A High Frequency Dual Inverted Mesa QCM Sensor Array With Concentric Electrodes

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ABSTRACT Quartz Crystal Microbalance (QCM) is a candidate technology for high sensitivity application. This mass sensor’s property depends on the piezoelectric property of the AT-cut quartz crystal. QCM has been widely employed for gas detection due to its comparative advantages including high mass sensitivity, potential for array configuration, low cost, ease of fabrication as well as wide range of available sensitive material compatibility. However, its application has been limited due to two main disadvantages of the non-uniform mass sensitivity across the electrodes and lower frequency of operation. In this work, in order to overcome these disadvantages, a novel concentric electrode structure combined with the dual inverted mesa structure has been proposed and implemented. It is demonstrated that the developed and optimized concentric electrode has provided a uniform displacement across the QCM’s sensing electrode. In addition, the dual inverted mesa design has been implemented in the QCM array in which a high fundamental resonant frequency of 33 MHz has been achieved without interference between the adjacent channels in the sensor array. The interference between the adjacent high frequency QCM channels has been eliminated and therefore, each QCM can function as an individual gas sensor when coated with the designed sensing layers. For 33 MHz, the interference has been eliminated at the optimal center to center between electrode distance \( c_{2c} \). In the case of 10 MHz array, \( c_{2c} \) value of 6 mm is achieved which is lower than the value of \( c_{2c} \) achieved on a traditionally un-etched QCM array which was found equal to 6.5 mm. Therefore, the proposed QCM array design is shown to reduce the overall size while enhancing the device sensitivity, uniformity and performance.

INDEX TERMS Array configuration, concentric electrode, dual inverted mesa structure, frequency interference, high frequency, micromachining, piezoelectric resonator, quartz crystal microbalance, QCM, uniform displacement profile.

I. INTRODUCTION
Quartz crystal microbalance (QCM) is a highly sensitive mass sensor which can detect nano gram level of mass variations [1], [2]. The QCM has been employed in a wide range of low concentration gas detecting applications including detection of environmental pollutants [3], [4], biomarkers in human breath [5], aroma in food [6], poisonous gases [7] and chemical vapors [8], [9]. This is due to their advantage of simple structure and low cost coupled with high mass sensitivity. The conventional QCM sensor geometry consists of AT – cut quartz crystal sandwiched between two metallic electrodes [10]. The sensor operates based on the inverse piezoelectric property of the quartz crystal [11]. Inverse piezoelectric effect can be defined as the property of the quartz crystal to undergo shear deformation when an electric potential is applied [12]. When this applied electric potential is alternated at a designed frequency, the crystal undergoes thickness shear mode of oscillation in the resonant frequency. However, the resonant frequency of the quartz crystal microbalance (QCM) increases or decreases based on the addition or removal of mass [10] as shown in Fig. 1. Thus, by measuring these variations in the resonant frequency using a high frequency oscillator, the amount of absorbed mass can be measured [13]. By adding a sensitive layer on top of the active sensing electrode, the QCM can be pivoted as a chemical gas sensor [14]. In this configuration, the mass variation is associated with the adsorption or desorption of the
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**FIGURE 1.** Quartz Crystal Microbalance (QCM) sensor showing the mass and frequency change relationship when the target gas comes into contact.

Gas molecules in the device sensing layer [15]. QCM offers several advantages compared to other gas sensors such as the simple design, potential high sensitivity, low cost, ease of fabrication, and compatibility of array configuration to detect multiple targets in a complex environment [16], [17]. Despite the comparative advantages of QCM in chemical gas detection application, it suffers from two main disadvantages that limit the device application in low gas concentration applications.

In conventional QCM, the electrode structure influences the distribution of displacement and sensitivity profile. This results in a non-uniform sensitivity across the sensing electrode, and therefore, degrades the device performance [18]. In order to counteract this disadvantage, a new optimized electrode structure has been proposed in this paper called as the concentric electrode design in the section II and the corresponding displacement profile is compared against the conventional QCM electrode geometries.

