

THE EFFECT OF SPIN COATING'S SPEED ON GRAPHENE-OXIDE'S LAYER TOPOGRAPHY AND QCM VISCOELASTICITY

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Abstract

Spin coating speed controls the deposition process, affecting the formation of the layer's topography of graphene oxide and the viscoelastic properties of quartz crystal microbalance (QCM). In this work, the graphene oxide was deposited on the QCM surface by spin coating method with speeds ranging from 500 to 800 rpm. The layer's surface topography was subsequently measured and analyzed using a roughness multiparameter, which included the area average (S_a), area roughness deviation (S_q), localized surface roughness (S_z), surface skewness (S_{sk}), and surface kurtosis (S_{ku}). The S_a , S_q , and S_z , which were decreased as the spinning speed increased. The measurements indicated that the graphene oxide layer deposited at 500 rpm had a rough and clumpy texture, while at a high speed of 800 rpm, the layer had a much smoother texture. A detailed analysis using the S_{ku} and S_{sk} parameters revealed that the clumpy texture in the 500 rpm sample contains tapered sharp graphene oxide structures and a small amount of porosity. On the other hand, the sharp structures were significantly reduced, and porosity was dominant in the sample deposited at high speeds of 700 rpm and 800 rpm. The effect of the spinning speed and the topography on the viscoelasticity was studied using an impedance analyzer. The impedance of the layer deposited at 500 rpm was relatively higher than that at 800 rpm. The frequency shift was also relatively significant for the sample deposited at low speed and was reduced at higher speed. The impedance analysis demonstrated that the layer's viscosity is affected by its mass, roughness, texture, and porosity.

Keywords: Graphene Oxide; Roughness; Spin Coating's Speed; QCM; Viscoelastic

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Introduction

Functional coatings have recently become a widely developed study for improving material properties. Functional layers based on carbon allotropic materials have demonstrated remarkable properties across various technological aspects, especially in sensor applications (Tiwari *et al.*, 2016). Graphene oxide is a thin layer of allotropic carbon that has sp^2 hydration with an atomic distance of about 0.142 nm (Kim *et al.*, 2009). The most attractive characteristics that graphene oxide can provide in the sensing structure include a large specific surface area and wealthy functional groups as sensing layers (Navarrete *et al.*, 2023). The thin sheet-like structure of graphene oxide facilitates electron transport, making it an ideal candidate for coating quartz crystal microbalance (QCM) (Tang *et al.*, 2021). When applied as a thin layer on a QCM, graphene oxide increases gas adsorption and, consequently, material sensitivity.

QCM has a piezoelectric working principle on quartz crystals with material loading on their surface, affecting the resonant frequency. Changes in mass result in observable frequency shifts induced by molecular interactions with the QCM surface. Modifying the surface of the thin QCM layer can enhance sensor properties, primarily by increasing gas adsorption (Aflaha *et al.*, 2023). Thus, a graphene oxide layer enhances molecular interactions when exposed to gas analytes.

Several deposition methods can be employed to create a thin layer of graphene oxide on the surface of a QCM. Some of the deposition methods previously explored include chemical vapor deposition (Chang *et al.*, 2016), dip coating (Kim *et al.*, 2019), and ultrasonic spray deposition (Masrie *et al.*, 2020). Research conducted by Lee *et al.* in 2019 involved the spray coating of graphene oxide on a QCM. However, this method necessitates the optimization of parameters for a homogenous thin film, including layer thickness, spray speed, spray pressure, solution viscosity, and spray distance (Lee *et al.*, 2019).

Another method for depositing graphene oxide layers on the QCM surface is the spin coating, known for its relatively high degree of homogeneity (Jarka *et al.*, 2017). Several factors can affect the quality of thin film formation, including spin coating speed (Oo *et al.*, 2022), rotational time (Na *et al.*, 2015), and solution concentration (Garg *et al.*, 2017). The spin coating speed will affect the formation of the layer due to the centrifugal forces acting on the layer (Liu *et al.*, 2022).

In this study, the surface topography of the coating was evaluated in terms of surface roughness. In previous research (Moreira *et al.*, 2020), the impact of spin coating on roughness was assessed using a single parameter, namely the surface average roughness (S_a). However, multiple roughness properties, such as texture, sharpness, and porosity, influence functional layer properties, which are significant in viscoelasticity analysis. Therefore, analyzing the coating quality through multiparameter analysis, such as surface root mean square (S_q), localized surface roughness (S_z), surface kurtosis (S_{ku}), and surface skewness (S_{sk}), provides more comprehensive insights into surface topography.

The present work investigates how spin coating speed affects the topography and viscoelastic properties of QCM. Layer topography notably influences QCM properties (Putri *et al.*, 2023), primarily due to variations in the deposited layer's mass (Georgieva *et al.*, 2018). A QCM equipped with a viscoelastic layer exhibits energy loss during oscillations due to loading (Masruroh, 2022). The viscoelastic properties are analyzed through the investigation of frequency shifts and impedance.

