

Synthesis of Hybrid Graphene Oxide Membranes for Water Purification

Group 1-04

Abstract

This study aims to create a low-cost hybrid graphene oxide membrane to purify water through heavy metal ion and organic dye adsorption, as well as the removal of bacteria. Graphene Oxide (GO) was synthesized from Graphite powder using a method without many of the limitations of Hummer's method, such as the evolution of toxic gas from the process. The GO powder was further modified to create Carboxyl GO (CGO), and investigated to determine the benefits of this modification. Fourier Transform Infrared (FTIR) Spectroscopy, Ultraviolet-Visible (UV-Vis) Spectroscopy and X-Ray Diffraction (XRD) tests were performed to ascertain the identity of the GO and CGO powders. GO and CGO were dispersed in Cu^{2+} , Zn^{2+} and organic dye solutions and stirred overnight, with the percentage absorbance being measured with a colourimeter for heavy metal ions, and concentration of residual dye was determined by measuring the absorbance at a maximum absorbance wavelength using a UV-Vis spectrophotometer. To counteract some complications arising from removing the powders from the dispersions, hybrid GO and CGO membranes were prepared, and Cu^{2+} , Zn^{2+} and organic dye solutions were passed through them, and absorbance is taken in a similar manner. Antibacterial Tests were also performed with *Escherichia coli* bacteria, using the spread plate method. These tests had shown GO and CGO to be highly capable in removing Cu^{2+} (GO: 82.02%; CGO: 98.13%), Zn^{2+} (GO: 75.85%; CGO: 91.16%) and the organic dyes Methylene Blue (GO: 96.83%; CGO: 89.95%) and Methyl Orange (GO: 88.68%; CGO: 99.19%), as well as having remarkable antibacterial properties (GO: 92.08%; CGO: 92.61%). Even after desorption, GO and CGO membranes retain significant Cu^{2+} (GO: 63.90%→72.29%; CGO: 68.17%→52.78%), Zn^{2+} (GO: 75.58%→68.68%; CGO: 59.25%→62.91%), Methyl Orange (GO: 67.78%→80.55%; CGO: 44.30%→58.00%) and Methylene Blue (GO: 93.76%→100%; CGO: 96.62%→65.33%) adsorption capabilities.

1. Introduction

Water is an essential resource for sustaining human life and development. Nevertheless, advancements in technology and rapid industrialization across the globe has meant that water bodies worldwide have been contaminated with various pollutants, such as heavy metals and organic dyes. Heavy metals are non-biodegradable and highly toxic if ingested, leading to long-term detrimental effects on health (Jan *et al.*, 2015).

Graphene Oxide is the oxidised form of graphene. It is a single-atomic-layered material formed by the oxidation of graphite which is cheap and readily available. It is also easy to process since it is dispersible in water (Lauren, 2018). The material has been widely researched in recent times, due to its many capabilities including in adsorbing organic dyes (Molla *et al.*, 2019), and possesses antibacterial and antimicrobial properties (Di Guilo *et al.*, n.d.). Its antibacterial activity towards *Escherichia coli* also exceeds that of similar compounds such as reduced graphene oxide, graphite and graphite oxide (Liu *et al.*, 2011).

The synthesis of Graphene Oxide is typically done through Hummer's method, where graphite flakes and NaNO₃ are stirred in concentrated H₂SO₄ solution, while KMnO₄ can be added to oxidise the graphite into graphene oxide (Paulchamy & Lignesh, 2015) Following purification, filtration and drying can then be used to obtain a Graphene Oxide powder. However, this method, while well known, still has various flaws. These range from toxic gases such as NO₂ and N₂O₄ being evolved to residual nitrate contaminating the product. (Yu *et al.*, 2016) Hummer's method can also cause complications in cleanup, due to the presence of Na⁺ and NO₃⁻ ions in the wastewater. (Chen *et al.*, 2013) Furthermore, NaNO₃, a crucial component of the Hummer's Method, is a strong oxidizer and can cause various health complications, simply from being inhaled.

