Thickness shear mode quartz crystal resonators with optimized elliptical electrodes*

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Quartz crystal resonators (QCRs) with circular electrodes have been widely used for various liquid and gas sensing applications. In this work, quartz crystal resonators with elliptical electrodes were studied and tested for liquid property measurement. Mindlin's theory was used to optimize the dimension and geometry of the electrodes and a 5-MHz QCR with minimum series resistance and without any spurious modes was obtained. A series of AT-cut QCRs with elliptical electrodes of different sizes were fabricated and their sensing performances were compared to devices with circular electrodes. The experimental result shows that the device with elliptical electrodes can obtain lower resonance impedance and a higher Q factor, which results in a better loading capability. Even though the sensitivities of devices with elliptical and circular electrodes are found to be similar, the sensor with elliptical electrodes has much higher resolution due to a better frequency stability. The study indicates that the performance of QCRs with elliptical electrodes is superior to that of traditional QCRs with circular electrodes.

Keywords: quartz crystal resonator, elliptical electrode, liquid sensing

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1. Introduction

Thickness shear mode quartz crystal resonators (QCR) with circular electrode have been widely used in sensing applications such as thin film deposition control, gas detection and liquid viscosity measurement.[1−3] Recently, there has been growing research interest in the effect of electrode geometry. Richardson and Bhethanabotla *et al.*[4] presented a QCR with a ring electrode that produces uniform mass sensitivity distribution across the device. In Refs. [5]–[8], the authors studied the modified-electrode QCR, which was used to probe the electrical properties of the analyte. Wang *et al.*[9] studied the effects of electrodes with continuously varying thickness on energy trapping in thickness shear mode quartz resonators. Mindlin[10] obtained an important theoretical result on electrode shape and derived a formula for the optimal electrode shape of AT-cut quartz resonators. In his theory, the optimal electrodes are nearly elliptical, with the major axis exceeding the minor axis by 25%. Based on Mindlin's theory of optimal electrodes, Yang *et al.*[11,12] and Xu *et al.*[13] obtained some numerical results and plotted the optimal electrode shape and size of an AT-cut and a SC-cut quartz crystal resonator, which indicates that the optimal electrodes of those cuts are both nearly elliptical. However, the results have not been verified experimentally. Moreover, the sensing performance of a QCR with optimal electrodes has not been studied. When applied to liquid property sensing, a traditional QCR with circular electrodes shows high resonance impedance and a low Q factor, which results in low loading capability and frequency stability. Electrode shape and size are important factors affecting resonance parameters.[14,15] Therefore, it is necessary to study QCR sensors with the optimal electrode shape and size.

In this paper, the experimental results for optimal eletrodes of QCR are presented. With the shape of optimal electrodes of AT-cut QCR known as elliptical, we proceed to study its effect on the sensing performance of the QCR, which includes loading capability, sensitivity, frequency stability and resolution.

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2. Theory

For an AT-cut quartz plate resonator, it is defined that x_1 and x_3 are the in-plane coordinates and x_2 is the plate normal. In the plane of the plate, along a direction determined by an angle ϕ, which is measured from the positive x_1 axis toward the positive x_3 axis, the optimal electrode length L is given in Ref. [10]. According to Mindlin's theory of the optimal electrode shape and size,[10] the optimal electrode length L is given by

$$l = \frac{L}{2h} = \pi \left[\frac{(\alpha/3 + \hat{\gamma}_{11}) \cos^2 \phi + \hat{\gamma}_{55} \sin^2 \phi}{2(R + 4k_{26}^2/\pi^2)} \right]^{1/2}, \quad (1)$$

where

$$\hat{\gamma}_{11} = \frac{\gamma_{11}}{3k^2 c_{66}}, \quad \hat{\gamma}_{55} = \frac{\gamma_{55}}{3k^2 c_{66}}, \quad \alpha = \frac{1+3R}{1+R}, \quad (2)$$

$$\gamma_{11} = \frac{s_{33}}{s_{11}s_{33} - s_{13}^2}, \quad \gamma_{55} = \frac{1}{s_{55}}, \quad R = \frac{2\rho' h'}{\rho h}, \quad (3)$$

$$k^2 = (1 + R - 8k_{26}^2/\pi^2)(\pi^2 \bar{c}_{66}/12c_{66}), \quad (4)$$

$$k_{26}^2 = e_{26}^2/(\bar{c}_{66}\varepsilon_{22}), \quad \bar{c}_{66} = c_{66} + \frac{e_{26}^2}{\varepsilon_{22}}, \quad (5)$$

and $2h$ is the plate thickness.