Materials and Methods

Synthesis and Deposition of Graphene Oxide Thin Film

The preparation stage begins with preparing a graphene oxide solution purchased from Suzhou BKnano 2D materials. The solution has a concentration of 2 mg/ml, sheet diameter of 20-25 μ m, disperse rate of 0.1 g/L, and purity of 99%. Put 20 ml of graphene oxide solution into a beaker glass. The graphene oxide solution was homogenized in a glass beaker using a magnetic stirrer. The stirrer was set at 1000 rpm for 1 hour with a hot plate temperature of 25°C. The solution was then sonicated in an ultrasound cleaner bath with a frequency of 40 kHz for 180 minutes. The purpose of ultrasonication was to prevent agglomeration and reduce oxygen content in the graphene oxide solution.

For the deposition process, take ten microliters of the synthesized graphene oxide solution using a micropipette. Then, carefully drip the graphene oxide solution onto the QCM substrate. The QCM substrate, with a resonant frequency of 10 MHz, was purchased from Greatmicro Electronic in Surabaya,

Indonesia. Ensure the QCM is placed inside the sample test tool before dripping the graphene oxide solution. Thin film deposition was carried out using a spin coater (as shown in Figure 1(a)). The substrate was dripped with graphene oxide solution while rotating for 30 seconds with variations of spin coating speed, namely 500 rpm, 600 rpm, 700 rpm, and 800 rpm (Figure 1(b)). The speed variation treatment process was carried out to obtain an even distribution of the thin layer. Pre-annealing was carried out using an oven to remove moisture from the thin film (as illustrated in Figure 1(c)). Then, it was heated at 100°C for 30 minutes.

Raman Spectroscopy Characterization

The layers were initially characterized using Raman spectroscopy. This characterization aimed to confirm the presence of graphene oxide in the deposited layer. Raman spectroscopy (Horiba Scientific - InVia Raman Spectroscope) utilized a 532 nm excitation laser.

Topography Measurement System (TMS) Characterization

The surface topography of the layer was examined by measuring its surface roughness using a white light surface profiler (TMS-1200 by Polytech). The system employs the Mirau interferometer principle, which applies monochromatic light to the thin film sample. The resulting interference patterns were processed using TMS 3.2 software to determine the shape and topography values of the layers formed.

Impedance Analyzer Characterization

An impedance analyzer is used to measure the impedance value to determine the viscoelasticity of QCM before and after deposition. The impedance analyzer used is the Bode 100 type (OMICRON Electronics Asia Ltd., Hong Kong), which measures the complex impedance of a circuit as a function of frequency. An impedance analyzer can provide a graphical relationship between the impedance value and the phase. The resulting values provide information about the viscoelastic properties of QCM.

Results and Discussion

Raman Spectroscopy

Figure 2 displays the Raman spectroscopic spectrum, exhibiting two characteristic peaks, the G and D bands, typical of carbon allotropes. The G band results from C-C single bond stretching, with an intensity of 731 at 1598 cm⁻¹. The D band, at 1352 cm⁻¹, exhibits an intensity of 718. The ratio

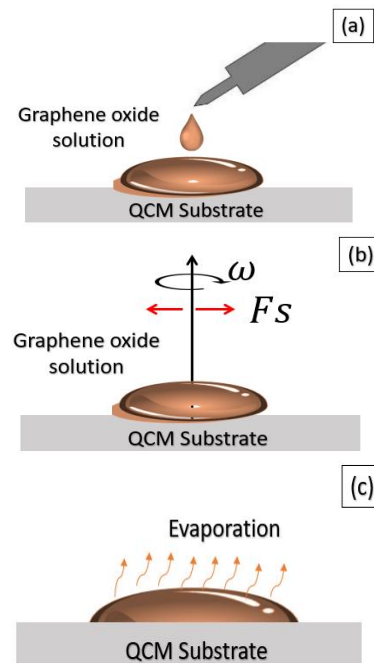


Figure 1. Thin film deposition process with spin coating method, a) drops solution, b) spin process, c) evaporation solution

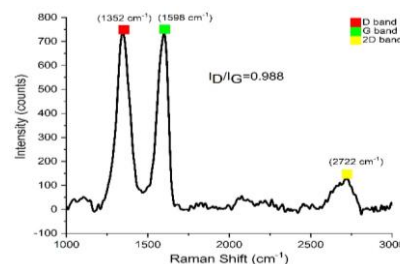


Figure 2. Raman spectroscopy of graphene oxide

between the intensities of the D and G bands (I_D/I_G) is 0.988, indicating that the layer formed on the QCM surface is graphene oxide.

The graphene oxide structure is indicated by an I_D/I_G intensity ratio of less than 1. Conversely, a value greater than one signifies reduced graphene oxide (rGO). The G-band mode results from the phonon energy of atoms in the sp^2 state of carbon, while the appearance of the D-band mode is due to the irregularity of the crystal structure (Brolly *et al.*, 2016). In this study, a 2D band is identified at a wavenumber of 2722 cm⁻¹. A lower intensity of the 2D band compared to the G band suggests a reduction in the layer thickness (Muzyka *et al.*, 2018). Raman spectroscopic observations have confirmed that the formed layer is a graphene oxide structure, and further characterization of its surface topography is warranted.