Due to the complications arising from Hummer's method, many have sought alternatives that do not require the use of NaNO₃, as it is the root cause of many of the issues of this method. In research from Zaaba *et al.*, 2017, a modified Hummer's method has been suggested. They replaced the use of NaNO₃ with H₂SO₄, H₃PO₄ and doubled the amount KMnO₄ recommended in Hummer's method. The Graphene Oxide produced through this method has advantages over Hummer's method, such as better conductivity and eliminating the emission of toxic gases during production. Chen *et al.*, 2013, synthesized graphene oxide using graphite powder, H₂SO₄ and KMnO₄ under an ice bath followed by an oil bath and the addition of H₂O₂. This gave a yield of 92%±3% which is very close to that for graphene oxide synthesized using Hummer's method (96%±2%). The graphene oxide synthesized was also nearly identical to that made by Hummer's method in the areas of composition, structure and morphology.

Graphite could bring forth even more benefits when its synthesized Graphene Oxide form is further modified. One example of such modification is the synthesis of Carboxyl Graphene Oxide (CGO) by Li *et al.*, 2017, which allowed the creation of high-performance capacitors. Si & Samulski, 2008, also chemically reduced graphene oxide by using sodium borohydride, removing most of the oxygen-containing functional groups, creating graphene that possesses electrical conductivity similar to that of graphite.

Graphene oxide is also frequently used to create membranes with various benefits. By fabricating a combination of polycrystalline large-area High Strength Metallurgical Graphene (HSMG®), graphene oxide, hydrazine and an in-situ interfacial polymerized polyamide on polysulfone porous scaffolding, composite filters with ion blocking efficiency of over 95% were synthesised for use in water desalination (Romaniak *et al.*, 2020). Graphene oxide is also suitable to be used as a membrane for water purification since it possesses strong antifouling properties that help prolong its lifespan (Hu *et al.*, 2016).

This study aims to create a low-cost hybrid graphene oxide membrane to purify water through heavy metal ion and organic dye adsorption, as well as the removal of bacteria.

1.1. Hypotheses

The hypotheses of this study are that: Graphene Oxide can be synthesized from graphite; Hybrid Graphene Oxide membranes can adsorb heavy metal ions; Organic dyes can be adsorbed/broken down by hybrid graphene oxide membranes; Carboxyl Graphene Oxide maintains its antibacterial properties.

2. Materials and Methods

2.1. Materials

All chemicals were obtained from Sigma Aldrich. All apparatus was obtained from the Biology and Chemistry Laboratories in the SRC.

2.2. Methods

2.2.1. Synthesis of Graphene Oxide

Graphite powder (1.5 g) was added to concentrated H₂SO₄ (95%, 35 mL) under stirring in an ice bath. Under vigorous agitation, KMnO₄ (4.5 g) was added slowly to keep the temperature of the suspension lower than 20 °C. Successively, the reaction system was heated at 40 °C and vigorously stirred for about 0.5 h. Then, 75 mL water was added, and the solution was stirred for 15 min at 95 °C. Additional 250 mL water was added and followed by a slow addition of 7.5 mL H₂O₂ (6%), turning the colour of the solution from dark brown to yellow. The mixture was filtered and washed with 5% aqueous HCl solution (125 mL) to remove metal ions. The resulting solid was washed with deionised water and then diluted to 600 mL, stirred overnight and sonicated for 30 min to exfoliate it to GO. The GO dispersion was then centrifuged at 3000 rpm for 20 min to remove the unexfoliated graphite. The GO powder was

characterized by FTIR and UV-Vis Spectroscopy, as well as X-Ray Diffraction (XRD) to ascertain its identity.

2.2.2. Preparation of Carboxyl Graphene Oxide (CGO)

GO (0.75 g) was first dispersed in distilled water. 50 mL of HBr was added to the GO suspension and the mixture was stirred for 12 h at room temperature. Oxalic acid (15.0g) was then added and the mixture kept stirring for another 4 h. CGO was obtained after centrifugation, lavation and vacuum drying at 50 °C. The CGO was characterized by FTIR and UV-Vis Spectroscopy, as well as X-Ray Diffraction (XRD) to ascertain its identity.

