Mass Sensitivity Analysis of a Newly Developed Quartz Crystal Microbalance with Ring-dot Electrode Configuration and Reduced Mass Loading Area

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Abstract

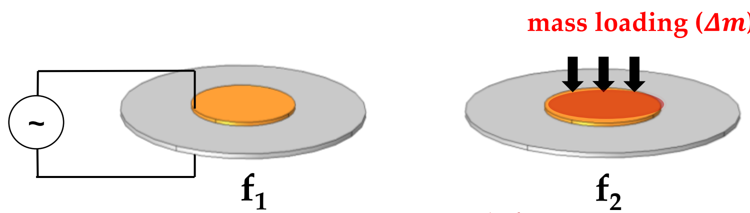
The conventional QCM design comprises a circular electrode configuration with an evenly distributed mass loading area. However, their mass sensitivity distribution is found to be non-uniform due to the inherent energy trapping effect. In this paper the recently developed QCM with a ring electrode and a ring-dot electrode configuration are evaluated. Finite Element Analysis is used to design and evaluate the conventional circular electrode QCM and the proposed ring electrode and ring-dot electrode QCM configurations, where the mass loading area is reduced up to 30% compared with the conventional sensor. Simulations are conducted to determine the sensor's resonant frequency shifts for an added mass per unit area of 20 μg/mm^2. The results indicate that newly designed ring and ring-dot electrode configurations operate at a higher resonant frequency. The observed frequency shift for the designed circular electrode, ring electrode, and ring-dot electrode configurations on a 333 μm thick quartz substrate are 85 kHz, 82 kHz, and 83 kHz, respectively. It is shown that the ring electrode and new ring-dot electrode configurations achieve a higher resonant frequency and offer a comparable sensing performance despite comprising of up to 30% reduced mass loading area in comparison to the conventional circular electrode configuration.

Keywords: Resonant frequency; Frequency shift; Finite Element Analysis; Ring electrode; Ring-dot electrode

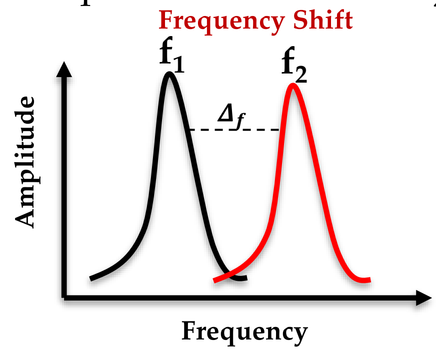
Graphical Abstract

Mass Sensitivity Analysis of a Newly Developed Quartz Crystal Microbalance with Ring-dot Electrode Configuration and Reduced Mass Loading Area

On applying AC voltage, the QCM oscillates at its resonant frequency due to the piezoelectric property



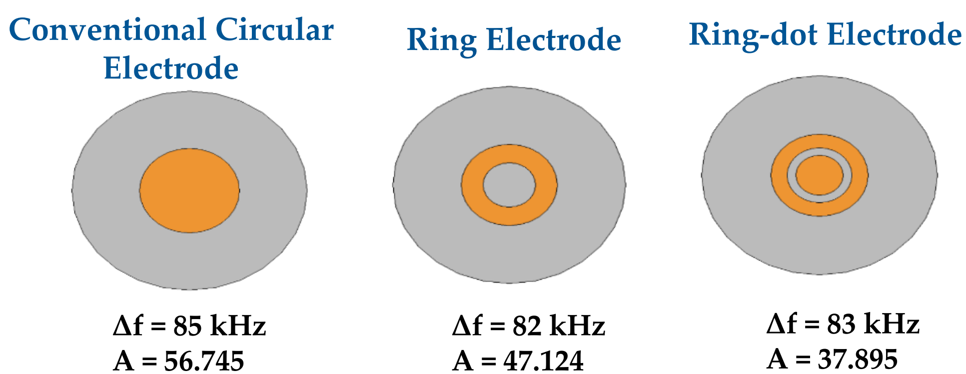
The size of the mass loading area (A) influences mass loading on the QCM, thus affecting the frequency shift (Δf) and the device sensing performance.



Mass loading on a QCM causes a shift in the resonant frequency. Thus, the sensing performance can be determined by frequency shift using the relation:

Frequency shift ∝ added mass

$$\Delta f \propto \Delta m$$



Objective

- To design a single channel Quartz Crystal Microbalance with conventional circular electrode, ring electrode and the recently developed ring-dot electrode configuration and simulate its resonant frequency and frequency shift for an added mass using Multiphysics simulations.

- To investigate the influence of mass loading area on frequency shift of the QCM in order to potentially improve the device mass sensitivity while utilizing a reduced mass loading area.

