Design of an efficient Quartz Crystal Nanobalance: a Finite Element Study

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Abstract

The mass sensitivity (> 1 micrograms) of current commercial analytical instruments, such as thermal gravimetric analyzers, severely limits their utility for measurement of valuable but poorly soluble materials such as synthetic proteins or DNA fragments. A quartz crystal microbalance (QCM), based on a transverse shear mode piezoelectric crystal operating at high frequencies, is gaining popularity in chemical and bio-sensing applications due to higher mass sensitivities as compared to the traditional analyzers and lesser sensitivity to vibrations. However, these devices suffer from non-uniformity of sensitivity distribution along the sensor surface thereby limiting their use for the determination of mass. Overcoming this limitation would lead to the development of a robust sensor with improved mass sensitivities and reduced sensitivity to vibrations, as compared to the currently available microbalances. The sensitivity profile can be influenced by a number of factors the electrode design and surface properties of the crystal. In the current work, we develop a finite element (FE) model of the QCM to investigate the mass sensitivity and its radial distribution on the sensor surface for various electrode designs. Such a model will aid in the development of versatile nano-balances with a uniform sensitivity distribution.

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Introduction

A quartz crystal nanobalance (QCN), based on a transverse shear mode piezoelectric crystal operating at high frequencies, is gaining popularity in chemical and bio-sensing [1] applications due to higher mass sensitivities as compared to the traditional analyzers and lesser sensitivity to vibrations. One potential use of a QCM is as a 'droplet gravimeter' which can measure the mass of non volatile residues (NVR) in a droplet of solvent [2]. The transverse shear mode QCM device can provide nanogram level sensitivity thereby enabling detection limits of parts per million. However, these devices suffer from non-uniformity of sensitivity distribution over the sensor surface thereby limiting their use for the determination of mass. Overcoming this limitation would lead to the development of a robust 'nanobalance' with improved mass sensitivity and increased frequency stability, as compared to the currently available microbalances. This can potentially revolutionize the mass measurement systems, in applications such as droplet gravimetry. The sensitivity profile of a QCN can be influenced by a number of factors such as the electrode design, size, and surface properties of the crystal. It has been established the mass sensitivity decays with distance from the center of the electrode. Controlling this sensitivity variation to achieve a constant sensitivity across the sensor surface, so that the mass sensitivity is independent of the placement of mass, will result in an efficient nanobalance.

Mathematical modeling provides an efficient and cost-effective way to investigate the performance of QCN devices before an actual device is fabricated [3, 4]. The finite element technique provides the ability to accurately model complex structures, and to optimize design parameters. In the current work, we develop a finite element (FE) model of the QCN to investigate the mass sensitivity and its radial distribution on the sensor surface for various electrode designs. Such a model will aid the development of a versatile nanobalance with a uniform sensitivity distribution. The same model can be extended to study fluid-device interaction by utilizing a full bidirectional coupling with the fluid domain, when the device is used in liquid-sensing applications.

1. Computational details:

An AT-cut quartz crystal, cut at 35.35 degrees to the z-axis, is a well known substrate for use as quartz crystal microbalance (QCM) due to its high sensitivity [5]. The QCN device (operating frequency 9 MHz) was also based on an AT-cut quartz crystal, cut at 35.35 degrees to the z-axis, with gold electrodes on the top and bottom surfaces. The device was 8.0 mm diameter and 0.185 mm thick with 5.0 mm diameter gold electrodes having a thickness of $0.15*10^{-3}$ mm on either side of the QCN. The density of gold and QCN were 18500 kg/m^3 and 2649 kg/m^3, respectively. These dimensions and properties are summarized in Table 1. The Young's modulus and Poisson ratio for gold were $92.05*10^3$ MPa and

0.42, respectively. The anisotropic elastic modulus matrix C^s and and piezoelectric constant matrix e were taken from Lu *et al* [4].

$$C^s(MPa) = \begin{bmatrix} 86.74*10^3 & 267.15*10^3 & -8.25*10^3 & 0 & -3.66*10^3 & 0 \\ & 102.83*10^3 & -7.42*10^3 & 0 & 9.92*10^3 & 0 \\ & & 129.77*10^3 & 0 & 5.70*10^3 & 0 \\ sym & & & 68.81*10^3 & 0 & 2.53*10^3 \\ & & & & 38.61*10^3 & 0 \\ & & & & & 29.01*10^3 \end{bmatrix}$$

$$e(C/m^2) = \begin{bmatrix} 0.171 & 0 & 0 \\ -0.0187 & 0 & 0 \\ -0.152 & 0 & 0 \\ 0 & -0.0761 & 0.067 \\ 0.067 & 0 & 0 \\ 0 & 0.067 & -0.095 \end{bmatrix}$$

