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Resonance Properties 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共振特性

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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 CO_2), at which a gas phase and a liquid phase coexist.[1] Supercritical CO_2 functions as a moderate solvent, has zero surface tension, and has high permeability in fine structures. These properties are regarded as ideal for the processing of nano- and microscale substances, and research and development on the formation of such substances and processes have been carried out actively.[2]

A problem of applying supercritical fluids is the heterogeneity of their molecular distribution, i.e., fluctuations in their density. The authors previously reported the behaviors of a Rayleigh-type surface acoustic wave (SAW) resonator fabricated on 128˚ Y-X $LiNbO_3$ and a pure shear-horizontal-type SAW (SH-SAW) resonator fabricated on ST 90˚-X quartz in high-pressure CO_2.[3] Abrupt changes in resonance properties were observed at a certain pressure.[3] It was found that the impedance can be used for sensing the difference between gas and liquid phases.[3] On the other hand, it has been found that an SH-SAW on 36˚ Y-X $LiTaO_3$ has high sensitivity for the detection of physical properties of the liquid phase [4-6].

In this study, the resonance properties of an SH-SAW resonator fabricated on 36˚ Y-X $LiTaO_3$ and the impedance of an interdigitated electrode (IDE) fabricated on a nonpiezoelectric substrate[7] were measured in high-pressure CO_2.

2. Measurement of Resonance Properties

An SH-SAW resonator consisting of a single-electrode interdigital transducer (IDT) with a wavelength λ of 20 μm, 30.5 finger pairs, an overlap length of 100 λ, and shorted grating reflectors with 50 metal strips was fabricated on 36˚ Y-X $LiTaO_3$ using an aluminum film with a thickness of 0.03 λ. The setup for measuring the resonance properties in high-pressure CO_2 was the same as that reported previously.[3] A flow of liquid CO_2 was induced into a chamber in which sample was set using a liquid feed pump. The resonance property was measured as a function of the pressure in the chamber using a network analyzer.

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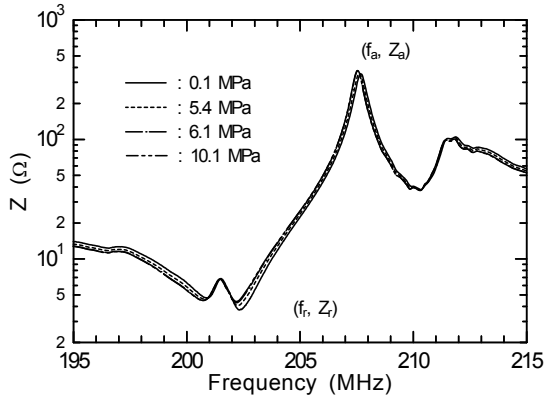

Fig. 1 Measured resonance properties.

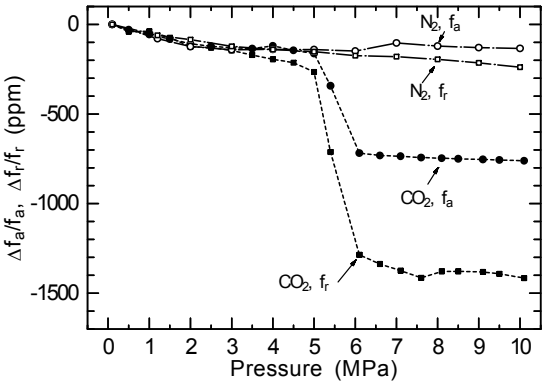

Fig. 2 Rates of change of f_r and f_a as functions of pressure.

Furthermore, to measure the change in the resonance properties due to only pressure, nitrogen (N_2) gas instead of CO_2 was also induced into the chamber.

Figure 1 shows the resonance properties (amplitude) of the SH-SAW resonator when the pressure was reduced from 10 to 0.1 MPa. The monitored temperature was approximately 18˚C. Therefore, the measured properties were those in the subcritical state slightly below the critical point. The resonance frequency f_r and antiresonance frequency f_a decreased with increasing CO_2 pressure, similarly to in our previous report.[3] The resonance impedance Z_r and antiresonance impedance Z_a increased with increasing pressure, slightly different behavior from that in the previous report,[3] in which Z_r increased and Z_a decreased with increasing CO_2 pressure; in other words, the resonance properties deteriorated with increasing CO_2 pressure.