Using the above formulae we have calculated the optimal electrode shape and size for a resonator with resonance frequency of 5 MHz (the corresponding plate thickness $2h = 0.33$ mm) and electrode/resonator mass ratio of $R = 0.004$.

The result shows that the optimal electrode shape is nearly elliptical. The semi-major axis is 5.4 mm and the semi-minor axis is 4.27 mm, as shown in Fig. 1. Theoretically, the resonator with the obtained optimal electrodes can achieve better energy trapping. In other words, its electrode size can be larger without causing spurious mode resonances. Therefore, the resonator can have both a lower series impedance and a single resonance mode simultaneously.

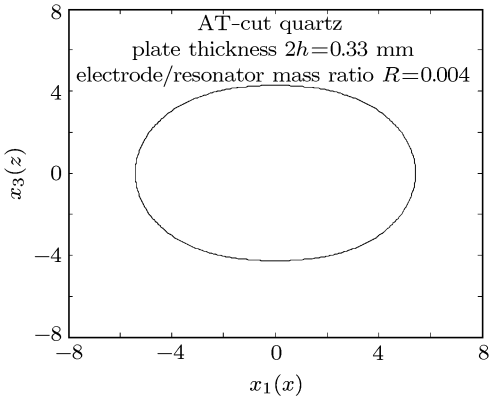


Fig. 1. Optimal electrodes of an At-cut quartz resonator with a fundamental resonance frequency of 5 MHz and an electrode/resonator mass ratio of $R = 0.004$.

3. Experiment

In this work, to explore the validity of the calculated optimal electrodes, we have designed and fabricated a series of AT-cut QCRs with elliptical electrodes of different sizes. The semi-minor axis of which range from 3.8 mm to 4.8 mm and the major axis exceeds the minor axis by 25%. In addition, for comparison, a series of AT-cut QCRs with circular electrodes are also fabricated, the radii of which range from 3.8 mm to 4.8 mm.

To obtain the values of resonance impedance and status of the spurious modes, the impedance characteristics of the QCRs with different electrode shapes and sizes were tested by impedance analyser Agilent4294A.

The sensing performance of QCRs with optimized elliptical electrodes was tested and compared with those of QCRs with circular electrodes, which includes loading capability, sensitivity, frequency stability and resolution

Sugar syrup water solutions of different concentrations were used to test the devices' loading capability, liquid viscosity and density sensitivity. The frequency stability of the devices in liquid was then tested, based on which, the resolution of the devices can be obtained and compared.

A crystal sensor holder was used for installing a QCR device and forming a liquid sensing cell. When in the liquid phase experiment, the crystal sensor holder was dipped into the measured medium. An Agilent4294A impedance analyser was used to test the devices' resonance impedances. The devices' frequency responses were measured using an SRS QCM100 phase-locked oscillator and an FC-1300 frequency counter

4. Results and discussion

4.1. Device optimization

According to Mindlin's theory, optimal electrodes satisfy Bechmann's number in every direction, that is to say, no spurious mode occurs for devices with electrodes smaller than the optimal electrodes, while electrodes larger than optimal electrodes can bring spurious mode. In addition, under the condition of no spurious mode, devices with elliptical electrodes can

use a larger electrode size, which leads to smaller resonance impedance compared to devices with circular electrodes.

Figure 2 shows the impedance characteristic curves of QCRs with elliptical electrodes of different sizes. For devices with electrodes of semi-minor axis=3.8 mm and those with electrodes of semi-minor axis=4.16 mm, which are smaller than the calculated optimal electrodes, their impedance spectra show no spurious modes. While, for devices with electrodes of semi-minor axis=4.4 mm and devices with electrode of semi-minor axis=4.8 mm, which are larger than the calculated optimal electrodes, spurious modes emerged.