The model was meshed using 24, 8131 three dimensional 20 node coupled field solid elements, in Ansys® [6]; nevertheless a very small piezoelectric coupling was applied for the gold electrode. The meshed model is depicted in Fig. 1. The applied voltage was 1 V and a damping coefficient of 2% was used. A harmonic analysis was carried out to study the frequency response in the range of 8.9 to 10.1 MHz, employing a total of 200 steps/intervals. The frequency response was then utilized to obtain the displacement profiles on the device surface, which in turn are used to compute the sensitivity distribution on the surface [7].

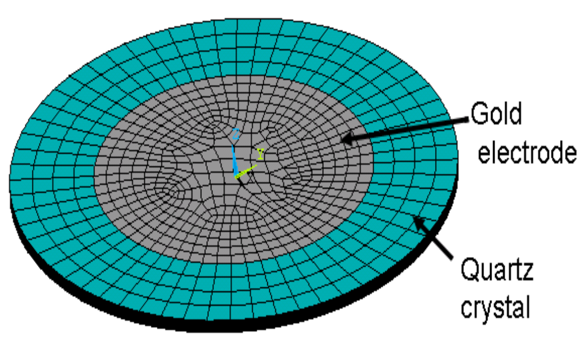


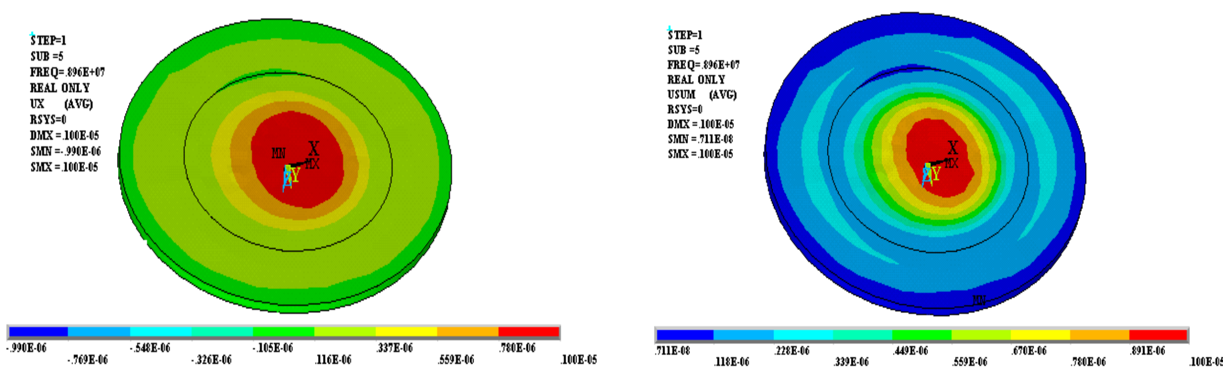
Figure 1: Schematic diagram showing the finite element model of the quartz crystal nanobalance with the gold electrodes

Table 1: Dimensions and properties of the QCN device components

	Piezoelectric crystal	Gold electrode
Diameter (mm)	8	5
Thickness (mm)	0.185	$0.15*10^{-3}$
Density (kg/ m^3)	2649	18500

2. Results and Discussion

The distributions of the radial and total displacement of the device surface are shown in Figs. 2 (a,b) which also indicate the first thickness shearing mode of the device. Figure 3 reflects the shearing motion of the device polarized in the X-direction, as also reported in the literature [8, 9]. Our results indicate that the displacement peaks at the center of the electrode and decays rapidly on moving away from the center (Fig. 4), which is in qualitative agreement with the experimental results reported in the literature [10]. In addition, the distribution of the displacement is elliptical rather than circular and is largely bounded by the electrode surface. The peak static displacements are of the order of a picometer (0.923 pm) and are mainly in the x direction. The displacements in the y and z directions are almost 2 orders of magnitude smaller than in the x-direction thereby suggesting the primarily X directional nature of the shearing waves.



(a) **(b)**

Figure 2: Contour plot showing the (a) radial displacement. (b) Total displacement of the device surface. The displacements shown above are in micrometers.

