Uniform Mass Sensitivity Distribution of Elliptically Designed Electrodes Based on a Quartz Crystal Microbalance

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ABSTRACT: Uniformization of mass sensitivity distribution is conducive to the application of the quartz crystal microbalance (QCM) in some fields. However, the sensitivity of the QCM sensor surface perpendicular to the displacement direction is higher than that of the displacement direction in the mass sensitivity distribution of ring and double-ring QCMs, which leads to poor reproducibility of the sensor. Considering the effect of the electrode structure on the mass sensitivity distribution, we found that for ring- and double-ring-type QCMs, when the elliptical single ring and double-ring electrode structures are combined, an approximately uniform mass sensitivity can be obtained in all directions. Therefore, this study proposes the elliptical single-ring and elliptical double-ring electrode structure design. Through theoretical calculations and three-dimensional finite element analysis verification, a systematic investigation is carried out to quantify the effect of the ratio of the minor axis to the major axis of the elliptical electrode on the mass sensitivity distribution in different directions, and the optimal ratio is found to be 0.8. Comparing the mass sensitivity of the new type of electrodes and the original electrodes, the result shows that the mass sensitivity distribution of the elliptical double-ring electrode structure is more uniform. Hence, these specially designed electrodes are conducive to improving the repeatability.

1. INTRODUCTION

The quartz crystal microbalance (QCM) has been widely used in many fields, including analytical chemistry, immunology, and drug development, due to its high-quality factor and high sensitivity. However, in practical applications, the sample to be tested cannot be uniformly and rigidly distributed on the entire electrode surface of the QCM, which leads to low repeatability of the measurement results. Low repeatability is one of the main reasons why the measurement results have certain error. The repeatability of measurements by the QCM is, to a large extent, limited by the unevenness of its sensitivity distribution.

For m−m- and n−m-type QCMs (QCMs with symmetrical and asymmetrical electrodes), the mass sensitivity distribution is similar to the Gaussian distribution. This means that the same mass distributed in different regions will induce a different frequency shift, which has a great effect on the measurement errors. To solve this problem, Josse et al. proposed a single-ring electrode structure whose mass sensitivity distribution presents a bimodal curve. Huang et al. designed dot-ring and double-ring QCMs to improve the uniformity of the mass sensitivity distribution. The existing investigation methods concerning the QCM mainly include experiments, the three-dimensional (3D) finite element method (FEM), and two-dimensional (2D) theoretical analysis. Before an experiment, a numerical or theoretical investigation on the basic characteristics and parameter optimization of the QCM is helpful to the experimental design. Numerical techniques and 3D FEM, in particular, are gradually replacing the analytical treatments. Compared to 2D theoretical analysis, 3D FEM is more flexible with regard to geometry.

There is a phenomenon in the 3D model of the ring- and double-ring-type QCM: the amplitude of vibration on the surface of the QCM sensor perpendicular to the direction of displacement is enhanced in the vibration distribution of the QCM. This phenomenon exists in the vibration distribution of circular and rectangular single-ring electrodes, as well as circular double-ring electrodes and even multiple concentric ring electrodes. However, there is no simple physical reason for why this phenomenon occurs. We have conducted the corresponding research and hypothesized that the phenomenon may be due to the strengthening of the trapping effect. It is necessary to improve the vibration amplitude in this

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direction to make the mass sensitivity distribution more uniform. To compensate for the defect of mass sensitivity distribution, this study combines the elliptical structure with the ring electrode structure and proposes the elliptical single-ring-type QCM. Compared with other electrode structures, the mass sensitivity distribution of the double-ring structure is more uniform, so we also propose the elliptical double-ring electrode structure. We then model and simulate these new types of electrodes to show their practical modal properties in finite element simulations. Through 2D theoretical analysis and 3D FEM, theoretical investigation and simulation verification of the new types of electrode structure QCMs are carried out.

2. RESULTS AND DISCUSSION

2.1. Theoretical Derivation of Mass Sensitivity. We collectively refer to single-ring electrodes and double-ring electrodes as ring electrodes. The top and side views of the QCMs with the elliptical ring electrodes are shown in Figure 1. Both types of electrodes are formed in an elliptical shape, where the major axis lies in the $x_1$ direction and the minor axis lies in the $x_3$ direction. The side of the elliptical electrode is used for sensing. The sensing surface of the QCM is usually divided into a fully electroded region E, a partially electroded region P, and a non-electroded region U. In the fundamental mode, the quartz crystal produces displacement in the $x_1$ axis direction and the acoustic wave propagates along the thickness direction of the crystal ($x_2$ direction).

