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Effect of Dip-Dry Coating Cycles on the Uniformity and Structure of Graphene Oxide and Reduced Graphene Oxide on Cotton Fabric Electrodes

Mohammad Azwan Safwan Harun^{1*}, Mastura Shafinaz Zainal Abidin^{1[0000-0002-3265-2164]**}, Shaharin Fadzli Abdul Rahman^{1[0000-0001-5106-5320]}, Zulkarnain Ahmad Noorden^{2[0000-0002-4639-2164]}, and Amirjan Nawabjan¹

¹Faculty of Electrical Engineering, Universiti Teknologi Malaysia, 81310 UTM Johor Bahru, Johor, Malaysia

²Institute of High Voltage & High Current, Faculty of Electrical Engineering, Universiti Teknologi Malaysia, 81310 UTM Johor Bahru, Johor, Malaysia

*mohammadazwansafwan@graduate.utm.my, **m-shafinaz@utm.my

Abstract. Graphene-based textile electrodes have received much attention for wearable energy storage due to light weight, flexibility and excellent electrical properties. However, conventional fabrication techniques usually suffer from poor coating uniformity and limited scalability and durability. This work investigates the effect of dip-dry coating cycles on the uniformity and structure of graphene oxide (GO) and reduced graphene oxide (rGO) coatings on cotton fabric electrodes. Circular cotton fabric substrates (diameter 24 mm) were coated by a simple and scalable dip-dry process with 2 and 10 coating cycles with GO and rGO aqueous dispersions. Scanning electron microscopy (SEM) revealed that increasing the coating cycles from 2 to 10 transformed the coating from a partial, non-uniform deposition to a dense and continuous graphene network encapsulating the cotton fibers. Energy-dispersive X-ray spectroscopy (EDX) confirmed this trend, the GO-coated samples showed a C/O weight ratio of 4.15 at 2 cycles and 1.50 at 10 cycles, reflecting the transition from localized to full-coverage deposition, while rGO-coated samples showed an increasing C/O ratio from 1.53 (2 cycles) to 1.92 (10 cycles), consistent with progressive rGO layer accumulation. Raman spectroscopy confirmed clear structural differences between GO and rGO coatings. The average D-band to G-band intensity ratio (I_D/I_G) ranged from 0.91 to 0.92 for the GO-coated samples and from 0.98 to 0.99 for the rGO-coated samples. These results demonstrate that the coating cycle number can be an important factor in the structural quality of graphene-coated textile electrodes and provide meaningful insight for the design of flexible and wearable supercapacitor applications.

Keywords: Graphene oxide (GO), Reduced graphene oxide (rGO), Dip-dry coating, Cotton fabric electrode, Wearable supercapacitor.

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1 INTRODUCTION

Since its first experimental isolation [1] graphene has attracted tremendous attention as a building block for next-generation flexible electronics due to its outstanding electrical conductivity and physical properties. The rapid expansion of flexible and portable electronics, smart textiles, and Internet of Things (IoT) enabled health monitoring systems has made wearable energy storage devices the focus of research [2]. Among the candidate materials for flexible electrodes, graphene and its derivatives, such as graphene oxide (GO) and reduced graphene oxide (rGO), are particularly attractive due to their high specific surface area, excellent electrical conductivity, mechanical flexibility, and chemical stability [3].

Textile materials, especially cotton fabric, are suitable substrates for wearable supercapacitors because they are flexible, breathable, inexpensive, and easy to include in clothing [4]. Several graphene-based and hybrid coatings have been studied to improve the performance of cotton-based electrodes. These include MXene/rGO/cellulose nanocrystal composites [5] silver nanoparticle/rGO coatings [6], polyaniline-coated rGO [7] and rGO/MnO₂/polyaniline composites [8]. Carbon nanotube (CNT) coatings and CNT/GO hybrid materials have also been explored as alternative conductive materials [9][10]. However, achieving a uniform coating on cotton fabric is still difficult because cotton has a porous and uneven fibrous structure. Therefore, the coating method plays an important role in determining the uniformity, electrical conductivity, and mechanical strength of the final electrode [11].