Topography

The topography analysis results are presented as surface roughness values, visually representing the deposited layer's surface texture. The surface of the graphene oxide layer formed peaks and valleys representing the topography of the deposited sheet particles. An increased roughness, with an even distribution, makes the graphene layer more effective in capturing gas molecules, allowing it to function as a sensor.

Figures 3(a)-3(e) illustrate the roughness profile of the analyzed layers in two dimensions. The image is generated by the reflection of monochromatic light from the graphene oxide positioned above the QCM. The reflected light gives rise to destructive and constructive interference patterns, visually representing the layer's texture and roughness. The color scale, displayed as the Z-axis, represents the layer's height. Darker colors indicate a higher layer, corresponding to a constructive interference pattern. The x-axis of the roughness graph is scaled from zero to 800 microns, indicating the coating's edge.

The spin coating speed significantly influences the formation of graphene oxide layers on the QCM. At 500 rpm (Figure 3(b)), a prominent peak at the center of the surface suggests the presence of agglomerate particles formed during deposition. At

600 rpm (Figure 3(c)), many agglomerates appear and are more evenly distributed in height. In contrast, at 700 rpm (Figure 3(d)), there are very few peaks with a relatively low height, and the distribution of peaks is less pronounced, tending to form pores. Finally, the surface roughness becomes more uniform at 800 rpm (Figure 3(e)). In contrast, the QCM before deposition (Figure 3(a)) exhibits a smoother texture than the graphene oxide layer.

Figure 3 shows that thin film deposition with high rpm results in a lower roughness profile. The deposition process with high spin coating speed will produce an even-thin layer (Figure 3(d)). Meanwhile, the speed at low rpm has a peak roughness profile in the center, indicating the agglomeration of graphene oxide particles (Figure 3(a)). The presence of agglomeration indicates the growth mode of the island layer. The island growth mode is due to the reduced interaction energy of the deposition with the substrate (Zubar *et al.*, 2018). As a result, the film accumulates at one point to form an island (Figure 4). The low-speed deposition process resulted in imperfect coating spreading, leading to a lack of coating interaction energy. The roughness profile is higher even at increased spin coating speed.

Previous research has indicated that an increase in the spin coating speed in rpm units

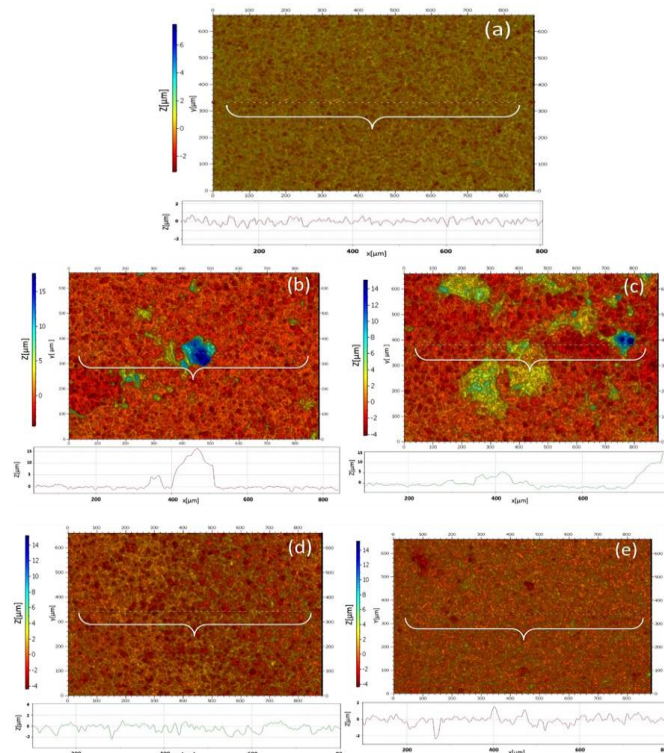


Figure 3. Profile 2D TMS a) before deposition; at different speeds b) 500 rpm, b)600 rpm, c) 700 rpm, d) 800 rpm

results in the material spreading further away from the substrate (Han *et al.*, 2021). As the layers spread, the thickness decreases, and the difference in roughness from the center to the edges is reduced. Higher spin coating speeds increase interaction energy, while rough topography on the layer suggests an island growth mode. Table 1 illustrates the impact of varying spin coating speeds on roughness parameter values.

TMS characterization results indicate that the graphene oxide layer's roughness corresponds to the deposited plane's area. The average surface roughness (S_a) represents the mean height of the layer calculated over a specific area. The value of S_a provides an overview of the surface roughness in the overall deposition profile (Hernandez and Hernandez, 2021). The root mean square surface roughness is denoted as Surface root mean square (S_q). S_q measures the overall deviation of average roughness from a flat surface to the top of the layer. The individual surface value (S_z) quantifies the difference in surface elevation between the average of the five highest peaks and the five lowest valleys within the measured area. These values of S_a , S_q , and S_z can be expressed as values (S_z). The values of S_a , S_q , and S_z are written mathematically as equations (1), (2), and (3) (Pawlus *et al.*, 2021).

$$S_a = \frac{1}{A} \iint_0^A |Z(x, y)| dx dy \quad (1)$$

$$S_q = \sqrt{\frac{1}{A} \iint_0^A |Z(x, y)|^2 dx dy} \quad (2)$$

$$S_z = \frac{1}{n} (S_{z1} + S_{z2} + S_{z3} + \dots + S_{zn}) \quad (3)$$

Where A is the surface area of the profile on the The Y-axis is used for measurement, and Z(x) is the

height variation of the surface profile for each data point.

