ will be reported.

4. Conclusions
The behaviors of SAW resonators fabricated on 128° Y-X LiNbO$_3$ and ST 90°-X quartz were measured in high-pressure CO$_2$. The rate of change of $f_a$ at a pressure between 5 and 6 MPa in the case of LiNbO$_3$ reached 1,300 ppm and was 3.5 times larger than that of an AT-cut QCM. It was found that the impedance, particularly $Z_a$, was effective for sensing the difference between gas and liquid phases without affecting the temperature. In the future, a theoretical verification of the above results and an investigation of the resonance properties in supercritical CO$_2$ will be reported.

Acknowledgment
The authors would like to thank Prof. E. Kondoh and Mr. K. Tamai at the University of Yamanashi for valuable suggestions and their assistance with experiments.

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