In addition, the conventional QCM designs are commonly limited with 5 MHz – 10 MHz mode of operation that hinders the device performance due to the lower resonant frequency [19]. The difference between the high and low resonant frequencies QCM is shown in the Table 1. The 5 MHz – 10 MHz QCMs are common because of their mechanical stability during operation and fabrication by having a thickness between 168 μm to 333 μm. It is shown that a higher resonant frequency provides high mass sensitivity [20]. However, in order to increase the resonant frequency of QCM, the thickness of the quartz has to be decreased which makes it fragile and difficult to handle during the fabrication process. Moreover, when employing QCM sensors in an array configuration, a higher resonant frequency in a conventional array increases the overall size of the device due to the higher influence of the high resonant frequency QCM channels [21]. Thus, in order to counteract these disadvantages of low frequency operation, large size and fragility, the proposed concentric electrodes are employed in a novel dual inverted mesa QCM design. In this work, a high resonant frequency of 33 MHz has been achieved in an unconventional array configuration with the similar robustness and mechanical stability during array formation as a lower resonant frequency QCM in the section III. Finite element analysis has been conducted to design and optimize the array by eliminating the interference between the adjacent channels in order to provide independency in operation for each QCM channel in complex environment with a presence of multiple gases.

Although there are other research works of QCM with much higher frequencies has been published [22]–[24], they are commonly limited with single channel of operation. This is due to the fact that the multiple channels with high resonant frequency possess the challenge of removing the frequency interference between the adjacent channels in order to independently operate and sense the gas. Thus, this paper is aimed to design and demonstrate considerably high QCM frequency of operation at 33 MHz with negligible interference in multiple channels using dual inverted mesa structure in the simulation environment.

**II. ELECTRODE STRUCTURE AND DISPLACEMENT PROFILE**

During the past few decades, the mass sensitivity of the QCM has been analytically modeled using the Sauerbrey’s equation [21], [25]. However, the Sauerbrey’s equation does not consider the effect of shape, structure or the material of the electrode which makes the Sauerbrey’s equation unsuitable for mass sensitivity modeling of a complex QCM structure [26]. Moreover, the mass sensitivity of the QCM depends on the vibrational amplitude at the localized point on the piezoelectrically active area in which the displacement pattern on the conventional circular electrode on a QCM can be described as a Gaussian curve [27]. This Gaussian pattern of the displacement in due to the energy trapping effect of the quartz [28]. In this structure, the acoustic waves are trapped towards the center of the quartz. In other words, quartz act as an acoustic lens and trap the energy towards the center portion of the quartz and gradually reduces in the form of a wave [25]. This distribution of displacement on the QCM can be analytically described as the Bessel equation as follows,

$$S_f (r, \Theta) = \frac{|A (r, \Theta)|^2}{2\pi \int_0^\infty r |A (r, \Theta)|^2 dr} \times C_f$$  \hspace{1cm} (1)
where, $S_f (r, \theta)$ is the mass sensitivity function with a unit of Hz/Kg, $C_f$ is the Sauerbrey’s sensitivity constant with a value of $1.78 \times 10^{11}$ Hz.cm$^2$/kg, $A_f (r, \theta)$ is the particle displacement amplitude function and $r$ is the distance from the center. The particle displacement amplitude function $A(r)$ is the solution of the following Bessel equation,

$$r^2 \frac{\partial^2 A}{\partial r^2} + r \frac{\partial A}{\partial r} + \frac{K_i^2 r^2}{N^2} A = 0$$  \hspace{1cm} (2)

where, $N = 2.0443$ according to the materials constant of the AT-cut quartz crystal, $K_i^2 = (\omega_i^2 - \omega_0^2)/c^2$, where $i = E, P, U$ ($E, P$ and $U$ represents the full electrode region, partial electrode region and non-electrode region, respectively), $c = \sqrt{c_{66}/\rho_q}$ is the acoustic wave velocity in the crystal, where $c_{66}$ is the elastic stiffness constant, $\rho_q$ is the density of the quartz, $\omega_0$ is the cut off frequency of full electrode region $\omega_f$, partial electrode region $\omega_p$ and non-electrode region $\omega_i$.