The dip-dry coating method is commonly used because it is simple, low-cost, and suitable for coating textile materials with GO and rGO dispersions on a larger scale [11]. Shateri-Khalilabad and Yazdanshenas [12] showed that cotton fabric could become electrically conductive after repeated dipping in GO solution followed by chemical reduction. Later, Karim et al. [13] applied a similar method on a larger scale to produce wearable graphene-based electronic textiles. Previous studies also showed that the number of coating cycles affects the amount of graphene deposited on the fabric. Chatterjee et al. [14] found that the graphene content of woven and knitted cotton increased when the GO concentration and number of dipping cycles were increased. Similar studies using viscose and para-aramid fabrics have shown that increasing the number of coating cycles can improve surface coverage and electrical conductivity. However, this improvement normally occurs only until an optimum number of coating cycles is reached [15][16].

The type of graphene material used, either GO or rGO, can also affect the electrical and structural properties of the coating. GO is commonly produced using the Hummers method [17] or a modified Hummers method [18]. It contains several oxygen-containing functional groups, including hydroxyl, epoxy, carbonyl, and carboxyl groups. These functional groups allow GO to disperse well in water, but they also reduce its electrical conductivity. On the other hand, rGO contains fewer oxygen-containing groups because part of its sp² carbon structure has been restored. As a result, rGO generally has better electrical conductivity than GO [19]. However, rGO has its own limitation. The rGO sheets can restack or form agglomerates when dispersed in water, which may lead to an uneven coating on the fabric surface [20][21][22]. In this

study, commercial GO and rGO dispersions were used directly as received. No additional dilution or reduction treatment was carried out.

Raman spectroscopy is commonly used to study the structural properties of GO and rGO. One important parameter obtained from Raman analysis is the D-band to G-band intensity ratio (I_D/I_G). This ratio is commonly used to indicate the level of defects and disorder in carbon-based materials [23][24]. Besides studying the coating structure, this work is also related to the possible use of the coated fabrics as electrodes for supercapacitors. Carbon-based materials have been widely studied for supercapacitor electrodes because of their potential to provide good capacitance and long cycle life [3][25][26].

Previous studies have shown that increasing the number of coating cycles can improve graphene deposition, surface coverage, and electrical conductivity on textile substrates [14][15][16]. However, there are still limited studies that directly compare GO and rGO coatings prepared using the same fabric, pretreatment method, and coating conditions. There is also limited information on how the number of dip-dry coating cycles affects the coating uniformity, surface morphology, elemental composition, and structure of GO- and rGO-coated cotton fabrics. Therefore, this study compares both materials under the same processing conditions using SEM, EDX, and Raman spectroscopy.

In this study, all cotton samples were first pretreated using acetone and deionized water to provide a clean and consistent surface before the coating process. Commercial GO and rGO dispersions were then coated onto the cotton fabric using 2 and 10 dip-dry coating cycles. The same processing conditions were used for both materials so that a direct comparison could be made. After coating, SEM was used to observe the surface morphology and coating coverage, while EDX was used to examine the elemental composition. Raman spectroscopy was used to study the structural characteristics of the GO and rGO coatings. By comparing GO and rGO at different coating cycles, this study aims to determine how the type of graphene material and the number of coating cycles affect the properties of the coated cotton fabric. The findings from this work can provide an initial understanding of the coating process before further optimization is carried out. Electrical and electrochemical measurements will be conducted in future work to determine the suitability of these coated fabrics as flexible textile electrodes for energy-storage applications.

2 EXPERIMENTAL

The preparation process for the cotton fabric electrodes is shown in Figure 1. Untreated cotton fabric was first cut into circular disks with a diameter of 24 mm using a precision cutting tool. Before coating, the cotton samples were cleaned to remove oils, waxes, surface contaminants, and other impurities that could affect the coating quality. For this pretreatment step, the fabric disks were fully immersed in 99% acetone and left to soak for 1 hour at room temperature. After soaking, the samples were removed from the acetone and rinsed thoroughly with deionized (DI) water to remove the remaining

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solvent and dissolved impurities. They were then dried in an oven at 80 °C for 30 minutes before the coating process.