The impedance characteristic curves of the QCRs with circular electrodes of different size are shown in Fig. 3. The impedance spectra of the devices with electrodes of radius =3.8 mm and those with electrodes of radius =4.16 mm show no spurious modes. While, for devices with electrodes of radius =4.4 mm and those with electrodes of radius =4.8 mm, spurious modes appear.

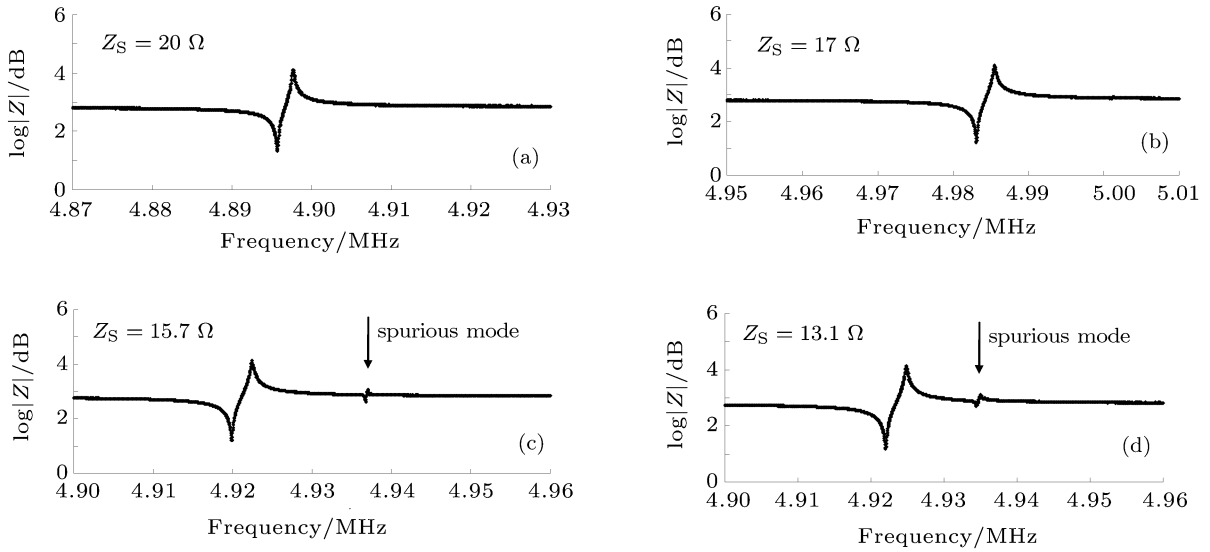


Fig. 2. Impedance of the resonator with elliptical electrodes. The electrodes size is given as "(semi-minor axis, semi-major axis)", the resonance impedance is noted as Z_S. (a) Electrode size (mm): (3.8, 4.75); (b) Electrode size (mm): (4.16, 5.2); (c) Electrode size (mm): (4.4, 5.5); (d) Electrode size (mm): (4.8, 6.0).