2.2.3. Preparation of Hybrid Graphene Oxide Membrane

GO membranes were fabricated using aqueous GO suspension on a commercial Whatman filter paper. 25 mg of GO was suspended in 20 ml of deionised water. Using a Buchner funnel, the GO suspension was deposited on the surface of the filter paper dropwise. GO suspension deposited on the membranes was dried in stagnant air. Air-dried GO on the film was then incubated in the oven at 90 °C overnight. Before use, a piece of filter paper was placed on top of and below the GO membrane layer each, and the pieces of filter paper were sealed with water. This was to prevent the dispersion of GO when it comes into contact with the necessary solutions, and to increase the contact time between the GO and necessary solutions since the rate at which these solutions flow through the filter is reduced. The above was repeated for CGO.

2.2.4. Heavy Metal Ion Adsorption Test

For Graphite (Gt), GO and CGO as adsorbents, 20 mL of Zn^{2+} (50 ppm) solution was prepared, to which 0.20 g of GO was added. The mixture was left to stir overnight before being centrifuged at 13000 rpm in 15 min intervals until a clear solution was obtained. The initial and final concentration of Zn^{2+} ions were determined by a colourimeter to calculate the percentage adsorption of heavy metal ions. The above procedure was repeated for Zn^{2+} with Gt, Zn^{2+} with CGO, Cu^{2+} with Gt, Cu^{2+} with GO and Cu^{2+} with CGO.

For GO and CGO membranes, 100 mL of Zn^{2+} and 100 mL of Cu^{2+} solutions were prepared. 50 mL of each solution was individually passed through the GO membrane 6 times. The initial and final concentration of Zn^{2+} and Cu^{2+} ions were determined by a colourimeter to

calculate the percentage adsorption of heavy metal ions. The above procedure was repeated for CGO membranes.

2.2.5. Organic Dye Sorption Test

Dilute solutions of methyl orange and methylene blue dyes (50 ppm) were prepared by dissolving the organic materials in deionised water. Calibration curves were plotted with a series of 5 different concentrations with absorbance measured by a UV-Vis spectrophotometer (Figures 2.2.5A and 2.2.5B).

For Gt, GO and CGO as adsorbents, 20 mL of each solution was mixed with 0.20 g of adsorbent and magnetically stirred overnight. The resulting solution was centrifuged at 13000 rpm in 15 min intervals until all adsorbent particles are removed. The concentration of residual dye was determined by measuring the absorbance at a maximum absorbance wavelength (464 nm for Methyl Orange; 668 nm for Methylene Blue) using a UV-Vis spectrophotometer.

For GO and CGO membranes, 50 mL of each solution was passed through GO and CGO membranes 6 times. The concentration of residual dye was determined by measuring the absorbance at a maximum absorbance wavelength (464 nm for Methyl Orange; 668 nm for Methylene Blue) using a UV-Vis spectrophotometer.

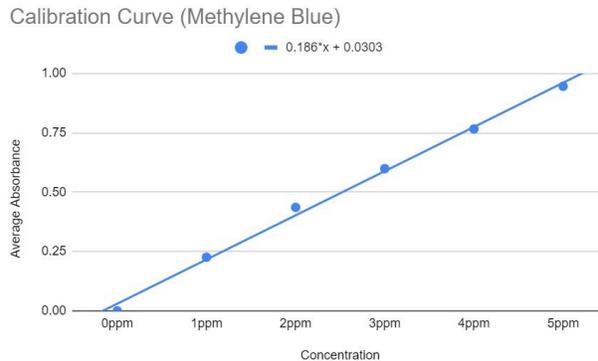


Figure 2.2.5A

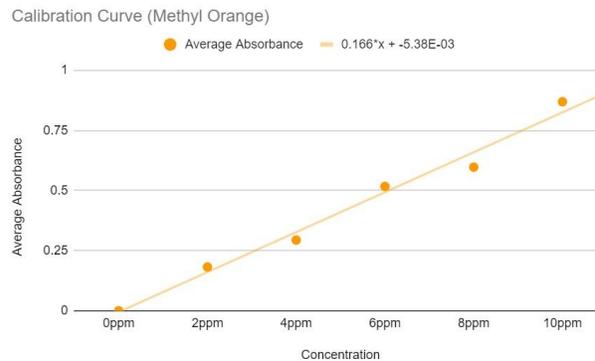


Figure 2.2.5B

2.2.6. Antibacterial Test

2.2.6.1. Preparation

To prepare the agar plates, 20 g of Luria-Bertani (LB) Agar powder was first dissolved in 500 mL of deionised water. Then, the mixture was shaken vigorously until all the powder had dissolved into the solution. Then, the mixture is autoclaved at 121 °C for 15 mins. The mixture was cooled with a water bath to 53 °C, and was subsequently poured into sterile petri dishes

inside a biological safety cabinet, filling them up approximately to the $\frac{1}{3}$ mark. The agar was then left to gel for 10 minutes.