Commercial GO and rGO water dispersions (99.5% purity, 2.0 wt%) were purchased from Nanografi Advanced Materials, Ankara, Turkey. The materials were used as received without any further dilution or additional reduction treatment. The cleaned cotton samples were coated with either GO or rGO using the dip-dry method. For Steps 5 and 6 in Figure 1, each cotton sample was completely immersed in the corresponding GO or rGO dispersion for 30 minutes at room temperature. This gave enough time for the dispersion to wet the cotton surface and penetrate the fibers. The 30-minute soaking time was selected based on preliminary trials. From these trials, soaking the samples for more than 30 minutes did not show any noticeable improvement in the amount of material deposited on the cotton fibers. After the soaking step, the samples were taken out of the dispersion and hung vertically to allow the excess liquid to drain. The coated samples were then dried in a laboratory oven at 80 °C for 30 minutes. This drying step helped remove residual moisture and promote better adhesion of the coating to the cotton fibers. The immersion and drying steps together constituted one coating cycle. To examine the effect of repeated coating on material deposition, the cycle was repeated 2 and 10 times, resulting in samples with progressively higher graphene loading. The two cycle counts 2 and 10 were chosen to represent the lower and upper bounds of the optimization stage an early, partially coated state at one end and a condition approaching coverage saturation at the other. This choice was informed by preliminary trials and is consistent with earlier dip-dry coating studies [14][15][16]. The resulting samples were labeled GO-2, GO-10, rGO-2, and rGO-10, as summarized in Table 1.

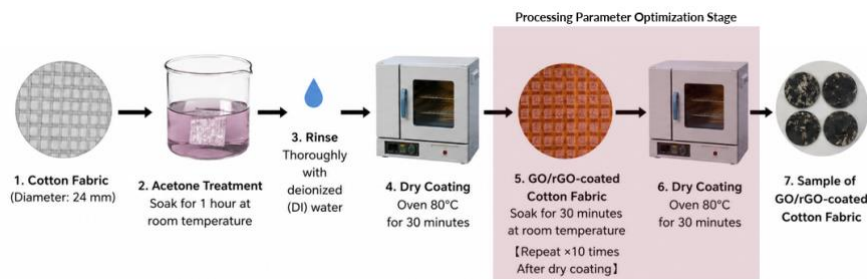


Figure 1: Process flow of the preparation, pretreatment, and dip-dry coating of GO- and rGO-coated fabric electrodes.

Table 1: Sample Labeling and Coating Parameters.

Coating Material on Cotton Fabric	2 Cycles	10 Cycles
Graphene Oxide (GO)	GO-2	GO-10
Reduced Graphene Oxide (rGO)	rGO-2	rGO-10

The surface morphology and elemental composition were examined using a Hitachi TM3000 scanning electron microscope (SEM) fitted with energy-dispersive X-ray spectroscopy (EDX), operated at 15.0kV in BSE COMPO mode. Raman spectra were

collected with an XploRA PLUS Raman spectrometer (HORIBA Scientific) using a 532 nm excitation laser.

3 RESULTS AND DISCUSSION

3.1 Effect of Coating Cycles on Surface Morphology

Figure 2 shows the SEM images of the bare cotton fabric and the GO- and rGO-coated samples after acetone and DI water pretreatment. As shown in Figure 2(a), the bare cotton fibers have a relatively smooth and clean surface. The cylindrical shape of the fibers can still be clearly seen, and no obvious deposits are present on the surface. This suggests that the pretreatment helped remove surface impurities without causing noticeable changes to the original fiber structure. The individual cotton fibers are also clearly separated, with no material bridging between them.

After 2 coating cycles, Sample GO-2 in Figure 2(b) shows only partial coverage of GO on the cotton fibers. Thin and flake-like GO sheets can be observed on some parts of the fiber surface, while several areas of the cotton remain exposed. This indicates that the coating was not yet fully formed after only 2 cycles. Most of the fibers can still be identified individually, and only a small amount of bridging can be seen between nearby fibers. This appearance is consistent with the early stage of GO deposition during the dip-dry coating process [14].