- To analyze the influence of the electrode dimensions of the ring electrode and the new ring-dot electrode configuration to achieve a higher resonant frequency in comparison to the conventional design, and potentially improve the device sensing performance for a reduced mass loading area.

Introduction to Quartz Crystal Microbalance Gas Sensors

Quartz Crystal Microbalance (QCM) is a micromachined gas sensor that can be utilized for measuring microgram level mass changes on the basis of change in frequency.

- It operates on the principle of the piezoelectric effect and comprises of a quartz crystal sandwiched between a top sensing electrode and a bottom reference electrode.

- Gold, Platinum and Silver are widely employed as metal electrodes for the QCM based on their high conductivity and inertness property.

- A sensing material is coated on the top sensing electrode which acts as a mass loading area in order to detect analytes.

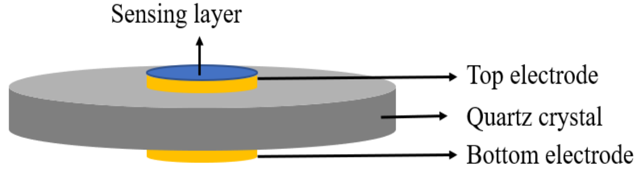


Fig. 1. Quartz Crystal Microbalance Structure (not to scale)

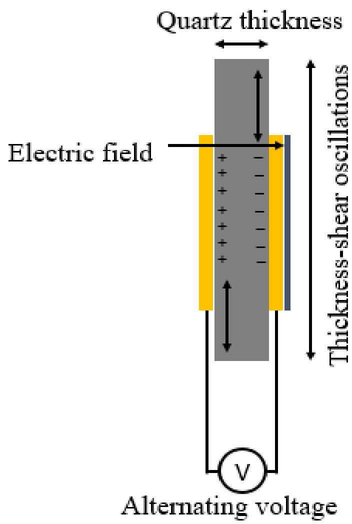


Fig. 2. Quartz Crystal Microbalance operating principle

Principle of Operation of QCM Gas Sensors

- Alternating voltage is applied to the QCM electrodes which creates an electric field thereby inducing a stress in the crystal which causes mechanical vibration or oscillation.

- AT cut quartz crystals vibrate in an anti-parallel fashion and hence produce thickness shear oscillations. This ensures frequency stability over a temperature range of -45 to 50°C and thus, can be used for detection in room temperature.

- The vibrational amplitude of the quartz is higher near the centre of the crystal and reduces in a gaussian pattern towards the edges. This is due to the energy trapping effect.

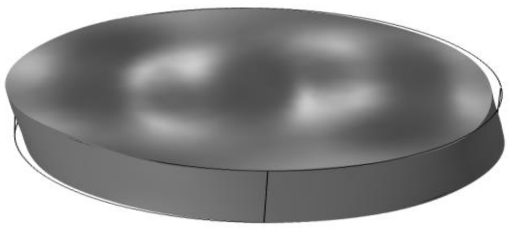


Fig.3. Thickness-shear deformation of a quartz crystal

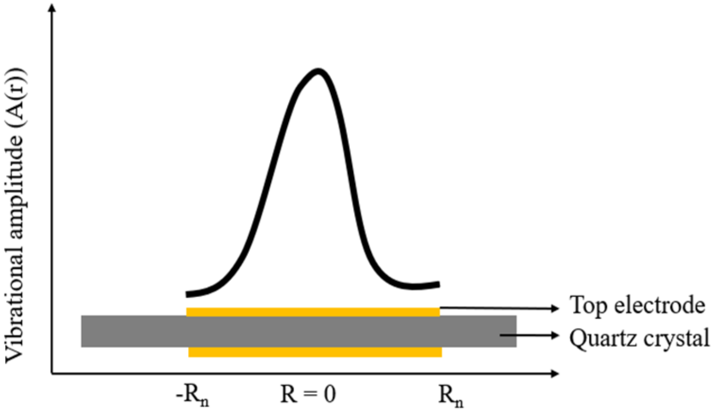


Fig.4. Energy trapping effect of QCM

Principle of Operation of QCM Gas Sensors

- The presence of the analyte on the mass loading area causes a change in the resonant frequency of the crystal as a result of additional stress. This change in frequency due to an added mass is utilized to identify the analyte.

- The change in the resonant frequency of the crystal due to mass deposition gives rise to a relation between mass and frequency which is governed by the Saurebrey equation,

$$\boldsymbol{\Delta_f = \frac{-2f_0^2}{A\sqrt{p_q\mu_q}}\Delta m}$$

- Δ_f = Change in frequency
- f_0 = Resonant frequency
- Δm = Change in mass
- A = Area of piezoelectric material
- p_q = Quartz crystal density
- μ_q = Shear modulus of quartz crystal

- The sensitivity of a device is the ability to detect small changes that are observed in its application. For QCM mass sensors, it is the ability to detect surface mass changes by the resonant frequency shift (Δf).