To prepare the bacteria, LB Broth was first prepared. 2.5 g of LB Broth powder was dissolved into 100mL of deionised water inside a glass bottle. The bottle was then shaken until all powder dissolved into the water. The solution was then autoclaved at 121 °C for 15 minutes. The broth was left to cool in a water bath at ~57 °C. In a biological safety cabinet, the external surfaces of the broth bottle and a 15 mL centrifuge tube were flamed momentarily before opening, followed by the flaming of the internal surface. 9-10 mL of broth was added to the centrifuge tube. The metal ring was flamed till red hot, and was then contacted with the empty area of the agar to cool. It was then used to scoop *Escherichia coli* (*E. coli*) bacteria. With the *E. coli* introduced to the centrifuge tube, the centrifuge tube was then flamed at the mouth before sealing. The mixture was then left to be shaken for 24 hours to incubate the bacteria.

2.2.6.2. Spread Plate Method

E. coli, agar plates and 100 mL of 0.85% (by weight) saline were prepared. Saline solution was aliquoted into 900 μ L cuvettes. 50 mL of 1.00 g/L GO and CGO solutions were also prepared and sonicated for 30 min.

Inside a biological safety cabinet, 3 (three) 15 mL test tubes were labelled GO, CGO and Control. 0.5 mL of *E. coli* broth culture and 4.5mL of LB Broth was added to each test tube. Then, 0.5mL of GO solution, 0.5mL of CGO solution and 0.5mL of sterile water were added to their respective test tubes. The above process was replicated thrice. The test tubes were shaken overnight at 30 °C to incubate the bacteria.

1.5 mL of *E. coli* broth culture was introduced into a curvete, and was diluted with LB broth until the absorbance of the mixture was 0.800 (± 0.050) at 600 nm, using a UV-vis spectrophotometer. Serial dilution with saline was subsequently carried out. 100 μ L of the diluted mixture was introduced onto the surface of the agar, then spread and sealed with parafilm. They were left to incubate for 24 h at 37 °C and a colony count was performed thereafter.

2.2.7. Desorption for GO and CGO

Heavy metal ion loaded GO/CGO membranes were washed with 40 mL 5% nitric acid three times, and then with deionised water until the surface was pH neutral. Heavy metal ion adsorption test was subsequently performed.

Organic dye loaded GO/CGO membranes were soaked in 95% ethanol overnight. The membranes were washed with deionised water before an organic dye adsorption test was performed.

3. Results and Discussion

3.1. Fourier Transform Infrared Spectroscopy

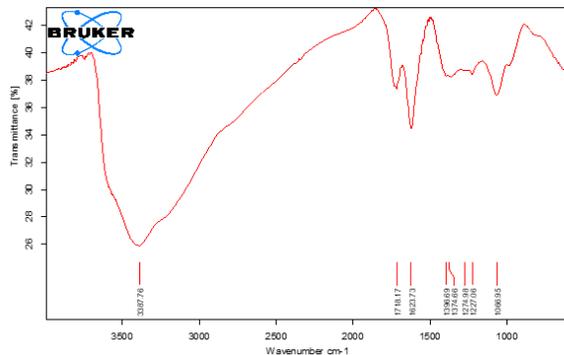


Figure 3.1A

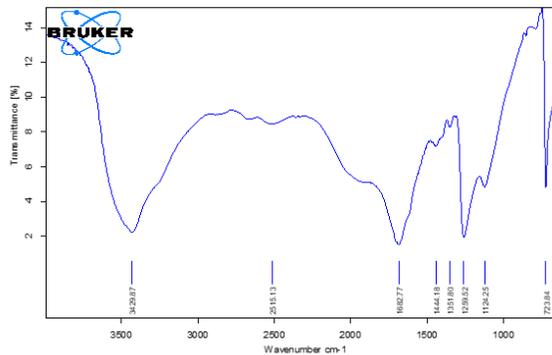


Figure 3.1B

Fourier Transform Infrared Spectroscopy was carried out on samples of GO (Figure 3.1A) and CGO (Figure 3.1B) after synthesis.