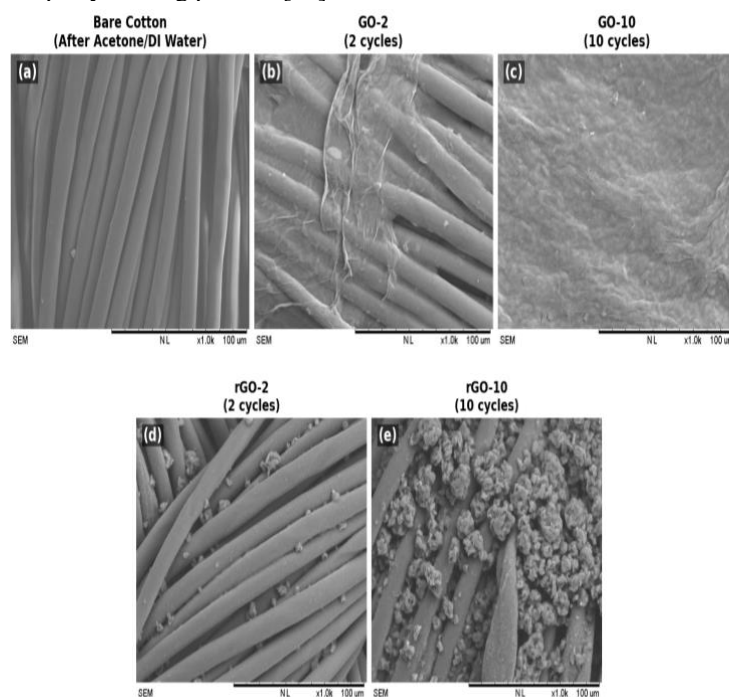


Figure 2: SEM images (×1000) of (a) bare cotton after acetone/DI water pre-treatment, (b) GO-2, (c) GO-10, (d) rGO-2, and (e) rGO-10

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In contrast, a significant change in fiber surface morphology was observed after 10 GO coating cycles (Sample GO-10), as shown in Figure 2(c). A denser and more continuous GO coating covers the cotton fibers, with less exposed fiber surface visible. The coating appears more homogeneous across the fiber surface compared to GO-2, and early signs of more inter-fiber bridging through the GO network are observable, suggesting the formation of a progressively interconnected conductive pathway with increasing cycle numbers.

As can be seen from Figure 2(d), the coating morphology for sample rGO-2 differs from its counterpart at the same cycle number that is GO-2 sample. The rGO flakes appear more aggregated and slightly rougher in texture, consistent with the tendency of rGO whose reduction removes stabilizing oxygen functional groups to undergo partial restacking and agglomeration on the fiber surface. Despite this, the rGO coating still provides partial fiber encapsulation with visible coverage of the cotton surface.

By contrast, sample rGO-10 (Figure 2(e)) shows by far the most extensive coverage of any sample examined. A thick, continuous rGO network wraps around the fibers and largely obscures the underlying fiber texture, bridging adjacent fibers to form an interconnected, three-dimensional conductive network across the fabric. These morphological observations are consistent with the progressive layer-by-layer accumulation reported in dip-dry coating studies [12][14][15].

To further illustrate the cycle-dependent coverage transition, Figure 3 presents SEM images comparing 2 and 10 cycle coatings for both GO and rGO. At $\times 500$ magnification, the transition from partial to near-complete fiber encapsulation with increasing coating cycles is clearly visible for both materials, while the GO coating exhibits a smoother, more conformal morphology than the rougher, more aggregated rGO coating.

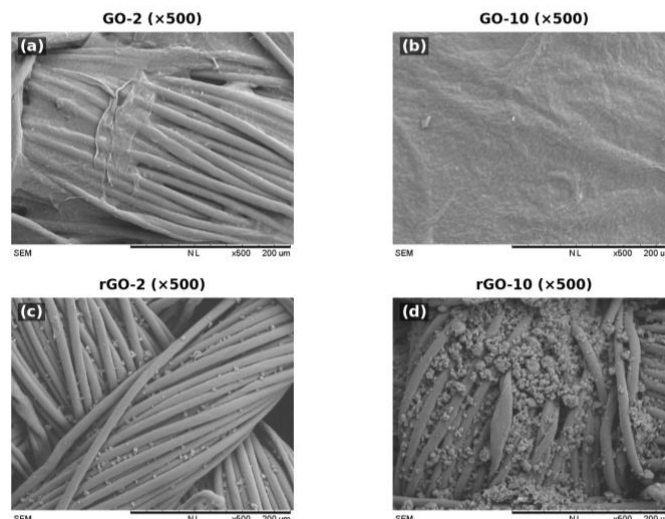


Figure 3: SEM images at $\times 500$ magnification: (a) GO-2, (b) GO-10, (c) rGO-2, (d) rGO-10 coated cotton fabric, showing the effect of coating cycle number on graphene coverage and network continuity.

