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QCM method using 100MHz SC-cut crystal units - examination of viscoelastic loads -

100MHz SC-cut 水晶振動子を用いたＱＣＭ法
- 粘弾性負荷に関するモデルについて

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1. Introduction

Quartz crystal microbalance (QCM) is a method of measuring the mass change of a material on its electrode by detecting the frequency change of a quartz crystal and has been applied in various fields[1-6]. Due to the growing interest in viscoelastic loading, the application of QCM has been extended to liquid systems. The kanazawa-Gordon equation submitted by Kanazawa in 1985 was proven to be applicable to water systems[6]. In 2007, Weihnacht and Bruenige, working on the monitoring of the clotting process, revisited the Kanazawa equation and proposed a general model to represent the behavior of QCM over a wider frequency range for fluids with viscoelastic properties[1,2]. AT-cut crystal units are widely used in QCMs. They are characterized by high sensitivity to frequency and ease of application in oscillators. Bruenige and his colleagues used an AT-cut crystal[2]. Bruenige and his colleagues use an AT-cut crystal[1,2].

For this study, we used a 100-MHz stress compensated cut (SC-cut) crystal resonator (3rd overtone) because there are few examples of using SC-cut sensors in QCM[5]. We clarified the temperature characteristics of an SC-cut crystal unit and analyzed it using the model submitted by Bruenige et al. In this article, we describe a simulation measurement system targeting aqueous glycerin solutions and verify the paper requested by Bruenige et al. Bruenige et al. focused on viscosity, which is an element of QCM, and showed that it is more accurate than the Kanazawa equation. They also simulated spurious modes.

2. Measuring system

Figure 1 shows the measurement system. The thermostatic chamber is connected to the thermostatic bath by a pipe, and the water in the chamber is kept at a constant temperature. When the sample is placed in the thermostatic bath and the temperature is changed, the resonance frequency changes. The C-mode and B-mode are measured at the frequencies of the 100-MHz SC-cut crystal unit.

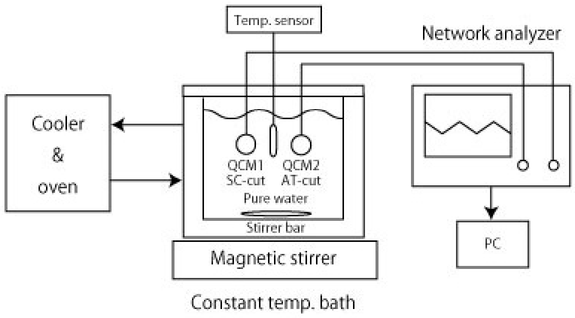

Fig.1 Measuring system.

3. Bruenige formula

Bruenige et al., who study viscoelastic loads such as blood coagulation, measured QCM admittance as a function of frequency using a network analyzer, which is expressed as

$$Y = i \cdot \frac{\omega c_p}{1 - k^2 \frac{\tan(\zeta)/\zeta}{1 + i \frac{\mathrm{Zload} \cdot tan(\zeta)}{2 \cdot z_q - i \cdot \frac{Zload}{\tan(\zeta)}}}} + i \cdot \omega c_s \quad (1)$$

The parameters are as follows

K ：Piezoelectric coupling coefficient (8.8% for AT-cut, 4.99% for SC-cut)
ω ：Angular frequency
Zq : Crystal acoustic impedance ($Zq = \rho_q \cdot v$ density： ρ_q Propagation speed： v)
ζ ：Normalized frequency ($\zeta = \omega dq/2v$ Crystal thickness: dq)
Cp : Capacity contributes to vibration
Cs : Remaining capacity
Z_{load} : Acoustic impedance of the load

Using the approximation equation, we can derive clear expressions for the change in resonant frequency ($\Delta\zeta$res) and anti-resonant frequency ($\Delta\zeta$a) (maximum and minimum absolute values of admittance Y) due to loading, assuming Cs = 0.

$$\tan(\zeta) \approx -\frac{1}{\zeta - \zeta_n}, \zeta_n = n\frac{\pi}{2}, n = 1,3,5 \ldots. \quad (2)$$

$$\Delta\zeta_{res} = -\frac{\mathrm{Im}(Zload)}{2 \cdot Z_q} + \frac{k^2}{\pi} - \sqrt{\left(\frac{\mathrm{Re}(Zload)}{2 \cdot Zq}\right)^2 + \left(\frac{k^2}{\pi}\right)^2} \quad (3a)$$

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$$\Delta\zeta_a = -\frac{\mathrm{Im}(Zload)}{2\cdot\rho_q v} - \frac{k^2}{\pi} + \sqrt{\left(\frac{\mathrm{Re}(Zload)}{2\cdot\rho_q v}\right)^2 + \left(\frac{k^2}{\pi}\right)^2} \quad (3b)$$

The complex shear modulus is G* = G' + iG" = iwη*, and the propagation velocity of the shear wave in liquid is vFl=√(G^*⁄ρfluid). The case of liquid loading and multiple cases are omitted.