Resonant Frequency Shift due to an Added Mass

- COMSOL Multiphysics 5.5 is utilized to build and simulate the resonant frequency of a QCM in the 3D space dimension.

- Resonant frequency of QCM is simulated and obtained by Adaptive frequency sweep. The frequency where maximum displacement is observed denotes the resonant frequency.

- In this work the frequency shift (Δf) for an added mass (Δm) is utilized to determine the device's sensing performance.

$$\boldsymbol{\Delta f \propto \Delta m}$$

- The 'added mass' entity of COMSOL is used to define a mass on the mass loading area of the QCM based on the principle,

$$\boldsymbol{F_A = -\rho_A\left(-\omega^2 u + a_f\right)}$$

- The force 'F_A' is mass acting upon the selected surface 'A', 'ρ_A' is the mass per unit area, 'ω' is the angular frequency, 'u' is the displacement and 'a_f' is external force contribution such as gravitational acceleration.

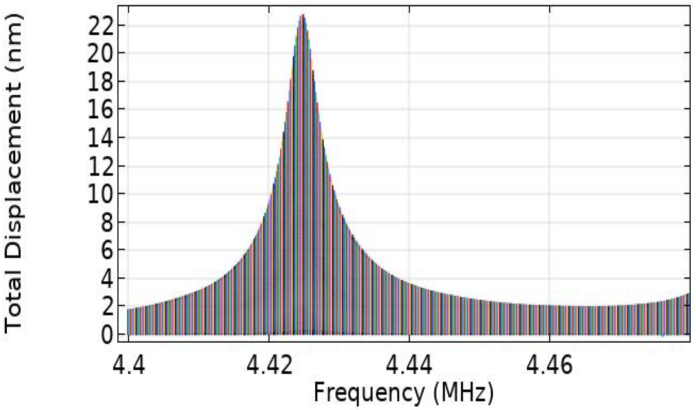


Fig.6. Line plot of Resonant Frequency of QCM

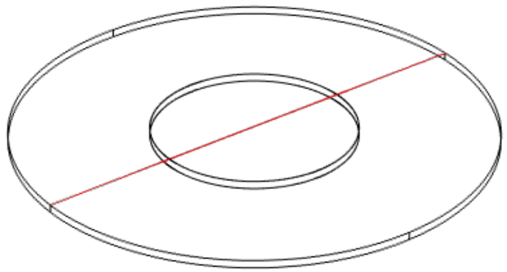


Fig.7. COMSOL 3D cut-line plot

Conventional Circular Electrode Configuration

- The circular electrode configuration is the conventional QCM electrode configuration.
- In this design, the entire top electrode is utilized for mass loading, thereby ensuring a higher frequency shift. However, it also consists of an unevenly distributed mass sensitivity due to energy trapping.
- The parameters used to build the circular electrode QCM are mentioned in Table 1.

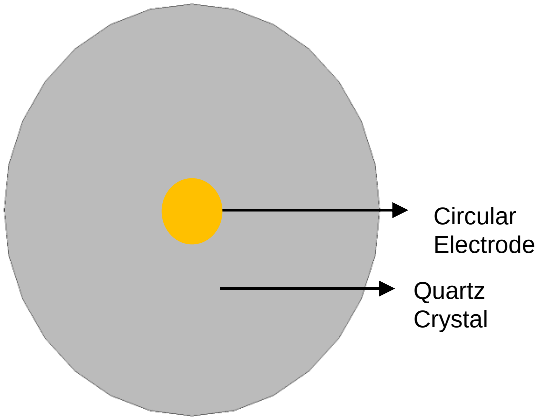


Fig.8. Conventional circular electrode configuration

<u>Design Parameters:</u>

Parameter	Value
Oscillator radius	10 [mm]
Oscillator thickness	333 [μm]
Dot electrode radius	4.25 [mm]
Electrode thickness	300 [nm]
Alternating voltage	10 [V]

Table 1. Conventional circular electrode QCM Design parameters

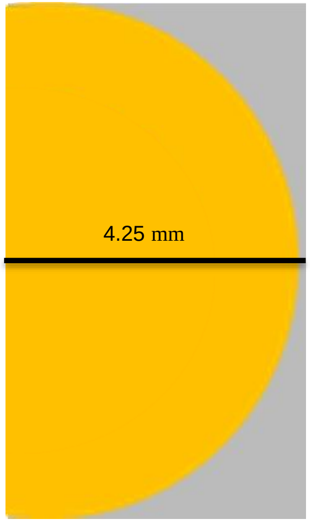


Fig.9. Conventional circular electrode radius

Conventional Circular Electrode Design

- The adaptive frequency sweep is performed to determine the resonant frequency and frequency shift of the circular electrode QCM.
- The simulation results indicate that the conventional circular electrode QCM achieved a frequency shift of 85 kHz for an added mass of 20 μg/mm^2.