3.2 Elemental Composition Analysis

EDX analysis was performed to quantify the elemental composition of the cotton fabric samples at each processing stage. Figure 4 shows EDX spectra for all samples in this study. Figure 5 summarizes the carbon (C) and oxygen (O) weight percentages, and the resulting C/O weight ratios, for the bare cotton (after acetone/DI water pretreatment), GO-2, GO-10, rGO-2, and rGO-10 samples based on the EDX spectra.

The bare cotton substrate after acetone/DI water cleaning shows a C content of 70.09 wt.% and O content of 29.91 wt.%, with a C/O weight ratio of 2.34, consistent with the predominantly cellulosic composition of natural cotton fibers (approximately 90% cellulose, which has a theoretical C/O atomic ratio of 1:1) [27]. The detected carbon signal in bare cotton arises from the cellulose backbone, while the oxygen signal reflects the abundant hydroxyl groups on the cellulosic surface.

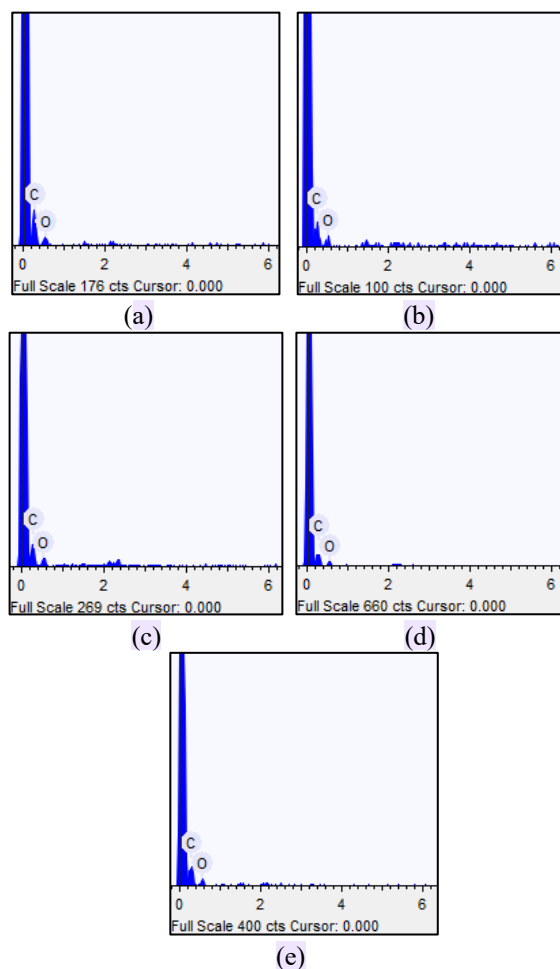


Figure 4: EDX spectrum of (a) bare cotton fabric after acetone and DI-water pretreatment, (b) GO-2, (c) GO-10, (d) rGO-2, and (e) rGO-10

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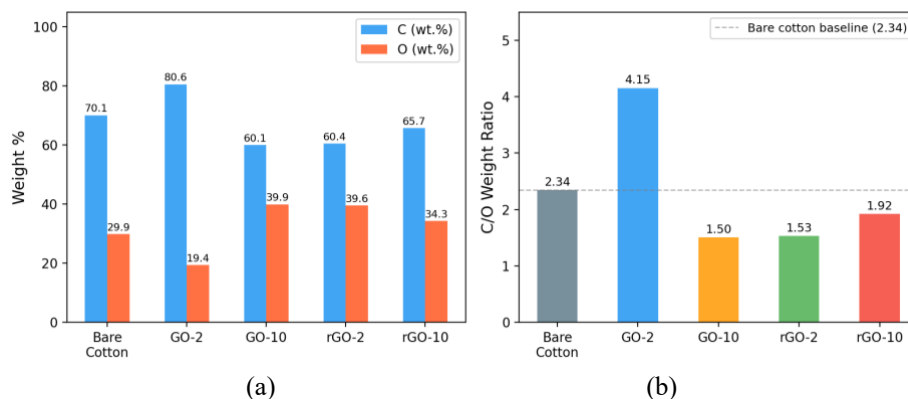


Figure 5: EDX analysis results: (a) C and O elemental weight percentages and (b) C/O weight ratios for bare cotton, GO-2, GO-10, rGO-2, and rGO-10 coated cotton fabric samples.