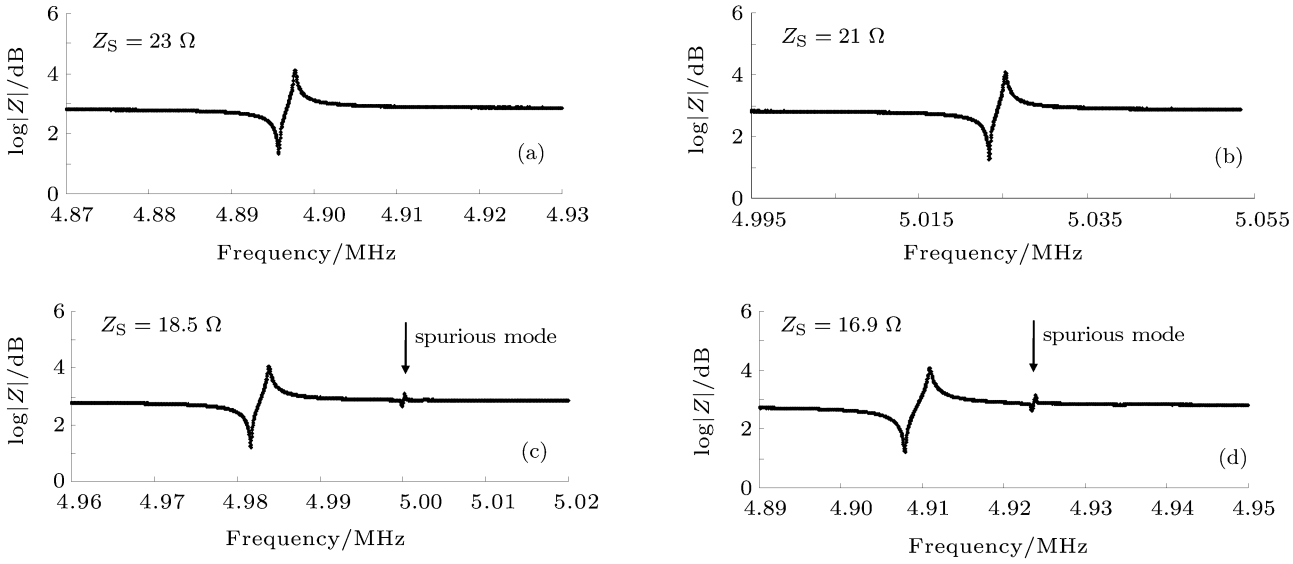


Fig. 3. Impedance of the resonator with circular electrodes. The electrode size is given as electrode radius, the resonance impedance is noted as Z_S. (a) Electrode radius (mm): 3.8; (b) Electrode size (mm): 4.16; (c) Electrode radius (mm): 4.4; (d) Electrode radius (mm): 4.8.

Under the condition of no spurious mode, for devices with elliptical electrodes, the obtained usable maximum electrodes area is 67.96 mm^2 (the semi-minor axis =4.16 mm, the semi-major axis =5.2 mm), the corresponding series resonance impedance is 17 Ω. While, for devices with circular electrodes, the obtained usable maximum electrodes area is 54.37 mm^2 (the electrodes radius=4.16 mm), the corresponding series resonance impedance is 21 Ω. The results show that under the condition of no spurious mode, a QCR with elliptical electrodes can use a larger electrode area, which results in smaller resonance impedance. The results agree well with Mindlin's theory of the optimal electrode. From the above experimental results, it can be seen that the obtained optimal elliptical electrodes have a semi-minor axis of 4.16 mm and a semi-major axis of 5.2 mm, which is close to theoretical result, namely semi-minor axis =4.27 mm and semi-major axis =5.4 mm. In Mindlin's theory of the optimal electrodes, some approximations are made. For example, the crystal plate is approximately infinite. Thus the optimal electrode obtained by experiment is slightly smaller than the calculated one.

4.2. Sensing performance of a QCR with elliptical electrodes

The sensing performance of a QCR with elliptical electrodes of semi-minor axis=4.16 mm and semi-major axis=5.2 mm was tested and compared to QCR with circular electrodes of radius=4.16 mm, which is a usable maximum size under the condition of no spurious mode. The measured sensing performance includes loading capability, sensitivity, frequency stability and resolution.

4.2.1. Loading capability

When a QCR is used for liquid sensing applications, liquid loading leads to an obvious increase of resonance impedance, which can result in a lower Q factor. Partially inverted sugar syrup water solutions with acoustic viscosities AV (AV is equal to the product of liquid viscosity and density, the unit of which is cp.g/ml) ranging from 19 cp·g/ml to 282 cp·g/ml were used to test the relationship between the resonance impedance and liquid loading. With an increasing concentration of solution, the viscosity and density become higher, and this results in a larger liquid loading impedance. Figure 4 shows the dependence of the resonance impedance of the two kinds of devices on the square root of acoustic viscosity AV of the sugar syrup water solution. It can be seen that a QCR with elliptical electrodes can obtain a lower resonance impedance compared to a QCR with circular electrodes. Figure 5 shows the resonance impedance curves of the two kinds of QCRs in sugar syrup water solutions with acoustic viscosities AV of 110 cp·g/ml. It is shown that a QCR with elliptical electrodes can obtain a larger Q factor than that with circular electrodes. It is indicated that the device with elliptical electrodes has a stronger loading capability.