For a Newtonian fluid, $\eta'' = 0$, so the real part of the load impedance equals the imaginary part. For small viscosities such as water and ethanol, Eq. (3a) becomes

$$\Delta\zeta_{res} = -\frac{\sqrt{0.5\rho_{fluid}\omega\eta'}}{2Zq} \quad (4)$$

This is the Kanazawa equation [6].

Quartz resonators usually have additional modes at frequencies higher than the desired main mode and affect each other when high viscosity or multiple viscosity materials are used. To reduce these spurious modes, Eq. (1) can be extended and Eq. (5) can be derived.

$$Y_{ext2} = \sum_{i=0}^{n} \frac{iwC_i}{1-(K_i\cdot K)^2\frac{\tan(\zeta_i)/\zeta_i}{1+i\frac{Z_{loadi}\cdot\tan(\zeta_i)}{2\cdot Z_q\cdot\left(1-\frac{iZloadi}{2\cdot Zq\cdot\tan(\zeta_i)}\right)}}} + i\omega C_s \quad (5)$$

4. Simulation

We verified that the Kanazawa equation and Bruenige equation are similar in frequency displacement for small-viscosity Newtonian fluids. We used a 9-MHz AT-cut crystal resonator by Bruenige et al. For the Kanazawa equation, the curve was obtained by substituting the parameters in the lower part.

Elastic shear modulus:μx(AT):2.95×10^{10}(kg•m-2)

Crystal Densityρ:2.65×10^{3}(kg•m-3)

Figure 2 shows that the Kanazawa and Bruenig equations are in agreement. Figure 3 shows the simulation results for the high-viscosity Newtonian fluid. We will attempt to derive these equations further for future work.

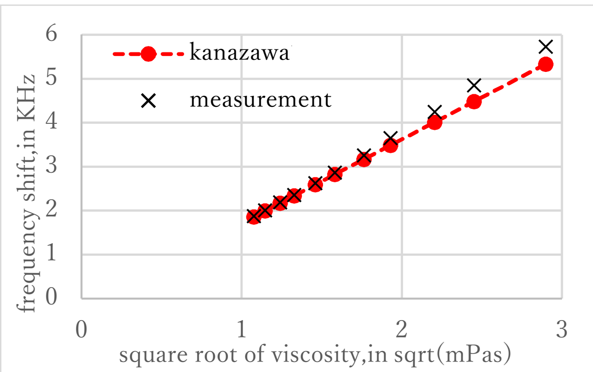

Fig.2 Low viscosity

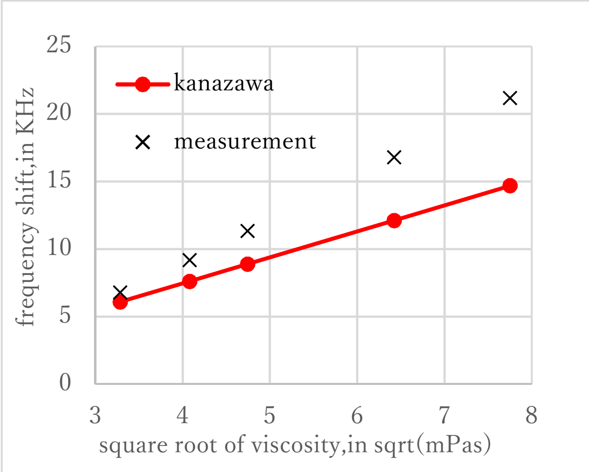

Fig.3 High viscosity

5. Conclusions

To clarify the temperature characteristics of a 100-MHz SC-cut crystal unit, we clarified the experimental equipment and conducted a si mulation experiment targeting the glycerin aque ous solution in an AT-cut crystal unit. We confi rmed that the model submitted by Bruenige et al. (low-viscosity Newtonian fluid) and the Kan azawa equation were in agreement and that the y were not in agreement with high-viscosity Ne wtonian fluid. We will aim for a more specific method for treating an SC-cut sensor as a QC M.

Acknowledgments

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