The EDX result for GO-2 shows a high carbon content of 80.57 wt.% and a C/O ratio of 4.15. This value is higher than those obtained for the bare cotton and GO-10 samples. The higher C/O ratio may be influenced by the underlying cellulose substrate rather than representing only the GO coating. As observed from the SEM image, the GO-2 sample has a thin and incomplete coating, with some parts of the cotton fibers still exposed. Because the EDX analysis was carried out at 15kV, the interaction volume can extend through the thin GO layer and detect signals from the cotton fiber underneath. Since bare cotton itself has a relatively high C/O ratio (2.34), this substrate contribution adds proportionally more carbon than oxygen to the measured signal for the thin, patchy GO-2 coating, elevating the apparent C/O ratio above that of the more complete, coating-dominated GO-10 sample (1.50). This substrate-interference effect diminishes once a denser, more continuous coating is formed at 10 cycles, so the GO-10 and rGO datasets are considered more representative of the coating material itself. We retain the GO-2 EDX data point, as it was consistent across repeated spectra and directly corroborates the partial and inhomogeneous coverage observed by SEM. However, it should be interpreted as substrate-influenced rather than as a direct measure of the GO-2 coating composition.

By contrast, GO-10 shows a lower C content (60.06 wt.%) and higher O content (39.94 wt.%), resulting in a reduced C/O ratio of 1.50. The increased oxygen content in GO-10 may be attributed to the greater amount and coverage of deposited GO resulting from the higher number of coating cycles. The abundant oxygen-containing functional groups in GO, including hydroxyl, epoxy, carbonyl, and carboxyl groups, contribute significantly to the detected oxygen signal [18][23]. The lower C/O ratio in GO-10 compared to GO-2 indicates that the GO coating at 10 cycles provides a more complete and representative measurement of the GO material's inherent high oxygen content and is less affected by substrate interference than the thinner GO-2 coating.

The rGO-2 sample exhibited a C content of 60.43 wt.% and an O content of 39.57 wt.% (C/O = 1.53), which is very similar to that of GO-10. In contrast, the rGO-10 sample exhibited a slightly higher C content (65.74 wt.%) and a lower O content (34.26

wt.%), resulting in an increased C/O ratio of 1.92. The higher C/O ratio in rGO-10 compared with rGO-2 is consistent with the expected effect of increasing coating deposition. As additional rGO layers were deposited, the relative contribution of the reduced graphene material, which contains fewer oxygen-containing functional groups than GO, to the EDX signal increased [19] [28]. The progressive increase in C/O ratio from rGO-2 (1.53) to rGO-10 (1.92) is consistent with the SEM observations of greater rGO coverage and accumulation on the cotton fiber surface with increasing coating cycle number.

To provide quantitative, non-destructive evidence of the surface changes associated with the coating build-up observed in the SEM images, atomic force microscopy (AFM) was performed on the coated cotton fabric samples over a $10 \times 10 \mu\text{m}^2$ scan area for each sample. In the present study, AFM characterization was completed for the GO-coated samples (GO-2 and GO-10), while measurements for the corresponding rGO samples are still in progress. Therefore, only the GO samples are included in the present AFM analysis. Table 2 summarizes the average surface height, average roughness (Ra), root-mean-square roughness (Rq), and peak-to-valley roughness (Rt) extracted from the AFM height data. For the GO-coated samples, the average surface height and surface roughness (Ra and Rq) increased from GO-2 to GO-10, as shown in Figure 6 and summarized in Table 2. This increase in surface height and roughness is consistent with greater accumulation and coverage of GO on the cotton fibers with increasing coating cycle number. The AFM results therefore provide additional quantitative evidence supporting the morphological changes observed in the SEM images.

Table 2: AFM Surface Height and Roughness Parameters ($10 \times 10 \mu\text{m}^2$ Scan Area).