The mass sensitivity of the QCM is an important design consideration, and it is calculated by the mass sensitivity function describing the mass sensitivity distribution on the surface of the QCM.\(^{25,26}\)

\[
S_f(r, \theta) = C_f \int_0^\infty \int_0^{2\pi} r A(r, \theta)^2 \, d\theta \, dr
\]

where $A(r, \theta)$ is the displacement amplitude function on the sensor surface, $r$ is the distance from that point to the center, $C_f$ is Sauerbrey’s sensitivity constant, and $S_f(r, \theta)$ is the mass sensitivity function on the sensor surface. The particle displacement amplitude function does not vary with the angle direction at the fundamental frequency.\(^8\)

Figure 1a shows a schematic of the elliptical single-ring-type QCM structure. The parameters $P_1$ and $P_2$ are the inner and outer radius, respectively, of the major axis of the elliptical single-ring electrode, and the parameters $Q_1$ and $Q_2$ are the inner and outer radius, respectively, of the minor axis of the elliptical single-ring electrode. Taking into account the different elasticity inside the surface of the plate, the propagation constants in the $x_1$ and $x_3$ directions are different, and the thickness vibration wave near the cutoff frequency is taken as the potential wave. The wave equation is approximated as\(^{27}\)

\[
\frac{\delta^2 A}{\delta (a_1x_1)^2} + \frac{\delta^2 A}{\delta (a_3x_3)^2} + (k^2 - k_\zeta^2)A = 0
\]

where $A$ is the particle vibration displacement, $k = \omega/\nu$ is the wave number of the excitation frequency, $k_\zeta = \omega_\zeta/\nu$ is the wave number of the cutoff frequency, $\omega$ and $\omega_\zeta$ are the excitation angular frequency and cutoff angular frequency, respectively, and $\nu$ is the propagation velocity in the crystal. The values of the anisotropy constants $a_1$ and $a_3$ in eq 1 depend on the material of the plate. In the case of an AT-cut quartz plate\(^{28}\)
Table 1. Comparison of Three Groups of FEM Results for Elliptical Single-Ring and Elliptical Double-Ring Electrodes

| Electrode                  | Group | Frequency (Hz) | Mass Sensitivity Distribution | ΔA    |
|----------------------------|-------|----------------|------------------------------|-------|
| Elliptical Single-Ring     | A     | 20367408       | ![Graph](image1)              | 0.09613 |
| Elliptical Single-Ring     | B     | 20359567       | ![Graph](image2)              | 0.01243 |
| Elliptical Single-Ring     | C     | 20348623       | ![Graph](image3)              | 0.06243 |
| Elliptical Double-Ring     | A     | 20395261       | ![Graph](image4)              | 0.02963 |
| Elliptical Double-Ring     | B     | 20388421       | ![Graph](image5)              | 0.01175 |
| Elliptical Double-Ring     | C     | 20382771       | ![Graph](image6)              | 0.06159 |
Figure 3. Vibration displacement distribution of QCMs with circular ring and elliptical ring electrodes: (a) circular single-ring electrode; (b) elliptical single-ring electrode; (c) circular double-ring electrode; and (d) elliptical double-ring electrode.

| Table 2. Design Parameters of the Circular Single-Ring Electrode |
|---------------------------------------------------------------|
| electrode parameters | \( R_1 \) | \( R_2 \) |
| value (mm)           | 0.4375   | 1.75    |

| Table 3. Design Parameters of the Circular Double-Ring Electrode |
|---------------------------------------------------------------|
| electrode parameters | \( R_1 \) | \( R_2 \) | \( R_3 \) | \( R_4 \) |
| value (mm)           | 0.2188   | 0.6563  | 0.875   | 1.75    |
\[ \gamma_{11} = 0.5140, \ 
\gamma_{55} = 0.6503 \]

where \( \gamma_{11} \) and \( \gamma_{55} \) are the bending and twisting rigidities and \( \epsilon_{66} \) is the stiffness constant.