The roughness profile S_a in Table 1 shows a decreasing trend with each spin coating speed increase. Increasing the spin coating's speed during the deposition process results in a decrease in surface roughness value. The S_q/S_a ratio has the highest value at 600 rpm spin coating speed. The S_q/S_a value indicates many peak distributions represented by particle buildup. Based on the Gaussian distribution, the comparison between the roots of the squared roughness and the average roughness, which has a value of more than 1.25, indicates a high roughness distribution (Kumar and Rao, 2012). The S_z supports this result as an indication of the layer with the highest peak. Huge S_z indicates higher agglomeration of layers. As a result, a homogeneous layer topography is obtained when the speed increases. The homogeneous layer indicated by the roughness of each point will be lower.

Additional insights into the sharpness of the peaks can be obtained from the Surface kurtosis (S_{ku}) parameter. S_{ku} measures the expected distribution value of the average roughness, describing asymmetry properties (Bajpai *et al.*, 2016). S_{ku} of more than 3 indicates a spiky surface corresponding to a Gaussian height distribution (Yang *et al.*, 2021). The spiky surface means the number of peaks is relatively high, and the valleys are deep enough. The graphene oxide layer at all variations of spin coating speed has a S_{ku} value exceeding 3, indicating that the layer is relatively spiky. The highest S_{ku} observed at 500 rpm suggests that the graphene oxide layer initially converges at a single point before being dispersed due to rotational motion. S_{ku} is calculated using equation (4) (Molnar, 2022).

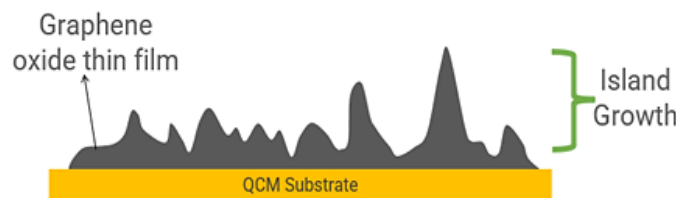


Figure 4. Island growth mode on a substrate

Table 1. Roughness parameter of graphene oxide thin film at different speeds

	S_a (nm)	S_q (nm)	S_q/S_a	S_z (nm)	S_{ku}	S_{sk}
Before Deposition	250.22	324.38	1.296	4840	4.18	-0.78
Spin 500	644.12	801.98	1.245	17280	16.89	1.19
coating's 600	528.71	767.73	1.452	15111	13.2	1.37
speed (rpm) 700	449.02	583.02	1.298	8001	4.27	-0.57
800	386.6	513.87	1.329	7910	5.54	-0.43

$$S_{ku} = \frac{1}{A} \cdot \frac{1}{S_q^4} \iint_0^A |Z^4(x, y)| dx dy \quad (4)$$

Another critical parameter is the surface skewness (S_{sk}). The S_{sk} measurement aims to determine the profile/surface symmetry on the average plane at deep valleys or high peaks, predicting surface porosity (Liu *et al.*, 2015). At speeds of 500 and 600 rpm, positive S_{sk} values indicate dominant peak height, whereas spin coating speeds of 700 and 800 rpm are marked by negative S_{sk} values, indicating the prevalence of valley depths and, thus, porosity. Surface skewness (S_{sk}) respectively based on equations (5) (Molnar, 2022).

$$S_{sk} = \frac{1}{A} \cdot \frac{1}{S_q^3} \iint_0^A |Z^3(x, y)| dx dy \quad (5)$$

The S_{sk} value measures profile/surface symmetry on the average plane and is more aimed at deep valleys or high peaks. S_{sk} gives priority to the appearance of surface porosity (Liu *et al.*, 2015). The S_{sk} value equal to zero indicates a slope balance between the peaks and valleys. Spin coating's speeds of 500 rpm and 600 rpm have positive S_{sk} values indicating a more dominant peak slope. The spin coating's speeds of 700 rpm and 800 rpm have a prevailing valley slope indicated by a negative S_{sk} value. The dominant valley shows the amount of porosity formed.

The overall roughness multiparameter values in Table 1 indicate reduced roughness with increasing spin coating speed. Surface roughness decreases at higher speeds. Increasing spin coating speed produces a more even roughness distribution across the entire QCM surface (Belhadj *et al.*, 2021). A layer with high roughness but uneven surface results in a low ability to capture analyte molecules.