In order to evaluate the Gaussian distribution of displacement in a circular electrode and to further customize the electrode structure for a uniform displacement profile, COMSOL Multiphysics software [29] is used for finite element analysis and modelling. The inbuilt kernel of the COMSOL is used to design the QCM geometry. The boundary parameters are predefined for easy access. Then, the quartz crystal is designed with gold metallic electrodes on the top and bottom. The AT cut quartz and gold materials are assigned with the inbuilt COMSOL materials. Followed by this step, the solid mechanics physics is used to define the boundary conditions. The boundary conditions are set to free state in order to avoid damping effect on the displacement profile. Then, the electrostatics physics is used to apply the alternating potential on the gold electrodes. MEMS module in the COMSOL is used for multiphysics piezoelectric property in COMSOL. For meshing, due to the large difference in the thickness between quartz and gold layers, the physics controlled mesh takes longer simulation time. Thus, the quartz and the electrodes are meshed separately with user controlled mesh. Triangular mesh with swept across the z-axis is used for the electrodes and the quadrilateral mesh is used for the quartz. Finer meshing size is used for the whole geometry. Followed by meshing, adaptive frequency analysis in the COMSOL is conducted to obtain the frequency response and displacement data. Followed by this analysis, post processing of displacement data across the axis of electrode is carried out to plot the displacement profiles of the electrodes.

**A. CONVENTIONAL ELECTRODES IN QCM**

A common electrode geometry used in the QCM is the circular electrode [30]. Ring and ring dot electrodes came as the alternatives for the circular electrode to reduce the energy trapping effect of quartz and provide uniform displacement [31]. However, these two electrodes have low mass loading area compared to the circular electrodes and lack uniformity in the distribution of displacement.

### TABLE 2. Dimensions of circular, ring and ring dot electrodes.

| Parameter                          | Dimension | Unit |
|------------------------------------|-----------|------|
| Radius of circular electrode       | 4.25      | mm   |
| Radius of ring electrode           | 4.25      | mm   |
| Radius of ring dot electrode       | 4.25      | mm   |
| Ring width of ring electrode       | 2.125     | mm   |
| Ring width of ring dot electrode   | 2.125     | mm   |
| Radius of dot in ring dot electrode| 500       | \mu m |
| Radius of quartz crystal           | 12        | mm   |

The geometrical design of the circular, ring and ring dot electrodes are shown in Fig. 2. These three different electrode geometries are designed in the COMSOL using its inbuilt kernel as per the dimensions given in the Table 2. The overall size of these electrodes and the radius of quartz crystal are kept constant to have a fair comparison of their displacement patterns. In order to obtain their displacement patterns, the adaptive frequency study has been performed in the finite element analysis tool and the displacement patterns of each electrode are compared in the results shown in Fig. 2. Fig. 2 illustrates that the displacement profile of the conventional circular, ring and ring dot electrode in which the circular electrode has a Gaussian curve pattern of displacement across the axis of electrode. The vibrational amplitude is high at the center of the circular electrode and gradually decreases when the distance between the center of the electrode increases. Owing to this non uniform distribution profile of the conventional circular electrode operation, the frequency shift...
caused in the QCM during the target gas interaction is lower compared to the frequency shift caused by an electrode with uniform displacement profile.

Subsequently, the ring electrode exhibited a higher displacement towards the edge of the electrode and lower displacement towards the center of electrode. Despite being more uniform than the circular electrode, the ring electrode have comparatively low mass loading area than circular electrode. Thus, in order to increase the mass loading area and uniformity in displacement pattern parallelly, ring dot electrodes are proposed and their displacement patterns are compared with other electrodes as shown in the Fig. 2.

The ring dot electrode displayed more uniformity in the displacement profile than the ring electrode. The vibrational amplitude is observed to be higher on the center portion of the ring dot electrode compared to the ring electrode.

Thus, from the following displacement profile investigation, the displacement profile exhibited by the ring dot electrode is found to be both uniform and has a comparatively high mass loading area than the other two electrodes. Therefore, the ring dot pattern is chosen for further customization to achieve the desired uniformity.