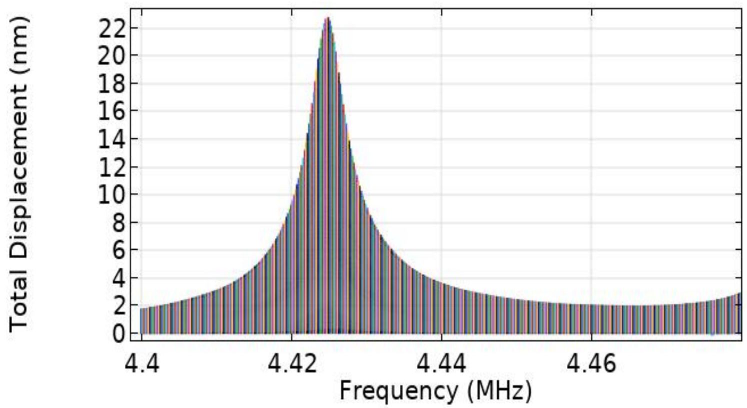


Fig.10. Resonant frequency of QCM for 0 added mass.

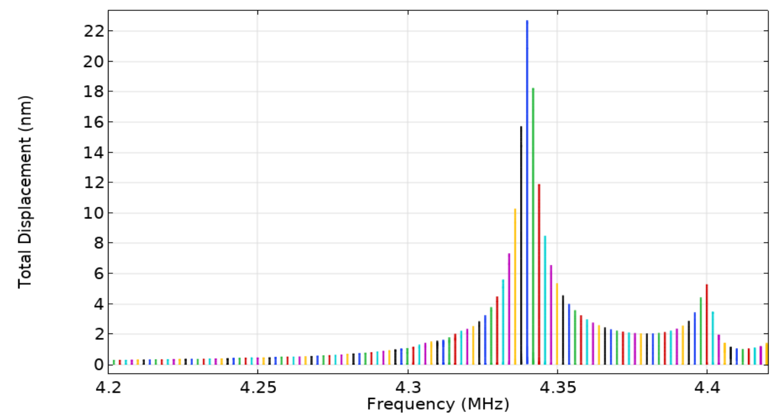


Fig.11. Resonant frequency of QCM for 200 μg added mass.

Electrode Configuration	Mass Loading Area (mm^2)	Frequency for 0 added mass	Frequency for 20 μg/mm^2 added mass	Frequency Shift (Δf)
Circular Electrode	56.745	4.425 MHz	4.340 MHz	85 kHz

Table 2. Conventional circular electrode QCM simulation result

Analysis of Effect of Mass Loading Area

- Utilizing the QCM with conventional circular electrode configuration, an analysis is performed to determine the effect of mass loading area on the frequency shift (Δf) of the QCM.
- The mass loading area is specified as a boundary on top of the top electrode representing a polymer layer of null thickness (no height).
- The mass loading area of the QCM is varied from values of 0.85 mm to 4.25 mm for an interval of 0.425 mm, which corresponds to a range of 10 % to 100 % coverage of the entire top electrode.
- The resonant frequency shift due to an added mass of 20 μg/mm^2 on the top electrode is determined for the different mass loading area.

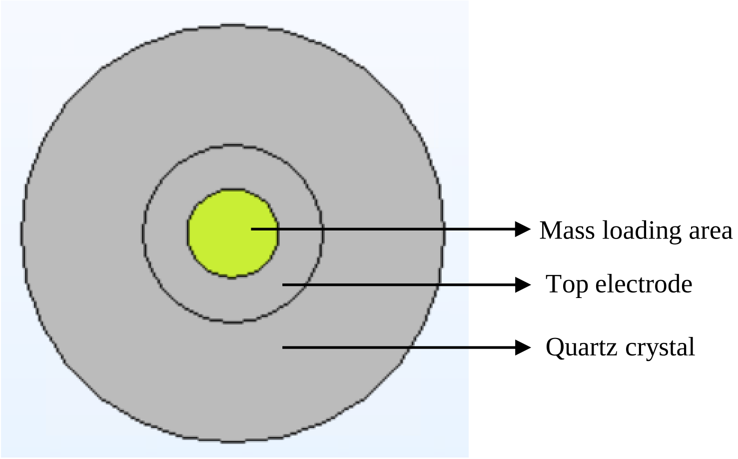
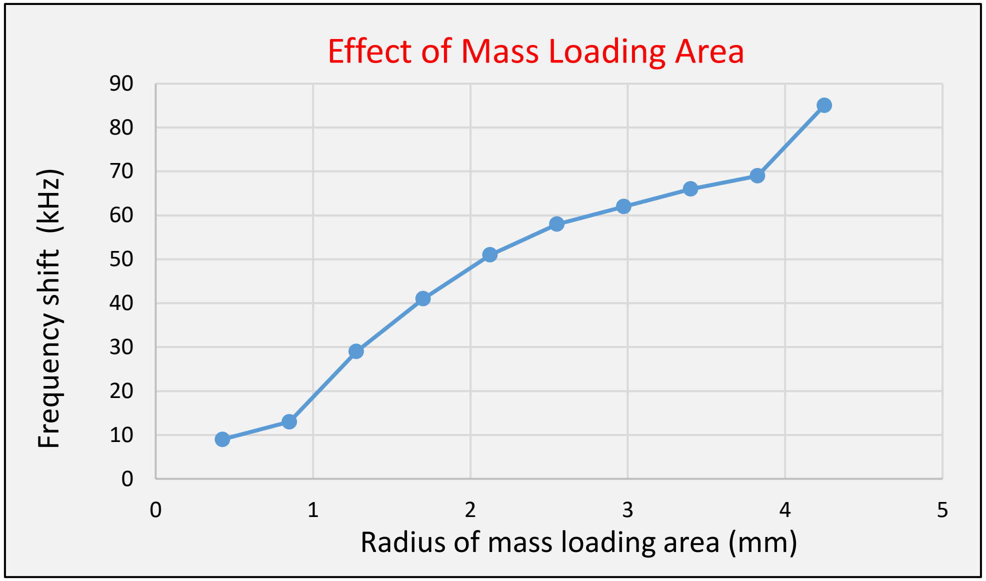