Viscoelasticity

The viscoelastic properties of the QCM after deposition with graphene oxide depend on the measured impedance value, which signifies a loading on the QCM. High impedance leads to extensive QCM loading, rendering it less rigid. Table 2 displays the change in impedance at each

spin coating speed. The study reveals that a speed of 500 rpm results in the most significant change in impedance value. This speed causes the QCM to oscillate due to the rough and uneven topography of the coating. It's evident from Table 2 that an increase in spin coating speed results in a marginal change in impedance (ΔZ). The ΔZ value is calculated as the difference between the impedance after deposition (Z_1) and before deposition (Z_0).

The impedance change (ΔZ) is accompanied by the shift of frequency (Δf), which indicates the loading of graphene oxide particles on the QCM. Δf is calculated as the difference between the frequency after deposition (f_1) and before deposition (f_0). A negative frequency shift value suggests a decreased QCM oscillation frequency from high to low. This drop in frequency occurs because the QCM's surface, coated with a layer, requires more time to vibrate, resulting in a smaller frequency value.

Impedance analyzer measurements reveal that a spin coating speed of 500 rpm produces a high-frequency shift due to its rough surface. A rough surface produces a more significant change in deposited mass than a smooth or flat-textured surface. The frequency value is closely related to the mass of graphene oxide deposited, and this relationship is expressed in the Saurbrey equation (Huang *et al.*, 2017):

$$\Delta m = -C_{QCM} \times \Delta f \quad (6)$$

Equation (6) calculates the mass of graphene oxide deposited on QCM. The magnitude of the change in mass is calculated from the frequency shift. The frequency shift signifies the ability of the QCM to oscillate. The low-frequency shift shows that the QCM remains rigid, and the Saurubey equation can be fulfilled.

Lower speeds cause graphene oxide particles to exhibit high roughness but become concentrated in the center of the surface. The topography of the agglomerated layer on the QCM surface significantly affects oscillation due to loading. The main factors influencing loading are the uneven layer topography and stiffness (Masruroh, 2022). Higher roughness results in more graphene oxide mass being deposited due to non-uniform distribution.

Table 2. Influence spin coating's speed of impedance change, frequency shift, and mass deposited

Spin coating's speed (rpm)	Z_0 (ohm)	Z_1 (ohm)	ΔZ (ohm)	f_0 (Mhz)	f_1 (Mhz)	Δf (Mhz)	Δm (µg)
500	15.656	79.478	63.822	10.005	9.9091	-0.0958	42.1705
600	15.162	48.404	33.242	10.005	9.9238	-0.0812	35.7236
700	15.719	41.776	26.057	10.005	9.9542	-0.0507	22.3142
800	15.669	17.716	2.047	10.005	9.9554	-0.0495	21.7985

The QCM, before deposition, possesses a series resonance frequency of 10 MHz, signifying the crystal's fundamental vibration frequency. After deposition, the resonant frequency shifts to 9.9 MHz. The deposited layer exhibits a maximum resonant frequency shift of 0.09 MHz, representing a 0.9% shift. The Saurubey equation can be fulfilled when the maximum frequency shift is less than 2% of the QCM's pre-deposition value (Qiao *et al.*, 2015). It indicates that the QCM is considered rigid and lacks viscoelastic behavior. Layers deposited at 500 rpm (Figure 5(b)) and 600 rpm (Figure 5(c)) show rounded peaks. The layers formed at 500 and 600 rpm show that the QCM leads to viscoelastic properties. The different peak shapes obtained at 700 (Figure 5(d)) and 800 rpm (Figure 5(e)) spin coating speeds deliver a sharp shape. A pointed peak

indicates a tendency for the QCM layer to become rigid. The rigid property of QCM signifies that the oscillation energy is not dampened. The spin coating's speed of 800 rpm has a peak similarity with QCM before deposition. This similarity indicates that there is no loading that inhibits QCM oscillations.

The phase impedance also represents a shift in the QCM resonance produced by the change in mass on the deposited surface (March *et al.*, 2015). The sharp peaks indicate the tendency of the layer to become stiff. In contrast, the rounded peaks indicate viscoelastic or elastic properties so that there is a scattering of QCM oscillation energy. A firm peak and a meeting point between impedance and phase suggest that the Butterworth-Van Dyke (BVD) model is still running well. The BVD model