According to eq 2, the following relationship between the sizes \( P_1, P_2, Q_1, \) and \( Q_2 \) of the elliptical single-ring electrode can be established, and then the energy trapping effect along the \( x_1 \) and \( x_3 \) directions is the same

\[ a_1P_1 = a_3Q_1, \quad a_1P_2 = a_3Q_2 \tag{4} \]

This circular ring electrode shape (Figure 1b) is adopted because its vibration analysis is comparatively easier with the help of coordinate transformation, and axially asymmetrical vibrations do not appear spuriously in this case.26 The electrodes are essentially optimal in the sense of Mindlin in that they approximately satisfy the criterion for Bechmann’s number in every direction.29

To treat the \( x_1 \) and \( x_3 \) directions in an equivalent manner, the coordinate plane \((x_1, x_3)\) of Figure 1a is transformed to the \((a_1x_1, a_3x_3)\) plane, as shown in Figure 1b. Here, \( R_1 \) and \( R_2 \) represent the outer and inner radii of the electrode after the coordinates have been transformed into the polar coordinates. When eq 2 is converted into polar coordinates \((r, \theta)\), we obtain

\[ r^2 \delta^2 A \frac{\partial^2}{\partial r^2} + r \frac{\partial A}{\partial r} + \frac{\partial^2 A}{\partial \theta^2} + (rk_r)^2 A = 0 \tag{5} \]

where \( k_r^2 = k^2 - k_c^2 \).

Assuming that the excited acoustic wave is mostly limited within the fully electroded region, the operating frequency \( f \) should satisfy the condition \( f_U < f < f_P < f_{\text{U}} \). Therefore, the solution of eq 5 can be written as

**Table 4. Comparison of Mass Sensitivity Distribution Uniformity of FEM Results for Four Electrode Structures**

| electrode                | circular single ring | circular double ring | elliptical single ring | elliptical double ring |
|--------------------------|----------------------|----------------------|------------------------|------------------------|
| frequency (Hz)           | 20.342564            | 20.382959            | 20.358459              | 20.393462              |
| \( \epsilon \)           | 0.2874               | 0.2506               | 0.2438                 | 0.2358                 |

**Figure 4.** Surface displacement distribution of QCMs with circular ring and elliptical ring electrodes: (a) circular single-ring electrode; (b) elliptical single-ring electrode; (c) circular double-ring electrode; and (d) elliptical double-ring electrode.
where \( k_0^R, k_1^R, \) and \( k_2^R \) are the expressions of \( k_i \) for the E, P, and U regions, respectively and \( f_0, N_0, U_0, \) and \( K_0 \) are the Bessel functions and the modified Bessel's functions, respectively. The unknown amplitude constants \( A, B, C, \) and \( D \) are obtained by solving a set of linear homogeneous equations, and the mass sensitivity function \( S(r) \) can be then determined.

Figure 1c shows the structure of the elliptical double-ring-type QCM. The parameters \( P_1 \) and \( P_2 \) represent the inner and outer radii of the inner ring electrode, and \( P_3 \) and \( P_4 \) represent the inner and outer radii of the outer ring electrode of the major axis of the elliptical single-ring electrode. Similarly, the parameters \( Q_1, Q_2, Q_3, \) and \( Q_4 \) represent the inner and outer radii of the inner and outer ring electrodes of the minor axis of the elliptical single-ring electrode.

From eq 2, the following relationships between the sizes \( P_1, \) \( P_2, \) \( P_3, \) \( P_4, \) \( Q_1, \) \( Q_2, \) \( Q_3, \) and \( Q_4 \) of the following elliptical double-ring electrode can be established.

\[
a_1P_1 = a_2Q_1, \quad a_4P_4 = a_3Q_4, \quad a_2P_2 = a_3Q_2, \quad a_1P_3 = a_2Q_3, \quad a_4P_3 = a_4Q_4
\]