Fig.12. QCM top electrode mass loading area

Analysis of Effect of Mass Loading Area



- The simulation results indicate that a gradual increase in frequency shift is observed for radii values up to 2.125 mm after which the frequency shift decreases. This confirms the presence of energy trapping in the device.

- The frequency shift also increases towards the edge of the electrode from 3.75 to 4.25 mm radius of mass loading area.

- The analysis results provides a scope for modifying the QCM electrode configuration to potentially achieve a higher frequency shift for a lesser mass loading area by selectively electroding the QCM surface.

Ring Electrode Configuration

- The ring electrode design is proposed to improve the mass sensitivity distribution across the QCM electrode by reconfiguring the electrode area, and potentially achieving a higher resonant frequency.

- The parameters used to build the QCM is based on a conventional 5 MHz circular QCM and are mentioned in the table 3. The outer electrode radius is fixed at 4.25 mm for fair comparison with the circular electrode design.

Design Parameters:

Parameter	Value
Oscillator radius	10 [mm]
Oscillator thickness	333 [μm]
Ring electrode width	0.75, 1.5, 2, 2.5 [mm]
Electrode thickness	300 [nm]
Alternating voltage	10 [V]

Table 3. Design parameters of Ring electrode configuration

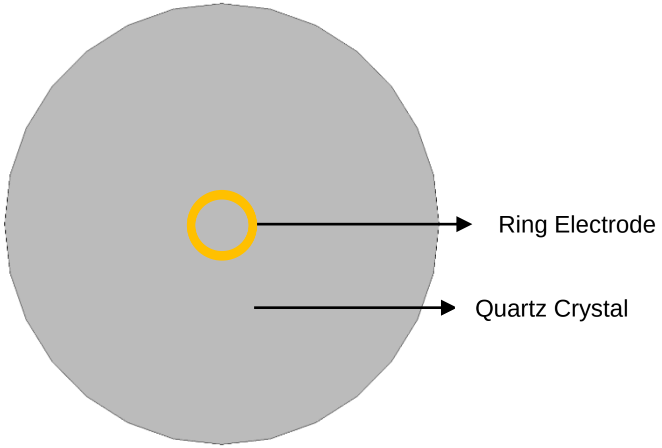


Fig.13. Ring electrode configuration

Ring Electrode Configuration – Effect of Ring width

- This analysis was performed in order to determine the effect of the ring electrode width on the frequency shift (Δf) of the QCM with ring electrode configuration.
- In this analysis, the ring electrode width was varied for different values ranging from 0.75 mm to 2.5 mm.
- The resonant frequency and frequency shift was determined for an added mass per unit area of 20 μg/mm².