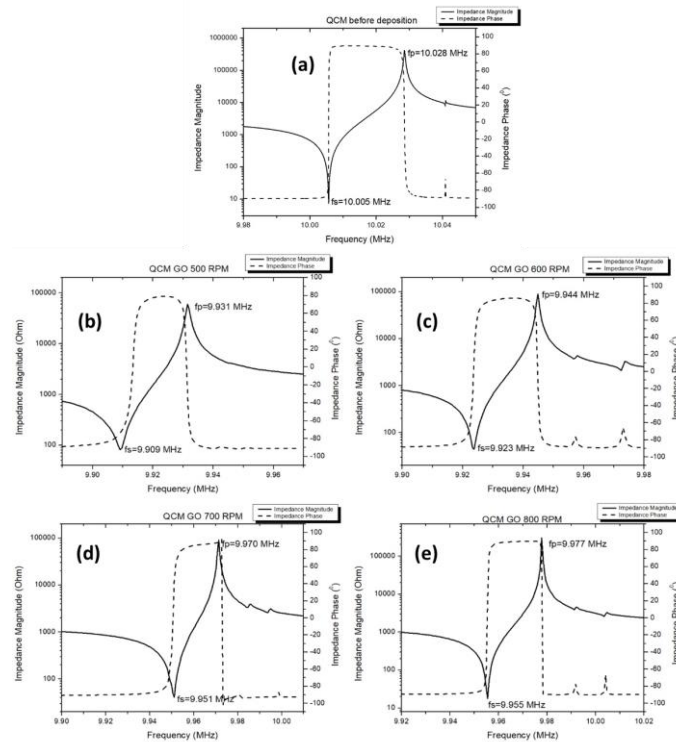


Figure 5. Deposition effect on magnitude impedance dan phase QCM a) before deposition; after deposition at different speeds b) 500 rpm, c) 600 rpm, d) 700 rpm, e) 800 rpm

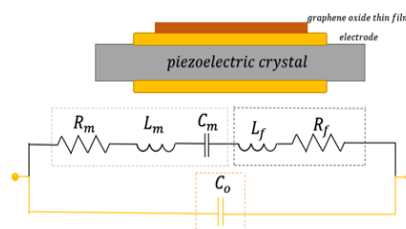


Figure 6. Crystal diagram QCM after deposition (top), electric model BVD (bottom)

provides an example of mechanical phenomena in QCMs using an electrical system with two equivalent arms, namely the moving and fixed arms. The motion arm is a combination of resistor (R), inductor (L), and capacitor (C) components. The fixed arm is represented by a capacitance parallel to the motion arm (Alassi *et al.*, 2017). The oscillation of the QCM stored energy results in a change in the value of the total impedance component on the movable arm (Figure 6). This is the reason why the series equivalent circuit is used to represent the oscillation behavior of the QCM (Table 2).

The QCM sensor without a deposit exhibits fundamental oscillation and broad phase impedance. However, the frequency decreases after deposition, and the phase impedance narrows. The QCM before deposition, modeled by BVD, consists of a series RLC motion arm (see Figure 6), where the quartz capacitance (C_m) represents the stored energy per oscillation, the quartz resistance (R_m) represents the energy loss, damping per oscillation from the crystal structure, and the quartz inductor (L_m) as the inertial element (Arnau, 2008). A layer deposited on the QCM surface introduces a series of components consisting of a resistance layer (R_f) and an inductor layer (L_f). When oscillating, the increased resistance component provides damping as a viscous property, and the inductor layer contributes to QCM elasticity. The QCM, after deposition with a spin coating speed of 500 rpm, exhibits a lower phase height due to the addition of higher resistance. Conversely, higher spin coating speeds show a slight phase enhancement with increasingly well-defined and pointed peaks attributed to the reduced mass dampening QCM vibration (lower resistance).

A significantly decreased crystal frequency can potentially interfere with QCM performance (Gu *et al.*, 2018). Surface topography with even roughness values allows low mass to be deposited. So, the more mass deposited, the higher the impedance value. The even and low mass layer makes a more minor frequency shift. The effect of spin coating's speed that allows deposition with low-frequency shifts has better oscillation capabilities of QCM.

Conclusions

This study utilizes roughness multiparameter and impedance analysis to elucidate the influence of spin coating speed on the surface topography and viscoelasticity of graphene oxide. The topography of the graphene oxide layer exhibits a peak-like texture at speeds of 500 rpm and 600 rpm. S_a , S_q , and S_z values decrease as spin coating speed increases. Additionally, the sharpness, as measured by the S_{ku}

parameter, is higher at lower spin coating speeds. Negative values for the S_{sk} parameter at speeds of 700 rpm and 800 rpm indicate prevalent porosity. Impedance analysis reveals that the change in impedance is more significant at lower spin coating speeds. Large agglomerates in the graphene oxide layer's topography lead to increased mass on the QCM surface, resulting in a more prominent frequency shift. In this study, the speed of 800 rpm exhibited the lowest frequency shift because less mass was deposited. Overall, the frequency shifts observed in this study mitigate the viscoelastic properties of the QCM, rendering it more rigid.