As seen in Figure 1c, the coordinate plane \( (x_1, x_2) \) of Figure 1d is transformed to the \( (a_1x_1, a_2x_2) \) plane. The parameters \( R_1, \) \( R_2, \) \( R_3, \) and \( R_4 \) represent the inner and outer radii of the inner ring electrode and the inner and outer radii of the outer ring electrode. When the operating frequency \( f \) satisfies \( f_0 < f < f_1 \), the solution of particle displacement amplitude \( A(r) \) for the QCM with the elliptical double-ring electrode can be rewritten as

\[
A(r) = \begin{cases} 
    A_0(k_r^R) & 0 \leq |r| \leq R_1 \\
    B_1(k_r^R) + C_0(k_i^E) & R_1 \leq |r| \leq R_2 \\
    D_1(k_r^R) + E_0(k_i^E) & R_2 \leq |r| \leq R_3 \\
    F_1(k_r^R) + G_0(k_i^E) & R_3 \leq |r| \leq R_4 \\
    H_0(k_i^R) & R_4 \leq |r| \leq \infty 
\end{cases}
\]

(6)

The unknown constants \( A, B, C, D, E, F, G, \) and \( H \) can be determined by solving a set of linear homogeneous equations as previously described, and the mass sensitivity function \( S(r) \) is then determined.

2.2. Elliptical Ring QCM Optimization. As mentioned in eq 1, the mass sensitivity \( S_l \) is related to displacement \( A(r) \). The mass sensitivity distribution can be improved by studying the vibration displacement distribution. The uniform mass sensitivity distribution is our aim in the sections.

2.2.1. Optimum Size of the Elliptical Ring Electrode. Mindlin obtained important theoretical results for the optimal shape and size of QCM electrodes using 2-D plate equations. The optimal electrodes satisfy the Bechmann’s number in every direction. Therefore, when the QCM is in thickness shear vibration, the electrode region vibrates in the phase without charge cancellation on the electrodes. That is to say, no spurious mode occurs for the QCM with electrodes smaller than the optimal electrodes, while electrodes larger than the optimal electrodes can bring the spurious mode. The shape of the optimal electrode differs minimally from that of an ellipse, the major axis exceeding the minor axis by 25%.29

According to eq 3, we can calculate \( a_1/a_3 \approx 0.7904 \). In other words, the ratio of minor axis to major axis of the elliptical ring electrode is about 0.7904, which is consistent with Mindlin theory.

2.2.2. Verification in FEA Software. FEM commercial software is used to verify the theoretical method, and the numerical simulator should be able to include geometric features of different electrode shapes in modeling and meshing to better describe real devices. Now our numerical calculations are verified using a finite element simulation modeling method.

All of the devices considered in this study are AT-cut quartz resonators having base operating frequencies of 20 MHz. Additionally, the thickness of the quartz plate is selected as \( h = 0.08 \) mm, the radius of the quartz plate is 3.25 mm, and the electrode thickness is 100 nm. The whole geometry models of elliptical ring-type QCMs are meshed together with electrodes and quartz plates, as shown in Figure 2. In order for the geometric models to be clearly seen, the quartz slates and electrodes are magnified in the direction of thickness by a factor of 10 and a factor of 1000, respectively. The harmonic analysis can provide intuitive and effective information about the vibration distributions of QCMs with different electrodes. The thickness shear mode can be obtained from the simulation with the electrodes set to corresponding electric potentials.

For elliptical single-ring and elliptical double-ring electrodes, we divide them into three groups for comparison, denoted as group A, group B, and group C. Group A is an elliptical electrode, whose ratio of minor axis to major axis is 70%; group B is an elliptical electrode, whose ratio of minor axis to major axis is 0.8; and group C is an elliptical electrode, whose ratio of minor axis to major axis is 90%. The FEM results are shown in Table 1. The difference between the maximum displacement amplitudes of the \( x_1 \) axis and the \( x_3 \) axis represents the difference in the mass sensitivity distribution in the two directions, and it is recorded as \( \Delta \lambda \).

As shown in Table 1, the resonance frequency increases gradually because of the increment of the proportion of the major axis of the elliptical electrode beyond the minor axis as well as the reduction of the corresponding mass inertia. In the mass sensitivity distribution in Table 1, the black curves represent the vibration displacement distribution along the \( x_1 \) axis and the red curves represent the vibration distribution along the \( x_3 \) axis. From the mass sensitivity distribution of the elliptical single-ring and elliptical double-ring electrodes, it can be observed that the mass sensitivity distribution of the \( x_1 \) axis and the \( x_3 \) axis tends to be consistent when the ratio of minor axis to major axis is 0.8. Furthermore, the ratio of the major axis to the minor axis between the numerical calculation and the FEM simulation is closely equal, which means that the simulation model is correct and effective.