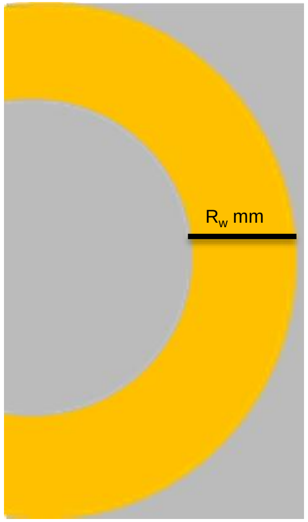
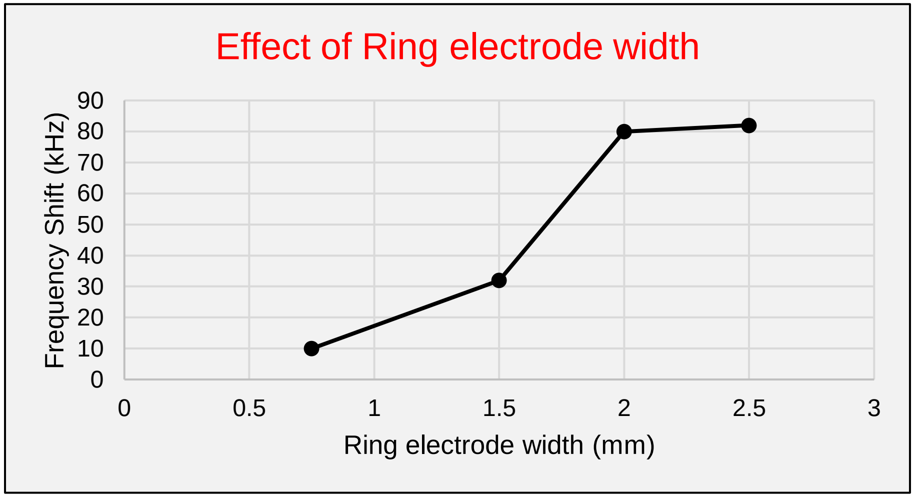


Fig.14. Ring electrode width

Ring width (R1)	Gap radius (G1)	Mass Loading Area (mm)²	Frequency for 0 μg/mm² mass	Frequency for 20 μg/mm² mass	Frequency Shift (Δf) (kHz)
0.75 mm	3.5 mm	12.567	4.742 MHz	4.732 MHz	10 kHz
1.5 mm	2.75 mm	32.987	4.526 MHz	4.494 MHz	32 kHz
2 mm	2.25 mm	40.841	4.49 MHz	4.41 MHz	80 kHz
2.5 mm	1.75 mm	47.124	4.466 MHz	4.384 MHz	82 kHz

Table 4. Simulation results of QCM with Ring electrode configuration

Ring Electrode – Effect of Ring width



- The simulation results indicate that the frequency shift increases with an increase in ring electrode width.
- A high frequency shift value of 80 kHz was achieved for a QCM with ring width 2 mm, while possessing 28% lesser mass loading area compared to the conventional circular electrode QCM.
- A higher frequency shift of 82 kHz was achieved for a QCM with ring width 2.5 mm, while possessing 18% lesser mass loading area compared to the conventional circular electrode QCM.

Ring-Dot Electrode Configuration

- The new Ring-Dot electrode configuration was proposed to improve the mass sensitivity distribution of the ring electrode, while ensuring a higher mass sensitivity by adding a central dot (circular) electrode.
- The parameters used to build the QCM is based on a conventional 5 MHz circular QCM and are mentioned in the table. The outer electrode radius is fixed at 4.25 mm for fair comparison with the circular electrode design.

Design Parameters:

Description	Value
Quartz radius	10 [mm]
Quartz thickness	333 [μm]
Dot electrode radius	0.5, 1.5, 2.5, 3.5 [mm]
Ring electrode width	0.5, 0.75, 1, 1.25, 1.5, [mm]
Electrode thickness	300 [nm]
Alternating voltage	10 [V]

Table 5. Design parameters of Ring electrode configuration

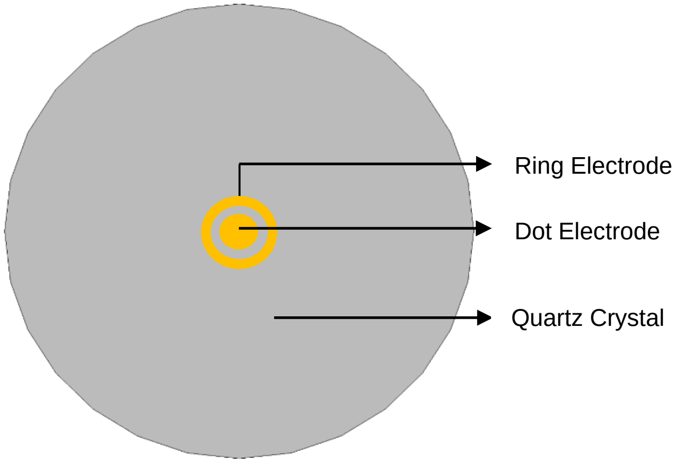


Fig.15. Ring-Dot electrode configuration

Ring-dot Electrode - Effect of Ring width

- This analysis was performed in order to determine the influence of the ring electrode width on the frequency shift (Δf) of the ring-dot electrode configuration.
- In this analysis, the dot electrode radius was kept constant at 5 mm, while the ring electrode width was varied from 0.5 to 1.5 mm by a step of 0.25 mm.
- The observed simulation results are depicted in Table 6.