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References

- Aflaha, R., Afiyanti, H., Azizah, Z. N., Khoirudin, H., Rianjanu, A., Kusumaatmaja, A., Roto, R., and Triyana, K. (2023). Improving ammonia sensing performance of quartz crystal microbalance (QCM) coated with nanofibers and polyaniline (PANI) overlay. *Biosensors and Bioelectronics*: X, 13:100300. <https://doi.org/10.1016/j.biosx.2022.100300>
- Alassi, A., Benammar, M., and Brett, D. (2017). Quartz crystal microbalance electronic interfacing systems: A review. *Sensors*, 17(12):2799. <https://doi.org/10.3390/s17122799>
- Arnau, A. (2008). A review of interface electronic systems for AT-cut quartz crystal microbalance applications in liquids. *Sensors*, 8(1):370-411. <https://doi.org/10.3390/s8010370>
- Bajpai, A.K., Bhatt, R., and Katare, R. (2016). Atomic force microscopy enabled roughness analysis of nanostructured poly (diaminonaphthalene) doped poly (vinyl alcohol) conducting polymer thin films. *Micron*, 90:12-17. <https://doi.org/10.1016/j.micron.2016.07.012>
- Belhadj, W., Timoumi, A., Alamer, F.A., Alsalmi, O.H., and Alamri, S.N. (2021). Experimental study and theoretical modeling of coating-speed-dependent optical properties of TiO₂-graphene-oxide thin films. *Results in Physics*, 30:104867. <https://doi.org/10.1016/j.rinp.2021.104867>
- Brolly, C., Parnell, J., and Bowden, S. (2016). Raman spectroscopy: Caution when interpreting organic carbon from oxidizing environments. *Planetary and Space Science*, 121:53-59. <https://doi.org/10.1016/j.pss.2015.12.008>
- Chang, S.J., Hyun, M.S., Myung, S., Kang, M.A., Yoo, J.H., Lee, K.G., Choi, B.G., Cho, Y., Lee, G., and Park, T.J. (2016). Graphene growth from reduced graphene oxide by chemical vapour deposition: Seeded growth accompanied by restoration. *Scientific Reports*, 6:22653. <https://doi.org/10.1038/srep22653>
- Garg, R., Elmas, S., Nann, T., and Andersson, M.R. (2017). Deposition Methods of Graphene as Electrode Material for Organic Solar Cells. *Advanced Energy Materials*, 7(10). <https://doi.org/10.1002/aenm.201601393>
- Georgieva, B., Nichev, H., Petrov, M., Koutzarova, T., Georgieva, V., and Dimova-Malinovska, D. (2018).