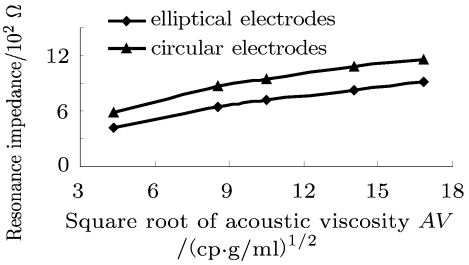


Fig. 4. Dependence of the resonance impedance of the two kinds of devices on the square root of acoustic viscosity AV of sugarcane syrup water solution.

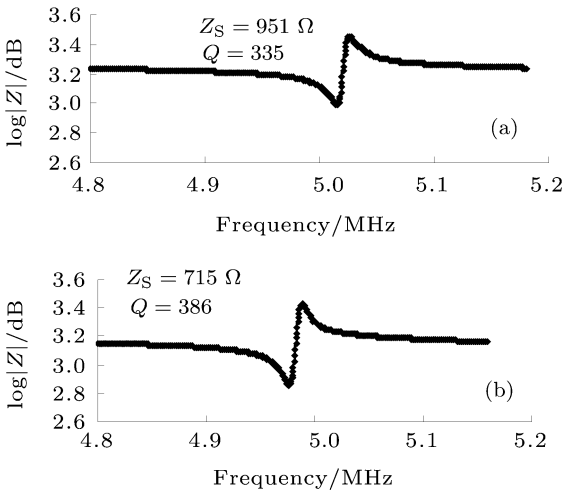


Fig. 5. Resonance impedance curves of the two kinds of QCRs in sugarcane syrup water solution with acoustic viscosity AV of 110 cp·g/ml: (a) circular electrodes and (b) elliptical electrodes.

The equivalent circuit of the QCR with one side facing the liquid is shown in Fig. 6. C_0 is the static capacitance of the device. L_1, R_1 and C_1 are the motional inductance, motional resistance, and motional capacitance of the unperturbed QCR respectively. L_2 and R_2 are the motional inductance and motional resistance contributed by liquid loading, respectively. R_1 and R_2 are given by Eqs. (6) and (7)[16]

$$R_1 = \frac{\eta_q \pi^2}{8k_0^2 C_0 \bar{c}_{66}}, \tag{6}$$

$$R_2 = \frac{\pi}{8k_0^2 C_0}\left(\frac{2\rho\eta}{\omega\bar{c}_{66}\rho_{\rm q}}\right)^{1/2}. \quad (7)$$

When the device is in resonance, the resonance impedance $Z_{\rm S}$ is approximately equal to the sum of R_1 and R_2, as shown in Eq. (8)

$$\begin{aligned} Z_{\rm S} &\approx R_1 + R_2 \\ &= \frac{1}{8k_0^2 C_0}\left[\frac{\eta_{\rm q}\pi^2}{\bar{c}_{66}} + \pi\left(\frac{2\rho\eta}{\omega\bar{c}_{66}\rho_{\rm q}}\right)^{1/2}\right]. \end{aligned} \quad (8)$$

In Eqs. (6), (7) and (8), $\eta_{\rm q}$ is the effective quartz viscosity, k_0 is the piezoelectric coupling factor, ω is the angular frequency at resonance, $\rho_{\rm q}$ is the quartz mass density, ρ and η are the density and viscosity of the liquid, respectively, $\bar{c}_{66}$ is the piezoelectrically stiffened shear modulus.

It can be seen that the resonance impedance $Z_{\rm S}$ is inversely proportional to the static capacitance C_0 of the device, which is directly proportional to the area of electrodes. Under the condition of no spurious mode, a QCR with elliptical electrodes can use a larger electrode area, which results in a larger static capacitance C_0. Thus, its resonance impedance Z_S is lower than that of a QCR with circular electrodes; the QCR with elliptical electrodes has better loading capability. This characteristic is helpful for the application of QCRs to high viscous systems.