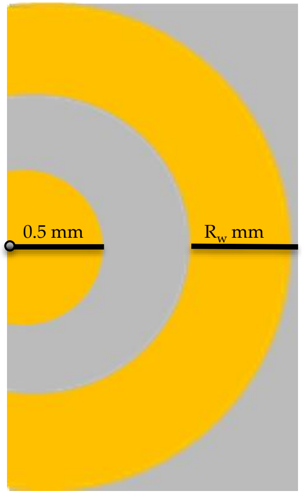
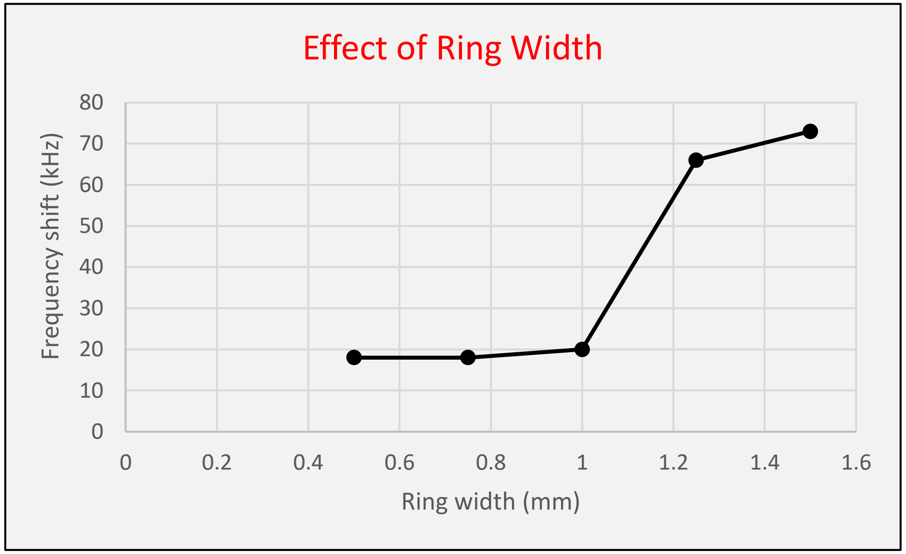


Fig.16. Ring-Dot electrode separation width

Ring width (R1)	Gap width (G1)	Mass Loading Area $(mm)^2$	Frequency for 0 $\mu g/mm^2$ mass	Frequency for 20 $\mu g/mm^2$ mass	Frequency Shift (Δf) (kHz)
0.5 mm	3 mm	13.352	4.673 MHz	4.655	18 kHz
0.75 mm	2.75 mm	19.046	4.816 MHz	4.798 MHz	18 kHz
1 mm	2.5 mm	24.347	4.815 MHz	4.795	20 kHz
1.25 mm	2.25 mm	29.256	4.596	4.53	66 kHz
1.5 mm	2 mm	33.772	4.566 MHz	4.493	73 kHz

Table 6. Simulation results of QCM with Ring-dot electrode configuration

Ring-dot Electrode - Effect of Ring width



- The simulation results indicate that the frequency shift increases with an increase in ring electrode width, while fixing the dot electrode radius. A steep increase in frequency shift is observed when increasing the ring electrode width from 1 mm to 1.25 mm.

- A higher frequency shift of 73 kHz is achieved for a QCM with ring electrode width of 2.5 mm, while fixing the dot electrode radius at 0.5 mm.

Ring-dot Electrode - Effect of Dot radius

- This analysis was performed in order to determine the effect of the dot electrode radius on the frequency shift (Δf) of the ring-dot electrode configuration.
- The resonant frequency and frequency shift was determined for an added mass per unit area of 20 µg/mm^2.
- In this analysis, the ring electrode width was kept constant at 0.75 mm, while the dot electrode width was varied from 0.5 to 3.5 mm.