**B. ASCENDING RING WIDTH PATTERN**

Despite the uniformity in displacement pattern and increased mass loading area of the ring dot electrode, it requires much more uniformity and higher vibrational amplitude. Thus, the ring dot electrode is added and customized with an additional ring with the equal ring width pattern \((RW1 = RW2)\) as shown in Fig. 3. When the displacement profile of this equal width double ring dot electrode is analysed, it shows that the displacement is higher towards the center of the electrode and decreases towards the outer ring which is similar to the displacement pattern of the conventional circular electrode.

Thus, in an unconventional way, the outermost ring width \((RW2)\) is increased in an ascending pattern in 1:1.5 ratio as shown in the Fig. 3. The dimension of the ascending ring width electrode is given in Table 3. This ascending order ring width pattern \((RW1 < RW2)\) results in an increase in the displacement amplitude of 7 nm on the outer ring as shown in Fig. 4 which is comparatively higher than the displacement of 3 nm attained with the equal ring width. As explained before, the energy trapping effect of quartz increases the vibrational amplitude towards the center of the electrode, thus, the rings are designed in a way that it indirectly provides uniform displacement across the electrode surface. Therefore, the proposed ascending ring width pattern has increased the uniformity of displacement profile compared to the other electrodes such as the circular and single ring electrodes.

However, the double ring dot electrode with ascending ring width pattern has exhibited a dip in the vibrational amplitude between the gap areas of the electrode. Thus, in order to further improve the displacement uniformity, the dual rings are added with additional ring in the similar ascending ring width pattern and the ring gaps are customized and investigated in the next section.

**C. PROPOSED CONCENTRIC ELECTRODE DESIGN**

The double ring electrode in the ascending ring width pattern is added with one more ring on a ring-width ratio of 1:1.5:2. In the first approach, three rings are designed with equal ring gaps as shown in Fig. 5. The displacement pattern for this design is observed to in a way in which the majority of vibrational amplitude is contained within the 2 nm – 5 nm range as shown in Fig. 6.

However, the vibrational amplitude is gradually depleted as the number of rings is increased which might lower the overall sensitivity of the QCM. Thus, in order to counteract this depletion in terms of overall vibrational amplitude, the ring gaps are customized to be in the descending order pattern while the ring-width remains same in the ascending order pattern. This new descending ring-gap pattern in the ratio of 1:0.7:0.4 combined with the ascending ring width pattern with a ratio of 1:1.5:2 is named as the concentric electrode which is shown in Fig. 5.
FIGURE 5. Schematic illustration of ascending ring width with equal ring gap electrode [RW1 < RW2 < RW3, G1 = G2 = G3] and ascending ring width and descending ring gap electrode [RW1 < RW2 < RW3, G1 > G2 > G3].

FIGURE 6. Displacement profile (a) Ascending ring width with equal ring gap electrode [RW1 < RW2 < RW3, G1 = G2 = G3] (b) Ascending ring width and descending ring gap electrode [RW1 < RW2 < RW3, G1 > G2 > G3].

The displacement pattern of the concentric electrode has exhibited a minimal and maximal vibration amplitude range between 5 nm to 8 nm approximately without any steep dip in the vibrational amplitude. This close proximity between the minimal and maximal range of 5 nm to 8 nm with the high minimal vibrational amplitude of 5 nm demonstrates that the concentric electrode provides a higher shift in the resonant frequency when the target gas comes into contact on any part of the electrode surface. Owing to this uniform and high minimum vibrational amplitude achieved at the all electrode surface, the target gas that comes into contact with the electrode creates a higher level of shift in the frequency than a conventional electrode which has non uniform displacement profile. The dimensions of these two electrodes are given in the Table 4.