Sample	Average Height (nm)	Ra (nm)	Rq (nm)	Rt (μm)
GO-2	68.18	114.0	154.9	1.457
GO-10	88.69	144.4	173.5	1.179

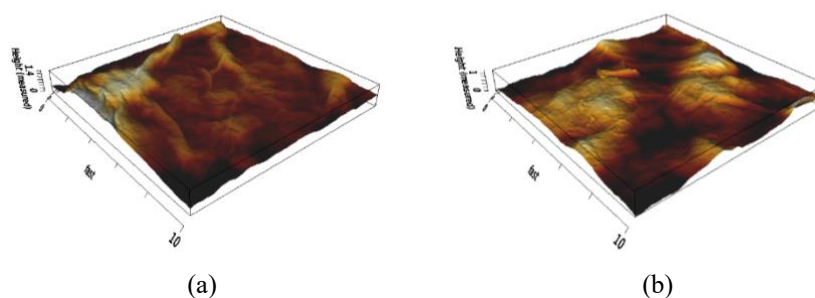


Figure 6: AFM height images ($10 \times 10 \mu\text{m}^2$ scan area) of (a) GO-2, and (b) GO-10 coated cotton fabric.

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3.3 Structural Characterization using Raman Spectroscopy

Raman spectroscopy was used to investigate the structural characteristics of GO and rGO coatings at 2 and 10 dip-dry cycles. Figure 7 shows the Raman spectra of all four samples over the spectral range from 0 to 4000 cm^{-1} . All four samples display the characteristic D-band ($\sim 1350 \text{ cm}^{-1}$) and G-band ($\sim 1580 \text{ cm}^{-1}$) associated with disordered sp^3 carbon and ordered sp^2 graphitic carbon, respectively [23][24]. A broad, low-intensity feature is also observed near $\sim 2700 \text{ cm}^{-1}$ for all four samples, corresponding to the 2D band. Its broadened, low-intensity profile relative to the G-band is characteristic of disordered, and multilayer stacked GO/rGO sheets rather than few-layer graphene and is consistent with the defect-rich structure indicated by the I_D/I_G ratios, it was not used for quantitative analysis in this study because reliable 2D-band fitting requires higher spectral resolution than used here.

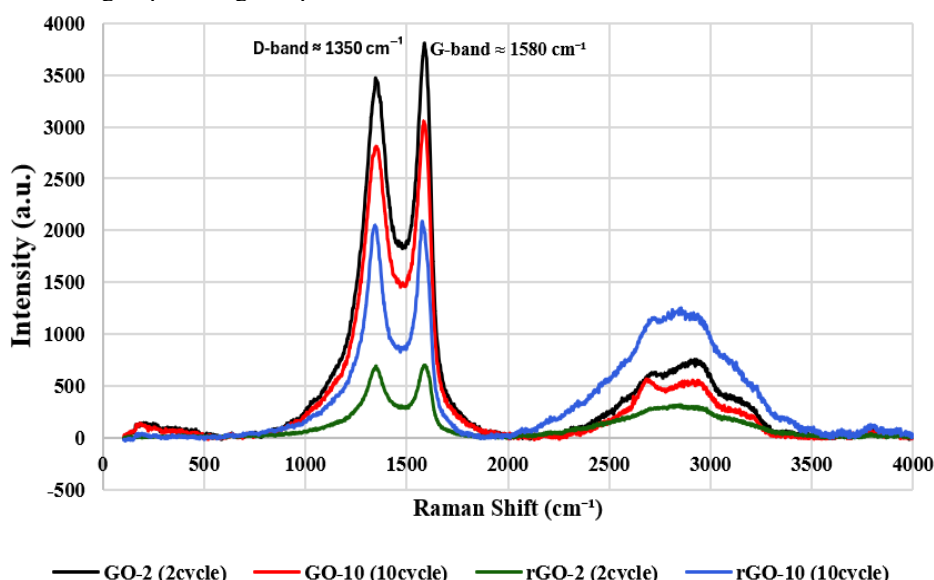


Figure 7: Raman spectra of GO-2, GO-10, rGO-2, and rGO-10 coated cotton fabric.

Table 3 presents the D-band and G-band peak positions and their corresponding intensities for the four coated cotton fabric samples, while Figure 8 summarizes the calculated I_D/I_G ratios. The GO-2 sample showed D- and G-band peaks at 1345.52 cm^{-1} and 1587.32 cm^{-1} , respectively, with an I_D/I_G ratio of 0.9115. For GO-10, the D- and G-band peaks were located at 1348.22 cm^{-1} and 1584.70 cm^{-1} , giving a slightly higher ratio of 0.9197. The small difference between the two GO samples suggests that increasing the coating cycles from 2 to 10 produced only a minor change in the structural disorder of the GO coating.

Table 3: Raman D-Band and G-Band Characteristics.