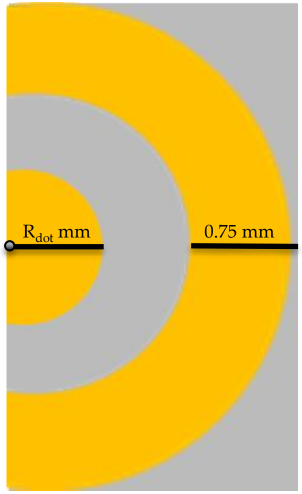
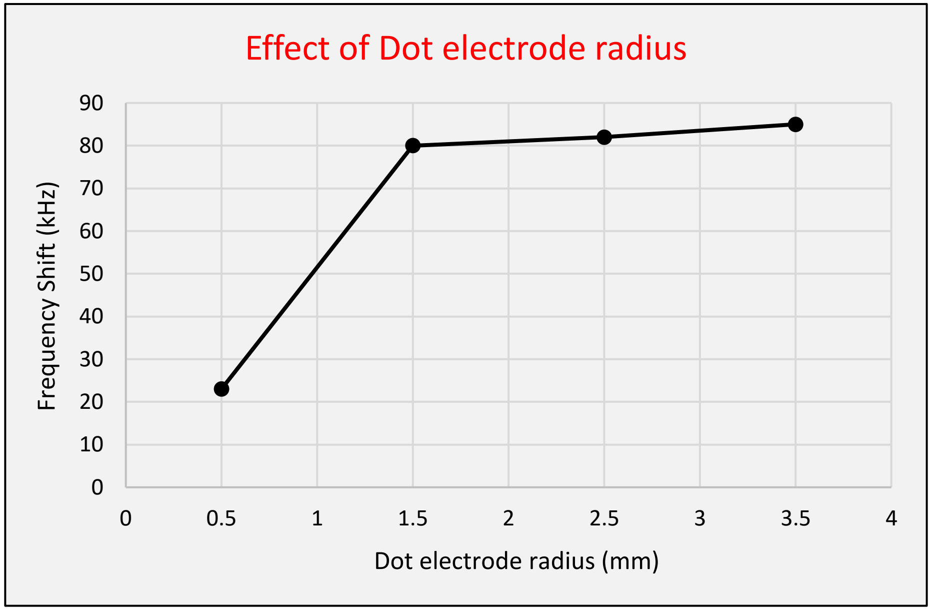


Fig.17. Ring-Dot electrode separation width

Dot electrode radius	Mass Loading Area (mm^2)	Frequency for 0 µg/mm^2 mass	Frequency for 20 µg/mm^2 mass	Frequency Shift (Δf) (kHz)
0.5 mm	19.046	4.821 MHz	4.798	23 kHz
1.5 mm	25.329	4.504 MHz	4.424	80 kHz
2.5 mm	37.895	4.45 MHz	4.367	83 kHz
3.5 mm	56.745	4.425 MHz	4.430	85 kHz

Table 7. Simulation results of QCM with Ring-dot electrode configuration

Ring-dot Electrode - Effect of Dot radius



- The simulation results indicate that the frequency shift increases with an increased dot electrode radius. A sharp increase in frequency shift is observed when increasing the dot electrode radius value from 0.5 mm to 1.5 mm.

- A higher frequency shift of 83 kHz is achieved for a QCM with ring-dot electrode of dot electrode radius 2.5 mm, while possessing 33% lesser mass loading area compared to the conventional circular electrode QCM.

Summary

Electrode Configuration	Mass Loading Area (mm^2)	Frequency for 0 $\mu g/mm^2$ mass	Frequency for 20 $\mu g/mm^2$ mass	Frequency Shift (Δf)
Conventional circular electrode	56.745	4.425 MHz	4.430 MHz	85 kHz
Ring electrode	47.124	4.466 MHz	4.384 MHz	82 kHz
Ring-dot electrode	37.895	4.45 MHz	4.367 MHz	83 kHz

Table 7. Comparison of QCM electrode configurations

- The QCM with conventional circular electrode of 4.25 mm radius attained a resonant frequency of 4.425 MHz, and a frequency shift of 85 kHz.
- The QCM with ring electrode configuration attained a higher resonant frequency of 4.466 MHz and achieved a high frequency shift value of 82 kHz while possessing 17% lesser mass loading area compared to the conventional design.
- The QCM with the new ring-dot electrode configuration attained a higher resonant frequency of 4.45 MHz and a higher frequency shift of 83 kHz while possessing 33% lesser mass loading area compared to the conventional circular electrode QCM.

Conclusion

- In this work, quartz crystal microbalance (QCM) with a conventional circular electrode, ring electrode and the recently developed ring-dot electrode configuration were designed, investigated and simulated for mass sensitivity analysis using Multiphysics simulations.
- The influence of mass loading area on frequency shift of the QCM was analyzed. The results indicated that mass loading on specific areas of the QCM electrode such as distance of 2.125 mm from center, and near the edges of the electrode can potentially improve the device sensing performance while utilizing reduced mass loading area. Based on this the ring electrode and the new ring-dot electrode configuration are built and investigated.
- The simulated ring electrode and the recently developed ring-dot electrode configurations achieve a higher resonant frequency and comparable frequency shift for an 17 to 30% reduced mass loading, when compared to the conventional design. This provides a scope to improve the mass sensitivity of the QCM by modifying parameters such ass the ring and dot electrode width and radius.

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Acknowledgements

The authors acknowledge the support by the Natural Sciences and Engineering Research Council of Canada (NSERC), Ontario Greenhouse Vegetable Growers (OGVG), CMC Microsystems, and the University of Windsor.