TABLE 4. Dimensions of ascending ring width electrode and concentric electrode.

| Parameter                  | Ascending Ring width (3 rings) | Ascending Ring Width and Descending Ring Gap (Concentric Electrode) |
|----------------------------|--------------------------------|---------------------------------------------------------------------|
| Ring width (RW1)           | 500 μm                         | 500 μm                                                              |
| Ring width (RW2)           | 750 μm                         | 750 μm                                                              |
| Ring width (RW3)           | 1 mm                           | 1 mm                                                               |
| Gap 1 (G1)                 | 500 μm                         | 500 μm                                                              |
| Gap 2 (G2)                 | 350 μm                         | 350 μm                                                              |
| Gap 3 (G3)                 | 200 μm                         | 200 μm                                                              |
| Radius of dot (RD)         | 500 μm                         | 500 μm                                                              |

The conventional circular electrode will provide a higher shift in the resonant frequency only towards the center part of the electrode, whereas, the newly developed concentric electrode provides the similar higher shift in the resonant frequency along the whole electrode surface. This is due to the influence of the customized electrode geometry that indirectly reduces the effect of energy trapping effect of quartz. The displacement profile comparison of the concentric electrode and conventional electrode with same overall electrode size is presented in Fig. 7. Fig. 7 shows that the concentric electrode has more overall uniformity in the distribution profile across the active electrode area in comparison to the conventional circular electrode. This overall increase in the vibrational amplitude of the concentric electrode increases the overall sensing efficiency of the sensor compared to the conventional circular electrode.

FIGURE 7. Displacement profile comparison of conventional circular electrode and concentric electrode in which the concentric electrode in QCM has a comparatively uniform displacement profile.

III. DUAL INVERTED MESA STRUCTURE

In QCM, the resonant frequency of the quartz crystal depends on the thickness of quartz at which the electrical excitation takes place [32]. The thickness of quartz and resonant frequency has an inversely proportional relationship between each other [11]. Owing to this relationship, the thickness of the quartz crystal needs to be reduced in order to increase the resonant frequency [33]. In addition, mass sensitivity of QCM increases exponentially with the increase in resonant frequency but has a downside of thinner quartz [34]. Thin quartz is difficult to handle during fabrication process and unsuitable for the practical applications such as electronic noses and other gas sensing applications.

Thus, the resonant frequency of the QCM has to be increased without sacrificing the robustness. In order to achieve this, a new customized design called the dual inverted mesa is proposed in this work. Then the latter designed concentric electrode is integrated. Dual inverted mesa QCM design can also be used to reduce the zone of influence of individual QCM channels in an array. This also leads to reduction in the overall size of the high frequency QCM array.
A. Robust and High Frequency- Single Channel QCM

In this dual inverted mesa QCM design, the edge of the quartz is kept relatively thick with a thickness of 333 µm while the inner cylindrical layer of quartz is reduced to 50 µm by etching and then coating with electrodes as shown in Fig. 8. This step thickness of 333 µm non vibrating quartz provides mechanical stability for the thin vibrating quartz of 50 µm during operation and handling. The 50 µm thickness of quartz has a resonant frequency of about 33 MHz which is mechanically supported by the six times thicker quartz support wall of 333 µm. As a result, there are possibilities for a mild increase in the energy trapping effect in this design. However, the concentric electrodes can effectively address the energy trapping effect and provide a more uniform displacement profile.

Thus, unlike conventional QCM sensor which is limited by low resonant frequency of operation, the dual inverted mesa QCM can operate at comparatively higher resonant frequency of 33 MHz and still provide mechanical stability similar to that of a conventional QCM design. Theoretically, the mass sensitivity of conventional 5 MHz QCM is 17.7 ng/cm²/Hz⁻¹ whereas the proposed 33 MHz QCM channels can individually provide a mass sensitivity of 0.4 ng/cm²/Hz⁻¹. This means that when a mass of 0.4 ng is adsorbed on the piezo electrically active crystal area of 1 cm², the resonant frequency of the oscillating quartz crystal reduces by 1 Hz. The level of detection (LOD) for a 0.1 Hz resolution is equal to 0.04 ng/cm²/Hz⁻¹ for a second.

In addition, this design can be even adapted to resonant frequencies above 33 MHz by reducing the thickness of the vibrating quartz and ensuring at least six times thickness of vibrating quartz to the non-vibrating quartz layer in order to effectively reduce the frequency interference.

B. Array Configuration – Dual Inverted Mesa Structure

In addition to the increased robustness and high resonant frequency, the dual inverted mesa structures can also be applied on the array configuration in order to minimize the zone of influence of each QCM channel in an array and subsequently reduce the size of high frequency QCM array.

