Proceedings of Symposium on Ultrasonic Electronics, Vol. 31 (2010) pp. 351-352
6-8 December, 2010

Behavior of Surface Acoustic Wave Resonators in Supercritical CO_2

超臨界 CO_2 中における弾性表面波共振子の振舞い

Katsuhiro Hayashi[1‡], Shoji Kakio[1] and Yasuhiko Nakagawa[2]
([1]Univ. Yamanashi, [2]Prof. Emeritus, Univ. Yamanashi)
林　克洋[1‡]，垣尾 省司[1]，中川 恭彦[2]　([1]山梨大院・医工，[2]山梨大名誉教授)

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 CO_2) 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 CO_2, 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 CO_2, 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 $LiNbO_3$ 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 ($LiNbO_3$) 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 CO_2 and its electrodes were bonded to an SMA receptacle by an aluminum wire.

g10me027@yamanashi.ac.jp

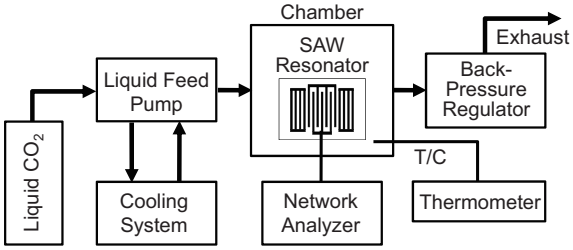

Fig. 1　Experimental setup.

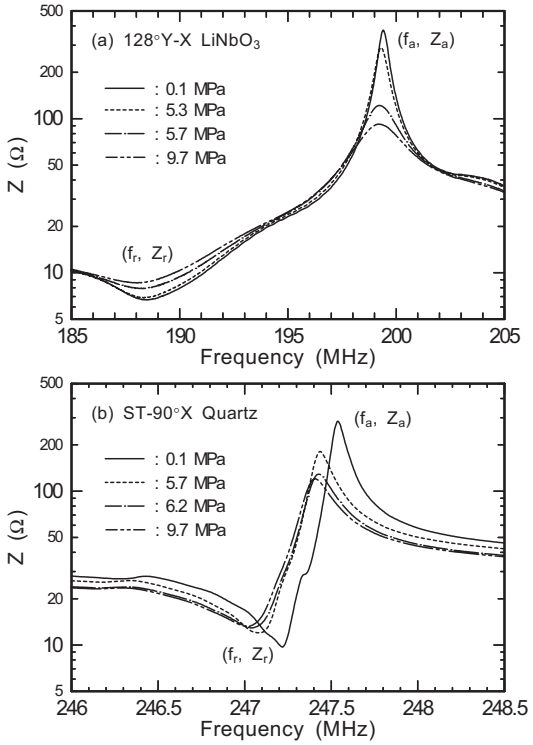

Fig. 2　Measured impedance properties.

3. Measurement of Resonance Properties

The experimental setup is shown in **Fig. 1**. A flow of liquid CO_2 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

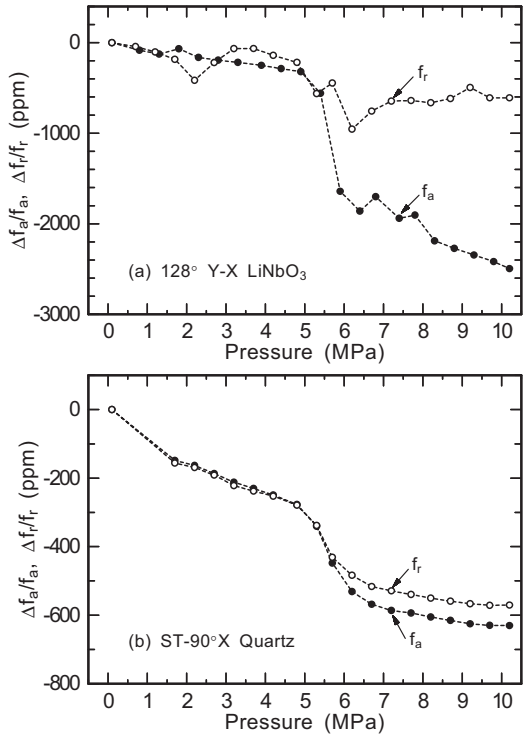

Fig. 3 f_r and f_a as a functions of CO_2 pressure.

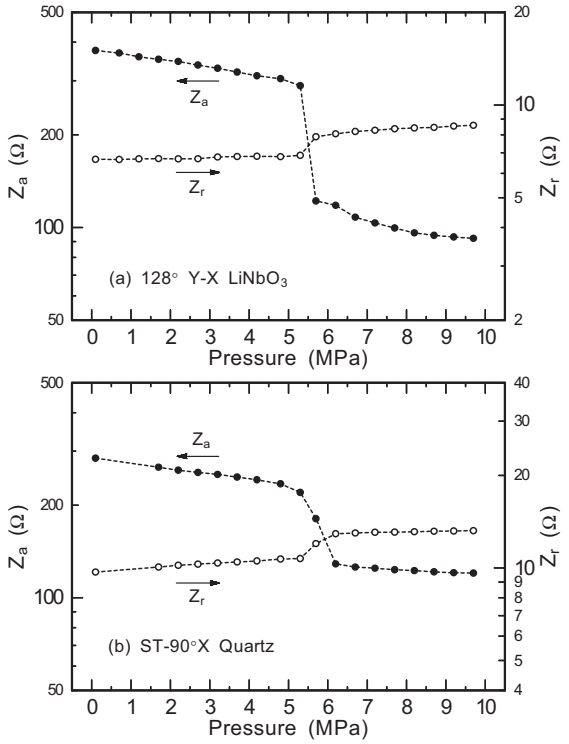

Fig. 4 Z_r and Z_a as a functions of CO_2 pressure.

chamber was monitored by a thermocouple. **Figure 2** shows the impedance properties (amplitude) of the SAW resonator when the pressure was reduced pressure 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_r of $LiNbO_3$. This behavior is similar to that reported for an AT-cut QCM.[2] 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 more clearly than f_r and f_a. Moreover, when the pressure was applied in the case of the $LiNbO_3$ sample, similar impedance properties to those at a reduced pressure were obtained without the effect of a temperature change.

Since the measured temperature in the chamber was approximately 26°C, the above properties were in the subcritical state slightly below the critical point at which a small change in pressure or temperature results in large changes in density and viscosity.[3,4] Therefore, it can be considered that the above marked changes in the frequency and impedance were due to changes in density and viscosity, respectively.

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.

References

1. T. Sako, I. Okajima:"Chorinkairyutai No Hanashi", Nikkan Kogyo Shinbunsya, 2006. [in Japanese]
2. K. Park *et al.*, J. Supercritical Fluids, **29** (2004) 203.
3. S. Angus *et al.*, Inter. Thermodynamic Tables of the Fluid State Carbon Dioxide. Oxford, England, Pergamon Press, 1976.
4. M. Matsubara: PhD Thesis, University of Yamanashi, Japan, 2010. [in Japanese]