Sample	D peak position (cm ⁻¹)	I _D	G peak position (cm ⁻¹)	I _G
GO-2	1345.52	3476.27	1587.32	3813.59
GO-10	1348.22	2812.87	1584.70	3058.38
rGO-2	1348.22	695.34	1587.32	702.65
rGO-10	1342.82	2055.19	1576.85	2093.07

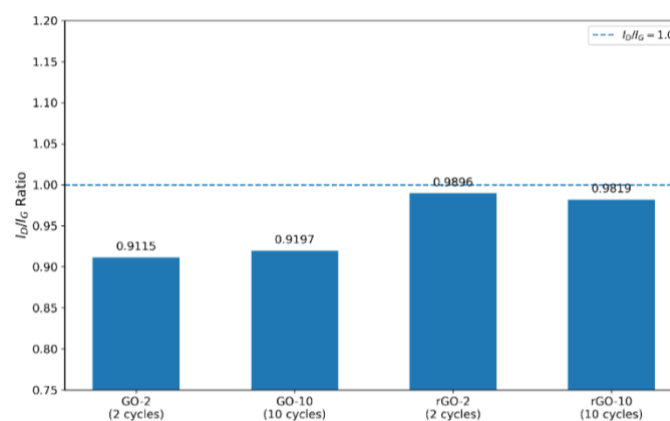


Figure 8: I_D/I_G ratio comparison for GO-2, GO-10, rGO-2, and rGO-10 samples.

The rGO samples showed higher I_D/I_G ratios than the GO samples. The rGO-2 sample recorded the highest ratio of 0.9896, with D- and G-band positions at 1348.22 and 1587.32 cm⁻¹. Meanwhile, rGO-10 showed an I_D/I_G ratio of 0.9819, with the D and G peaks located at 1342.82 and 1576.85 cm⁻¹, respectively. The higher ratios observed for the rGO samples indicate a greater level of structural disorder or smaller sp² carbon domains compared with the GO-coated samples.

A lower G-band position was also observed for rGO-10 compared with the other samples. This shift may be related to differences in the graphitic structure, sheet arrangement, and coating thickness resulting from repeated dip-dry cycles. Overall, the Raman results indicate that both the type of graphene material and the number of coating cycles affected the structural characteristics of the coating. However, as the analysis was based on single measurement point for each sample, the results should be treated as preliminary observations, and additional measurements are required to confirm the trends. Despite this limitation, the Raman analysis reveals distinct structural differences between GO and rGO coatings, highlighting the combined influence of the chemical state (GO vs. rGO) and the number of dip-dry coating cycles on the graphene coating formed on cotton fabric.

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4 CONCLUSIONS

This study investigated the effect of 2 and 10 dip-dry coating cycles on the uniformity and structural properties of GO- and rGO-coated circular cotton fabric electrodes with a diameter of 24 mm. The SEM results showed that increasing the number of coating cycles improved the coverage of graphene-based material on the cotton fibers. The 10-cycle samples generally formed a more continuous and connected coating than the 2-cycle samples. The EDX results showed that the C/O weight ratio of the GO-coated samples decreased from 4.15 for GO-2 to 1.50 for GO-10. In contrast, the C/O ratio of the rGO-coated samples increased from 1.53 for rGO-2 to 1.92 for rGO-10. These differences may be related to variations in coating coverage, material distribution, coating thickness, and the selected EDX measurement area.

Raman analysis also showed structural differences among the coated samples. The I_D/I_G ratios were 0.9115 for GO-2, 0.9197 for GO-10, 0.9896 for rGO-2, and 0.9819 for rGO-10. The GO samples showed only a small change in the I_D/I_G ratio when the number of coating cycles increased. For the rGO samples, the lower ratio observed for rGO-10 may indicate differences in sheet arrangement, structural disorder, or the size of the sp^2 carbon domains. The shift in the G-band position of rGO-10 also suggests a change in the carbon bonding environment, which may be influenced by coating thickness, sheet interaction, structural strain, or interaction with the cotton substrate.

Overall, the results indicate that increasing the number of dip-dry coating cycles improves the surface coverage and structural characteristics of GO- and rGO-coated cotton fabrics. These findings provide a basis for optimizing the coating process prior to subsequent electrical and electrochemical characterization, which will be required to evaluate the potential of the coated fabrics for flexible and wearable supercapacitor applications.

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