However, due to the monolithic quartz operation, the multiple QCM channels needs to be carefully placed at a distance from between other in order to avoid mechanical interference between the adjacent channels. This mechanical interference causes error in the frequency response of the neighboring channels which affects the final sensor output [35]. Thus, with an optimal distance between the channels, independency on each QCM channel without interferences can be gained and each QCM channel acts as an individual sensor on the monolithic quartz substrate. This enables multiple gas targets detection without interference.

Therefore, in order to achieve the minimum optimal distance between the adjacent channels on the monolithic quartz substrate, two QCM channels with dual inverted mesa structures are designed and placed adjacent to each other with a center to center distance (c2c) as shown in Fig. 9. The c2c is set to 4.5 mm and gradually increased with a step size of 1 mm. The device frequency response characteristics is simulated and investigated for each iteration to monitor the resonant frequency on both channels. The optimal c2c value for 33 MHz and 10 MHz with a vibrating thickness (VT) of 50 µm and 168 µm is obtained as 13 mm and 6 mm as shown in Fig. 10 and Fig. 11.

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The resonant frequency below the optimal c2c distance tends to mismatch between each other and gradually becomes...
Similar when it reaches the optimal \( c_2c \) distance. This is the point at which the zone of influence of adjacent QCM does not overlap. Thus, the minimal optimum distance between the center to center of electrodes \( (c_2c) \) ensures that absence of interference between the adjacent channels. Therefore, multiple QCM channels can operate as individual sensor on a same monolithic quartz substrate by maintaining this optimal \( c_2c \) distance.

This article builds on the previous work and research thesis that are also cited. In our previous works [36],[37], we have investigated the optimal \( c_2c \) value for a conventional 5 MHz un-etched QCM array in which we have achieved a \( c_2c \) value of 6.5 mm for a 5 MHz QCM array. The \( c_2c \) value between adjacent channels increases as the resonant frequency is increased. However, with the proposed dual inverted mesa structure and concentric electrodes, the optimal \( c_2c \) value for a 10 MHz array is achieved at 6 mm which is lower than 5 MHz QCM array. Thus, a comparatively high resonant frequency has been achieved on multiple QCM channels without interference on a minimalistic monolithic quartz wafer.

Furthermore, as a result, this new high frequency 33 MHz dual inverted mesa QCM with multiple channels provides a platform to detect multiple gases simultaneously in a complex environment with high mass sensitivity than the conventional QCM designs. For instance, if two target gases have to be detected in a multiple gas environment, the proposed sensor can be coated with the suitable sensing materials and can be employed to detect their concentration levels without inference from the neighboring channels. The proposed gas sensor has advantages including high mass sensitivity, low voltage requirement, array operation, and simple structure at comparatively low cost. In comparison, the other gas sensors in its range are limited with disadvantages including comparatively high cost, high input voltage requirement, array incompatibility, complex design and fabrication.

### IV. CONCLUSION

Quartz Crystal Microbalance (QCM) sensor is commonly limited within 5 MHz – 10 MHz of operation and with a non-uniform displacement profile. With the proposed novel concentric electrode, the non-uniform displacement profile that leads to uneven distribution of the mass sensitivity has been made comparatively uniform. Thus, compared to the conventional circular electrodes in the QCM, the sensing efficiency has been effectively improved in the proposed concentric electrodes.

Furthermore, by employing the dual inverted mesa structures in the monolithic quartz crystal, high frequency of 33 MHz has been achieved with an optimal \( c_2c \) value of 13 mm between the adjacent channels without interference. Significantly, this increase in resonant frequency has been achieved without sacrificing the robustness of the sensor for better handling and operation. The conventional circular electrode has a higher energy trapping effect on the dual inverted mesa structure due to strong quartz mount. This energy trapping has been effectively managed with the proposed concentric electrode.

Therefore, two of the major disadvantages of the QCM sensor have been counteracted in this work. Thus, the developed high frequency dual inverted mesa QCM array can be employed to detect the presence or concentration levels of multiple gas targets with high sensitivity and uniform displacement profile across the sensing electrode.

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