For GO, a broad peak at around 3387 cm^{-1} can be attributed to the stretching mode of O-H bonds, revealing its presence in GO. The sharp peak found at 1623 cm^{-1} is a resonance peak due to the stretching and bending vibration of OH groups of water molecules adsorbed on graphene oxide. The peak at 1227 cm^{-1} denotes C-O-C stretching and the peak at 1066 cm^{-1} corresponds to the C-O group's vibrational mode (Khalili, 2016).

CGO differs from GO mainly by the oxygen groups decomposed during carbonisation (C-O: 1718 cm^{-1} , C-O-C: 1066 cm^{-1}), as well as the addition of the -COOH peak at 1259 cm^{-1} (Li *et al.*, 2017).

3.2. Ultraviolet-Visible Spectroscopy

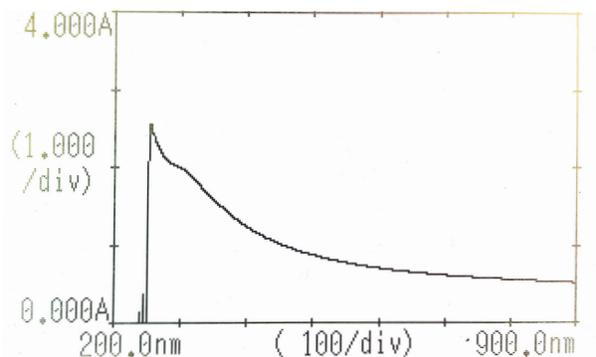


Figure 3.2A

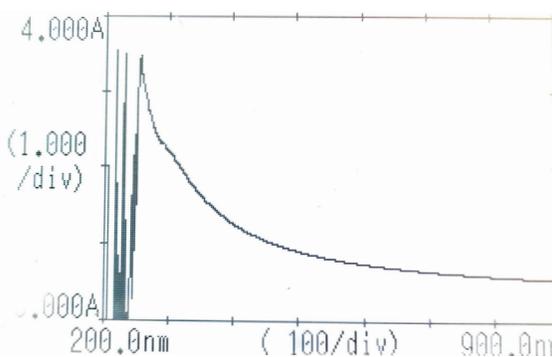


Figure 3.2B

Ultraviolet-Visible Spectroscopy was also carried out on samples of GO (Figure 3.2A) and CGO (Figure 3.2B) after synthesis.

GO showed a strong absorption peak at about 260 nm, attributed to the π - π transition of the C-C bonds. Weak absorption was observed at about 310 nm due to the n - π transition of C=O bonds. Hence, similar to the FTIR spectra, the UV-Vis spectra revealed the presence of many oxygen functional groups, such as hydroxyl and carbonyl on GO (Khalili, 2016).

CGO showed similar absorption peaks, with one at 260 nm and a shoulder at 310 nm, due to the fact that there is no addition or complete removal of any one type of bond detectable by UV-Visible Spectroscopy during the carbonisation process.

3.3. X-Ray Diffraction Analysis

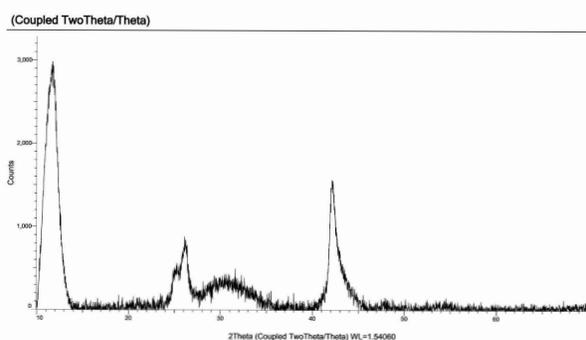


Figure 3.3A

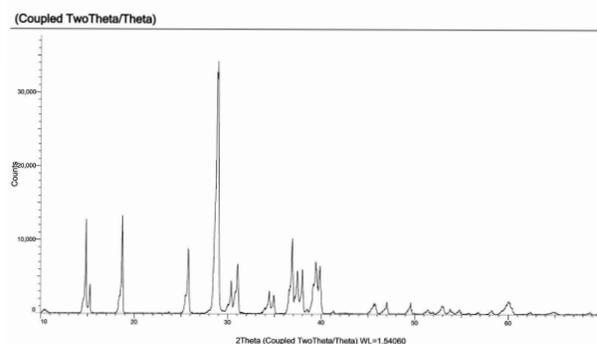


Figure 3.3B

X-Ray Diffraction analysis (XRD) was carried out on samples of GO (Figure 3.3A) and CGO (Figure 3.3B) after synthesis.