- Influence of loading QCMs with electrochemically-deposited ZnO on their NO₂-sensing properties. *Journal of Physics: Conference Series*, 992(1). <https://doi.org/10.1088/1742-6596/992/1/012026>
- Gu, C., Li, P., Jin, F., Chen, G., and Ma, L. (2018). Effects of the imperfect interface and viscoelastic loading on vibration characteristics of a quartz crystal microbalance. *Acta Mechanica*, 229(7):2,967-2,977. <https://doi.org/10.1007/s00707-018-2155-x>
- Han, D.H., Kim, D., Yun, H.W., Lee, J., Lee, U. gi, Chung, H. S., and Kim, W.B. (2021). Effects of aging on the thickness of a homogeneous film fabricated using a spin coating process. *Journal of Coatings Technology and Research*, 18(3):641-647. <https://doi.org/10.1007/s11998-020-00429-x>
- Hernandez, M.P. and Hernandez, G. (2021). Quantitative Surface Morphology Analysis Using Roughness Parameters. *Revista Cubana de Física*, 38(1):10-16.
- Huang, X., Bai, Q., Hu, J., and Hou, D. (2017). A practical model of quartz crystal microbalance in actual applications. *Sensors*, 17(8):1,785. <https://doi.org/10.3390/s17081785>
- Jarka, P., Tański, T., Matysiak, W., Krzemiński, L., Hajduk, B., and Bilewicz, M. (2017). Manufacturing and investigation of surface morphology and optical properties of composite thin films reinforced by TiO₂, Bi₂O₃ and SiO₂ nanoparticles. *Applied Surface Science*, 424(Part 2):206-212. <https://doi.org/10.1016/j.apsusc.2017.03.232>
- Kim, J.Y., Shin, D.O., Kim, K.M., Oh, J., Kim, J., Kang, S.H., Lee, M.J., and Lee, Y.G. (2019). Graphene Oxide Induced Surface Modification for Functional Separators in Lithium Secondary Batteries. *Scientific Reports*, 9(1):1-8. <https://doi.org/10.1038/s41598-019-39237-8>
- Kim, K.S., Zhao, Y., Jang, H., Lee, S.Y., Kim, J.M., Kim, K.S., Ahn, J.H., Kim, P., Choi, J.Y., and Hong, B.H. (2009). Large-scale pattern growth of graphene films for stretchable transparent electrodes. *Nature*, 457(7230):706-710. <https://doi.org/10.1038/nature07719>
- Kumar, B.R. and Rao, T.S. (2012). AFM studies on surface morphology, topography, and texture of nanostructured zinc aluminum oxide thin films. *Digest Journal of Nanomaterials and Biostructures*, 7(4):1,881-1,889.
- Lee, S.W., Choi, B.I., Kim, J.C., Woo, S.B., Kim, Y.G., Yoo, J., and Seo, Y.S. (2019). Reduction and compensation of humidity measurement errors at cold temperatures using dual QCM humidity sensors based on graphene oxides. *Sensors and Actuators, B: Chemical*, 284:386-394. <https://doi.org/10.1016/j.snb.2018.12.154>
- Liu, L., Li, X., and Nonaka, K. (2015). Light depolarization in off-specular reflection on submicro rough metal surfaces with imperfectly random roughness. *Review of Scientific Instruments*, 86(2). <https://doi.org/10.1063/1.4908172>
- Liu, Q., Yan, Y., Meng, L., Zhang, Z., and Zhou, P. (2022). Influence of Airflow Disturbance on the Uniformity of Spin Coating Film Thickness on Large Area Rectangular Substrates. *Coatings*, 12(9). <https://doi.org/10.3390/coatings12091253>
- March, C., García, J.V., Sánchez, Á., Arnau, A., Jiménez, Y., García, P., Manclús, J.J., and Montoya, Á. (2015). High-frequency phase shift measurement greatly enhances the sensitivity of QCM immunosensors. *Biosensors and Bioelectronics*, 65:1-8. <https://doi.org/10.1016/j.bios.2014.10.001>
- Masrie, M., Badaruddin, S.A.M., Hussin, M.R.M., Nor, N.M.R.M., and Joe, J. (2020). Rapid Reduction of Graphene Oxide Thin Films on Large-Area Silicon Substrate. *Journal of Physics: Conference Series*, 1535(1). <https://doi.org/10.1088/1742-6596/1535/1/012027>
- Masruroh, M. (2022). The Impedance Analysis of a Viscoelastic Petalous Structured Stearic Acid Functional Layer Deposited on a QCM. *Sensors*, 22(19):7,504. <https://doi.org/10.3390/s22197504>
- Molnar, V. (2022). Asymmetric Height Distribution of Surfaces Machined by Hard Turning and Grinding. *Symmetry*, 14(8):1,591. <https://doi.org/10.3390/sym14081591>
- Moreira, J., Vale, A.C., Pires R.A., Botelho, G., Reis, R.L., and Alves, N.M. (2020). Spin-Coated Polysaccharide-Based Multilayered Freestanding Films with Adhesive and Bioactive Moieties. *Molecules*, 25(4):840. <https://doi.org/10.3390/molecules25040840>
- Muzyka, R., Drewniak, S., Pustelny, T., Chrubasik, M., and Gryglewicz, G. (2018). Characterization of graphite oxide and reduced graphene oxide obtained from different graphite precursors and oxidized by different methods using Raman spectroscopy. *Materials*, 11(7):1,050. <https://doi.org/10.3390/ma11071050>
- Na, J.Y., Kang, B., Sin, D.H., Cho, K., and Park, Y.D. (2015). Understanding solidification of polythiophene thin films during spin-coating: Effects of spin-coating time and processing additives. *Scientific Reports*, 5:1-14. <https://doi.org/10.1038/srep13288>
- Navarrete, M.C., Díaz-Herrera, N., and González-Cano, A. (2023). Deposition of Graphene Oxide on an SPR Fiber Refractometer for Sensor Applications. *Sensors*, 23(8):4,098. <https://doi.org/10.3390/s23084098>
- Oo, N.B., Wipataphan, P., Panomsuwan, G., Pewnim, N., Jongprateep, O., and Techapiesancharoenkij, R. (2022). Effect of Substrate and Spin-Coating Parameters on Structures of ZnO Nanorod and PvdF Film. *Suranaree Journal of Science and Technology*, 29(6):1-7.
- Pawlus, P., Reizer, R., and Wieczorowski, M. (2021). Functional importance of surface texture parameters. *Materials*, 14(18):1-29. <https://doi.org/10.3390/ma14185326>
- Putri, N.P., Suaebah, E., Rohmawati, L., Santjojo, D.J.D.H., Masruroh, M., and Sakti, S.P. (2023). Implications of the Electrodeposition Scan Rate on the Morphology of Polyaniline Layer and the Impedance of a QCM Sensor. *Trends in Sciences*, 20(3):6,411. <https://doi.org/10.48048/tis.2023.6411>
- Qiao, X., Zhang, X., Tian, Y., and Meng, Y. (2015). Modeling the response of a quartz crystal microbalance under nanoscale confinement and slip boundary conditions. *Physical Chemistry Chemical Physics*, 17(11):7,224-7,231. <https://doi.org/10.1039/C4CP05141F>
- Tang, X., Debliqy, M., Lahem, D., Yan, Y., and Raskin, J.P. (2021). A review on functionalized graphene sensors for detection of ammonia. *Sensors*, 21(4):1,443. <https://doi.org/10.3390/s21041443>
- Tiwari, S.K., Kumar, V., Huczko, A., Oraon, R., Adhikari, A. De., and Nayak, G.C. (2016). Magical Allotropes of Carbon: Prospects and Applications. *Critical Reviews in Solid State and Materials Sciences*, 41(4):257-317. <https://doi.org/10.1080/10408436.2015.1127206>
- Yang, Y., Knust, S., Schwiderek, S., Qin, Q., Yun, Q., Grundmeier, G., and Keller, A. (2021). Protein adsorption at nanorough titanium oxide surfaces: The importance of surface statistical parameters beyond surface roughness. *Nanomaterials*, 11(2):1-15. <https://doi.org/10.3390/nano11020357>
- Zubar, T.I., Sharko, S.A., Tishkevich, D.I., Kovaleva, N.N., Vinnik, D.A., Gudkova, S.A., Trukhanova, E.L., Trofimov, E.A., Chizhik, S.A., Panina, L.V., Trukhanov, S.V., and Trukhanov, A.V. (2018). Anomalies in Ni-Fe nanogranular films growth. *Journal of Alloys and Compounds*, 748:970-978. <https://doi.org/10.1016/j.jallcom.2018.03.245>