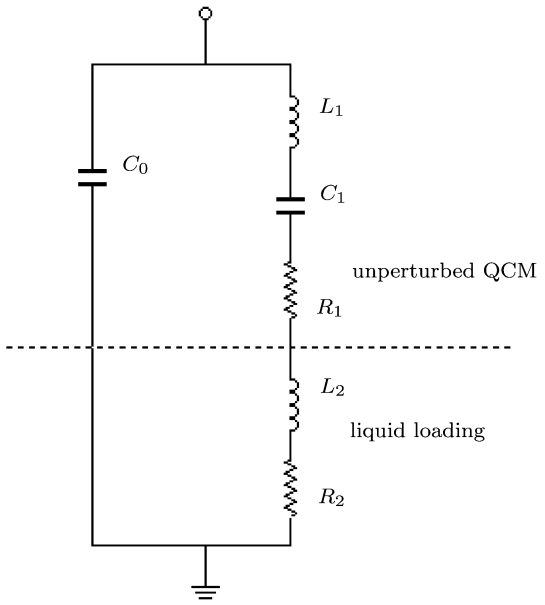


Fig. 6. The equivalent circuit of a QCR with one side facing liquid.

4.2.2. Sensitivity

Sugar syrup water solutions of different concentrations were used to test the devices' sensitivity to liquid acoustic viscosity. The result showed that the devices' frequency shift is linear to the change of the square root of liquid acoustic viscosity, as shown in Fig. 7. Their linear coefficients k_0 obtained are both about 725 Hz/(cp·g/ml)$^{1/2}$ from Eq. (9)

$$k_0 = \frac{\Delta f}{\Delta(\sqrt{AV})}, \quad (9)$$

where Δf is frequency shift, $\Delta(\sqrt{AV})$ is the change of the square root of acoustic viscosity of sugar syrup water solutions.

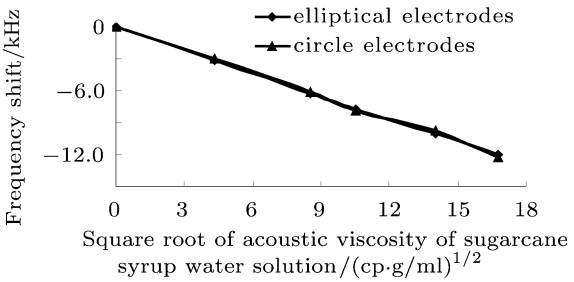


Fig. 7. Dependence of the frequency shift of the two kinds of devices on the square root of acoustic viscosities AV of sugar syrup water solutions.

It can be seen that the QCR with elliptical electrodes has similar sensitivity to the QCR with circular electrodes. This suggests that the sensitivity of a QCR is not obviously affected by the electrode geometry.

4.2.3. Frequency stability and resolution

The frequency versus time curves of the two kinds of devices in 20 wt% sugar syrup water solution were tested using FC-1300 frequency counter. The results are shown in Fig. 8.

For comparison, a reference frequency is used in the y axis of Fig. 8. For a normalization of the frequency stability measurements in the time domain, the IEEE has proposed the Allan deviation, shown by Eq. (10)[17]

$$\sigma_y^2(\tau) = \frac{1}{2(m-1)}\sum_{n=1}^{m-1}(\bar{f}_{n+1}(\tau) - \bar{f}_n(\tau))^2. \quad (10)$$

To evaluate the frequency stability of the sensors, Allan deviations were calculated for each device by varying the average time τ, from 1 s to 38 s, using 1200 samples, and the maximum value in this range was taken to be $\sigma_y(\tau)_{\max}$. The calculated $\sigma_y(\tau)_{\max}$ for QCR with elliptical electrodes is 3.56, while that of the QCR with circular electrodes is 4.77. This means

that the QCR with elliptical electrodes has higher frequency stability.