2.3. Effect of the Elliptical Ring Electrode. In this section, we conduct a numerical simulation to investigate the relationship between the mass sensitivity and the elliptical ring electrodes. The elliptical ring electrodes should be designed with the ratio of minor axis to major axis approximately equal to 0.8 according to the previous section. For the purpose of comparison, the resonator with circular ring electrodes having the same electrode parameters (e.g., quartz crystal radius and thickness) as elliptical ring electrodes is also fabricated.
2.3.1. Elliptical Single-Ring Electrode. Two typical resonator models, one with the circular single-ring electrode and one with the elliptical-ring electrode attached on the upper side, are simulated. Figure 3a,b shows the energy-trapping diagrams of QCMs with circular single-ring and elliptical double-ring electrodes for the fundamental mode. According to a large number of theoretical numerical calculations, the circular single-ring electrode parameters were adjusted to obtain the design parameters shown in Table 2. As shown in Figure 3a, it is obvious that there is a platform region of vibration amplitude in the center of the plate bounded by the circular single-ring electrode. The vibration in both directions is unevenly concentrated in the fully electrode region. As can be seen from Figure 3b, compared to the vibration of the circular single-ring electrode, the elliptical single-ring electrode is more consistent with the best trap effect and the vibration energy is evenly distributed throughout the electrode region. To observe the platform in the center of the plate more distinctly, we plot the normalized displacement distribution profiles along the x1 and x3 axis in Figure 3a,b. The vibration displacement distribution of the QCM with the circular single-ring electrode in the x1 direction is lower than that in the x3 direction, and the vibration distribution of the elliptical single-ring electrode is more uniform.

2.3.2. Elliptical Double-Ring Electrode. The mass sensitivity distribution of the double-ring electrode QCM is more uniform than that of other electrode structures. Therefore, an elliptical double-ring electrode is proposed for the QCM. According to our previous research results, the circular double-ring electrode design parameters are adjusted as shown in Table 3. As indicated in the energy-trapping diagrams of Figure 3c,d, the vibration distribution of the elliptical double-ring QCM is more uniform in all directions on the surface of the sensor compared with the circular double-ring QCM. It can be observed from the vibration displacement distribution profiles along the x1 and x3 axis in Figure 3c that the vibration displacement amplitude in the x1 direction of the outer ring region of the circular double-ring QCM is relatively high. However, in Figure 3d, the mass sensitivity distribution in the outer ring region of the elliptical double-ring QCM is uniform.

2.3.3. Comparison of Uniformity of the Mass Sensitivity Distribution. To visually reflect the uniformity of the mass sensitivity distribution of the four electrode structures, we use the coefficient of variation (\(c_v\)) of the node displacement data on the measured electrode surface to indicate the uniformity of the mass sensitivity of the electrode region. Table 4 shows the coefficient of variation of the four electrode structures. As can be seen from Table 4, the coefficients of variation of the elliptical ring electrode are smaller than those of the circular ring electrode. The variation coefficient of the elliptical double-ring electrode structure is the smallest among the four electrode structures. It can be concluded that the mass sensitivity of the elliptical ring electrode is more uniform than that of the circular ring electrode, and the uniformity of the mass sensitivity distribution of the elliptical double-ring electrode is better. These results can be obtained easily through the comparison between the coefficients of variation.

3. CONCLUSIONS

QCMs with elliptical single-ring and elliptical double-ring electrodes are presented based on theoretical analysis and FEM, through which the uneven vibration distribution phenomenon of the x1 and x3 axes on the surface of circular ring electrodes is solved. The mass sensitivity distributions of new types of electrodes are compared with those of circular ring electrodes. Our results show that the elliptical ring electrodes improve the uniformity of the mass sensitivity in comparison to the circular ring electrodes, and it is recommended that the ratio of minor axis to major axis of the elliptical ring electrode should be 0.8. Moreover, the mass sensitivity distribution of the elliptical double-ring-type QCM is more uniform. The scheme is also substantiated with FEM simulation. This electrode design reveals a path for improving the QCM detection sensitivity distribution for applications where a more uniform mass sensitivity is desirable.

**ASSOCIATED CONTENT**

**Supporting Information**
The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsomega.1c04957. Numerical calculation of elliptical ring electrode size optimization (PDF)

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