Sharp diffraction peaks at $2\theta = 11.7^\circ$ and $2\theta = 42.1^\circ$, which are similar to those of Ain *et al.*, 2019, were observed with GO, indicating its successful synthesis. The 11.7° peak also shows complete oxidation of graphite to GO due to the introduction of oxygen-containing functional groups (Li *et al.*, 2014).

CGO showed a minor peak at $2\theta = 10.3^\circ$, in alignment with Li *et al.*, 2014. The peak was smaller than that of GO, showing that CGO has larger spacing than GO.

3.4. Heavy Metal Ion Adsorption Tests

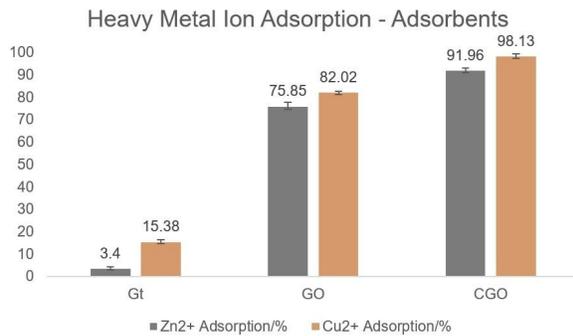


Figure 3.4A

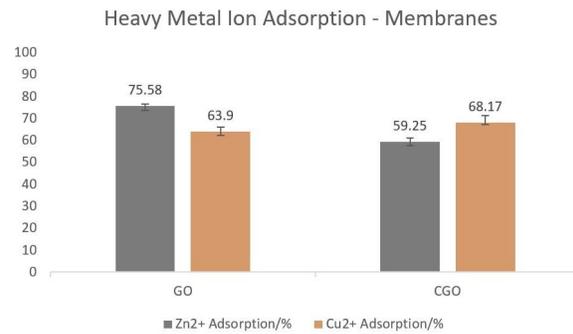


Figure 3.4B

GO had proven to have significantly superior heavy metal ion adsorption capability than pure Gt, at 82.02% compared to 15.38% for Cu²⁺ ions, and 75.85% compared to 3.40% for Zn²⁺ ions. This is because GO benefits from an ordered, structured, highly specific surface area, and flexible surface functionalization options, which help achieve high performance in heavy metal ion removal (Velusamy *et al.*, 2021). CGO possesses a greater heavy metal ion adsorption capacity than GO, due to the presence of more carboxylic acid groups, which enables stronger binding with the heavy metal ions (Bala *et al.*, 2007).

The performance of GO and CGO when used as membranes is poorer than that of its original form, and this can be attributed to the shorter contact time between the Zn²⁺/Cu²⁺ solution and the GO/CGO.

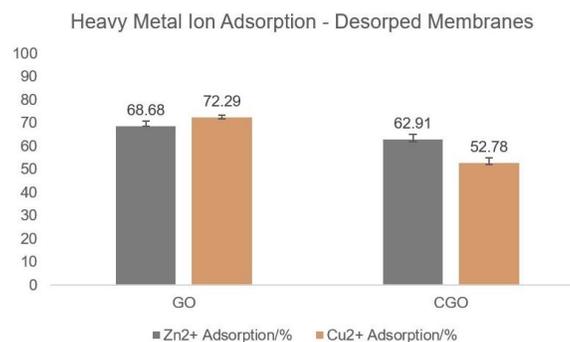


Figure 3.4C

After desorption, GO and CGO membranes have heavy metal adsorption properties in a similar range as compared to their virgin counterparts.