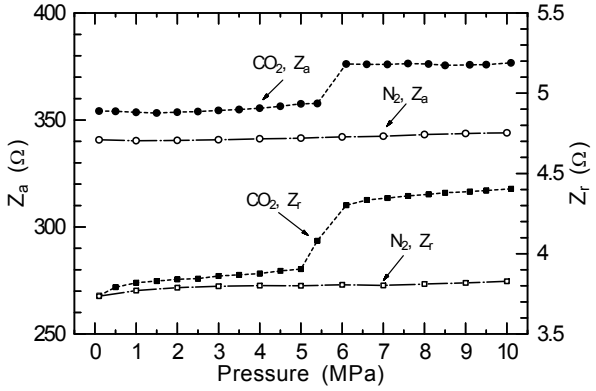

Fig. 3 Z_r and Z_a as functions of pressure.

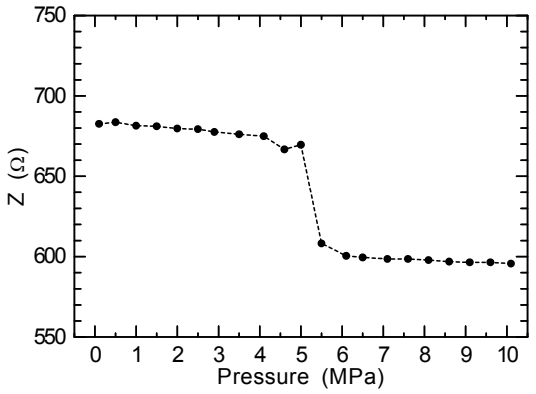

Fig. 4 IDE impedance as a function of CO_2 pressure.

The rates of change of f_r and f_a as functions of CO_2 pressure or N_2 pressure are shown in **Fig. 2**. f_r and f_a rapidly decreased between CO_2 pressures of 5 and 6 MPa. The rate of change of f_r was larger than that of f_a and reached approximately 1,000 ppm. On the other hand, f_r and f_a monotonically decreased with increasing N_2 pressure. From the comparison with the changes in the case of applying N_2 pressure, it is considered that the abrupt changes were caused by not only the pressure itself but also the density and viscosity as described later.

Figure 3 shows Z_r and Z_a as functions of CO_2 pressure or N_2 pressure. The abrupt changes between CO_2 pressures of 5 and 6 MPa were smaller than those of a Rayleigh wave on 128° Y-X $LiNbO_3$ in the previous report.[3] This difference is considered to be because a Rayleigh wave attenuates owing to the radiation of the energy of the longitudinal wave into the liquid. From the comparison with the results for N_2, it is also considered that the abrupt changes in impedance were caused by not only the pressure itself but also the density and viscosity.

The density and viscosity (shear viscosity) of CO_2 were calculated using the open software EOS-SCx.[8] When the CO_2 temperature was set to 20°C, discontinuous changes in the density and viscosity appeared at 5.7 MPa between the gas and liquid phases. The permittivity of CO_2 also changed discontinuously at the phase boundary. The phase velocity and attenuation of the SH-SAW are known to depend on the product of density and viscosity.[5] Therefore, for the SH-SAW resonator, the abrupt changes in the frequency were mainly due to changes in density and viscosity, and the abrupt changes in the impedance were caused by changes in not only the density–viscosity product but also the permittivity.

3. Impedance Measurement of IDE

An IDE with a period λ of 20 μm, 300 finger pairs, a metallization ratio of 0.5, and an overlap length of 100 λ was fabricated on a SiO_2 glass substrate using an aluminum film with a thickness of 0.015 λ. The impedance in high-pressure CO_2 was measured at a frequency of 10 MHz by a similar experimental setup to that used in the previous section. **Figure 4** shows the impedance of the IDE as a function of CO_2 pressure. The discontinuous change in the impedance observed between 5 and 6 MPa is considered to be due to the change in the dielectric constant of CO_2. Therefore, it was found that the impedance of the IDE on a nonpiezoelectric substrate can be used to sense the difference between gas and liquid phases.

4. Conclusions

The resonance properties of an SH-SAW resonator on 36° Y-X $LiTaO_3$ were measured in high-pressure CO_2. The abrupt changes in the resonance properties were considered to be due to discontinuous changes in the density, viscosity, and permittivity of CO_2. Moreover, it was found that the impedance of the IDE on a nonpiezoelectric substrate can be used to sense the difference between gas and liquid phases. In the future, an investigation of the resonance properties in supercritical CO_2 will be reported.

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