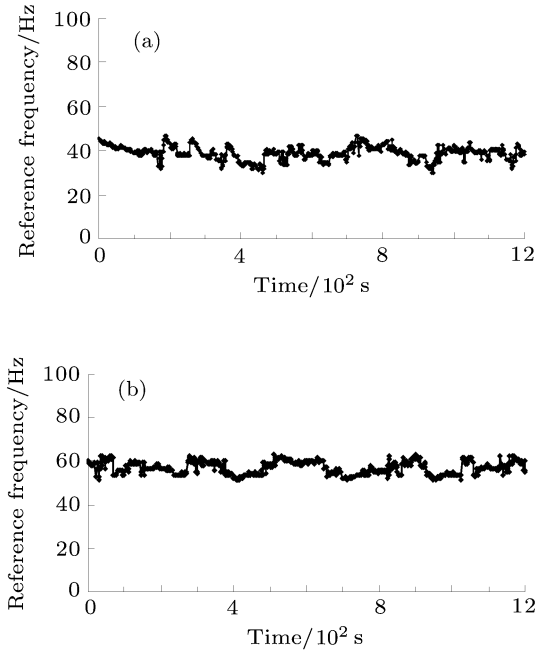


Fig. 8. Frequency stability measurement for QCRs with different electrodes with acoustic viscosity AV of 110 cp·g/ml: (a) circular electrodes and (b) elliptical electrodes.

This deviation allows us to estimate the frequency detection limit $\Delta f(\tau)$ using Eq. (11)

$$\Delta f(\tau) = \sigma_y(\tau)_{\max}, \tag{11}$$

then, the resolution of the sensor can be obtained by the relationship between the detection limit and the sensitivity coefficient k, as shown in Eq. (12)

$$\text{Resolution} = \frac{\Delta f(\tau)}{k}. \tag{12}$$

In the above equation (10), m is the number of samples $\bar{f}_n(\tau)$ is the average frequency of the n-th sample calculated over a time interval τ starting from an instant t_n, as shown in Eq. (13),

$$\bar{f}_n(\tau) = \frac{1}{\tau+1}\sum_{t=t_n}^{t_n+\tau} f(t), \tag{13}$$

where $f(t)$ is the resonance frequency in instant of the t-th second, k is the sensitivity coefficient.

From Subsection 4.2.2, it can be seen that the devices' frequency shift is linear against the square root of liquid acoustic viscosity with a linear coefficient of 725 Hz/(cp·g/ml)$^{1/2}$, namely

$$\frac{\mathrm{d}f}{\mathrm{d}(\sqrt{AV})} = 725\ (\mathrm{Hz}/(\mathrm{cp}\cdot\mathrm{g/ml})^{1/2}). \tag{14}$$

By using Eq. (14), the sensitivity coefficient k of devices' frequency shift versus liquid acoustic viscosity can be obtained, namely

$$k = \frac{\mathrm{d}f}{\mathrm{d}(AV)} = \frac{1}{2\sqrt{AV}}\cdot 725\ (\mathrm{Hz}/(\mathrm{cp}\cdot\mathrm{g/ml})^{1/2}). \tag{15}$$

For 20 wt% sugar syrup water solution, the measured acoustic viscosity AV is 110 cp·g/ml. Then, using Eq. (15), the sensitivity coefficient k can be obtained, namely

$$k = 34.6\ (\mathrm{Hz}/(\mathrm{cp}\cdot\mathrm{g/ml})). \tag{16}$$

Using Eqs. (11), (12), and (16), the resolution of QCR with elliptical electrodes is calculated to be 0.1 (cp·g/ml), while that of QCR with circular electrodes is 0.138 (cp·g/ml). It can be seen that QCR with elliptical electrodes can obtain obviously better resolution.

The result may be because QCRs with elliptical electrodes have a larger static capacitance C_0, which results in a smaller liquid loading impedance. According to Ref. [18], the Q factor is inversely proportional to the series impedance under the condition of $\Delta f \ll f_{\mathrm{s}}$ (f_{s} represents the series resonance frequency). Therefore, the QCR with elliptical electrodes can obtain larger Q factor, which results in better frequency stability and resolution.

5. Conclusion

In this work, QCRs with elliptical electrodes were studied both theoretically and experimentally. They are tested by liquid property measurement and compared to those with circular electrodes. The results showed that QCRs with elliptical electrodes can obtain lower resonance impedance and a higher Q factor, so they have better loading capability. In addition, compared to QCRs with circular electrodes, QCRs with elliptical electrodes have better frequency stability and similar sensitivity, indicating that the latter can obtain much better resolution. The results indicate that a QCR with elliptical electrodes has better performance compared to traditional a QCR with circular electrodes.

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