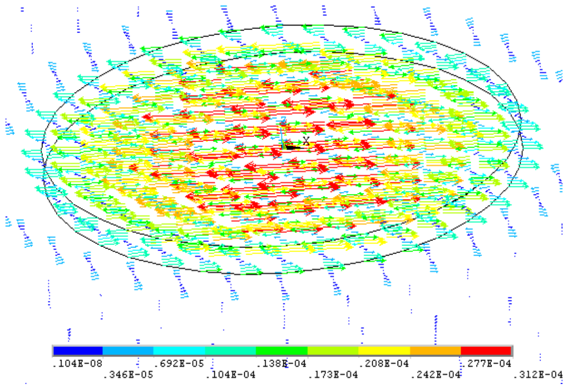


Figure 3: Vector plot showing the shearing motion of the device

Considering the influence of mechanical resonance, the dynamic displacements can be obtained from the static displacements by using the Quality factor (Q) [11]

$$U_{dyn}=U_{stat}*Q \qquad (1)$$

For a 9 MHz crystal, Q in air is reported to be 84195 [12]. This gives the value of peak displacement to be

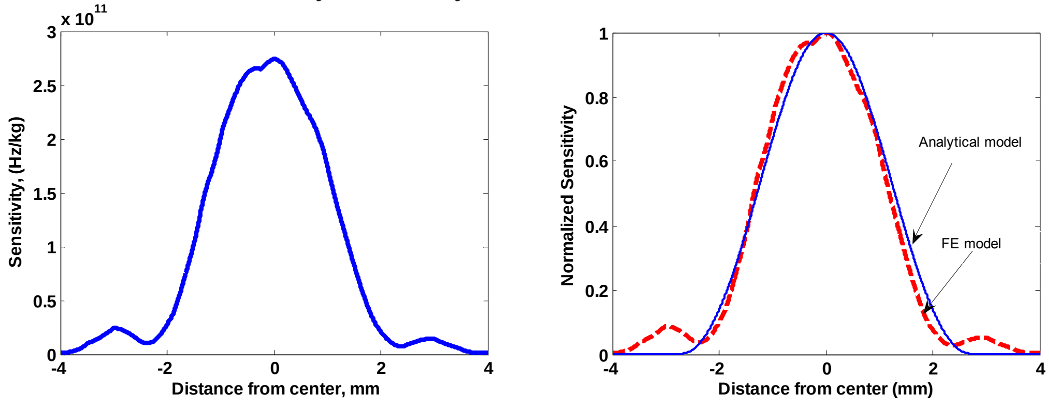
$$U_{dyn}=0.923*10^{-6}\ (\mu m)*84195$$
$$=77.7\ nm \qquad (2)$$

Thus, peak displacement for the current 9 MHz device, at a peak driving voltage of 1 V is 77.5 nm, which is in agreement with the experimentally observed displacements of a QCM obtained using scanning tunneling microscopy under vacuum [13]. The sensitivity distribution for the device surface was computed using the equation

$$S=\frac{|u(r)|^2}{2\pi\int_0^\infty r|u(r)|^2\,dr}C_f \qquad (3)$$

C_f is Sauerbrey's sensitivity constant, u(r) is the displacement on the device surface and r is the distance from the center. Cf = 1.78e+011 Hz/kg

The device sensitivity distribution is shown in Fig. 4a The sensitivity shows qualitative agreement with the sensitivity distribution obtained using analytical models [7] as evident from the distributions normalized using the corresponding maximum values depicted in Fig. 4b. Josse et al have analyzed n-m type electrode designs (n and m denote the unequal diameters of the top and bottom electrodes) using the analytical model [7]. The future work is focused on the investigation of the n-m type electrodes using the FE model developed in the current work, which can be thoroughly compared with the results from analytical models presented by Josse et al [7]. The developed FE models can then be reliably extended to study ring designs in order to explore the possibility of obtaining a flat sensitivity distribution based on known analytical results in addition to the investigation of more complex electrode configurations which cannot be addressed by the analytical models.



(a) **(b)**

Figure 4: (a) Sensitivity distribution on the device surface (b) Normalized Sensitivity distribution from the current FE model vs. analytical model of Josse et al. [7]

3. Conclusions

In this work, a three dimensional finite element model for a quartz crystal nanobalance device was developed to investigate the displacement and sensitivity distributions on the surface. Our results indicate that the displacement distribution of the device surface peaks at the centre and decays rapidly on moving to the device periphery; most of the displacement is confined to the electrode region. In addition, the shearing motion of the device is polarized in the X-direction as also reported in the literature. The developed model predicts surface displacements in accordance with the experimental values reported in the literature. Further efforts are underway to investigate different electrode designs to identify an electrode configuration which can provide high and uniform sensitivity across the device surface.

Acknowledgements

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