3.5. Organic Dye Adsorption Tests

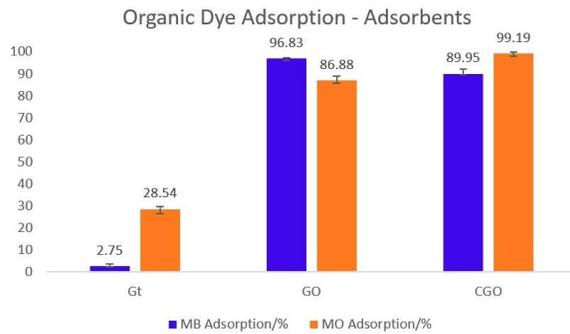


Figure 3.5A

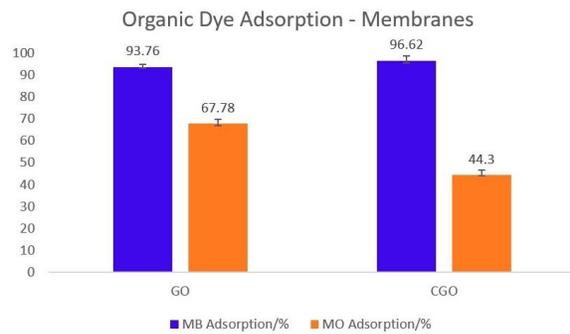


Figure 3.5B

As adsorbents, GO had significantly better adsorption of Methylene Blue (MB) and Methyl Orange (MO) than Gt. Between GO and CGO, GO outperformed CGO in MB adsorption (96.83% compared to 89.95%), while CGO outperformed GO in MO adsorption (99.19% compared to 86.88%).

The adsorption capabilities of GO and CGO as membranes is similar for MB adsorption, but poorer for MO adsorption. The similar, if not better adsorption of MB for GO and CGO membranes can be partially attributed to the fact that cellulose present in the filter paper used for the membranes can adsorb MB, increasing the overall adsorption capability. The decreased capability for MO adsorption for both types of membranes can once again be attributed to shorter contact time.

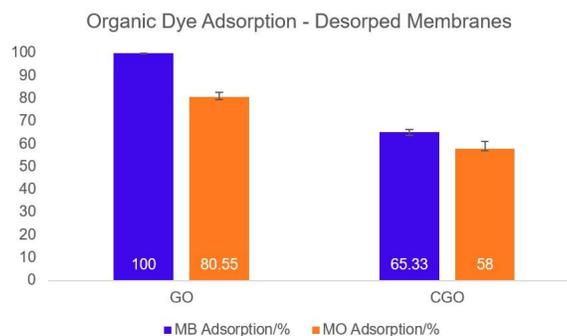


Figure 3.5C

GO membranes displayed improved performance after desorption, while that of CGO membranes was diminished.

3.6. Antibacterial Tests

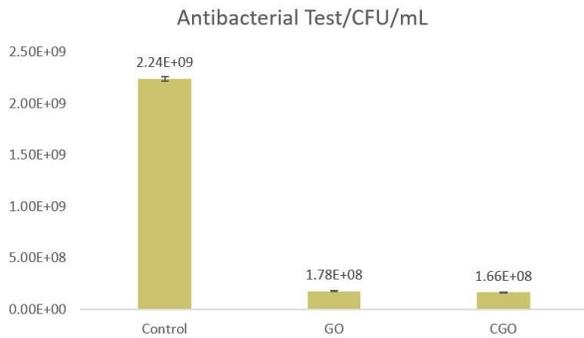


Figure 3.6A

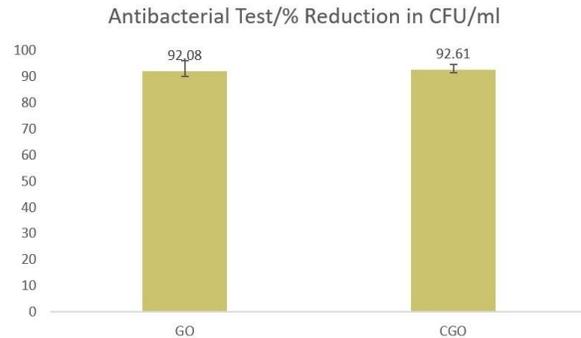


Figure 3.6B

GO is known to possess antibacterial properties, including the inhibition of *E. coli* (Liu *et al.*, 2011). When tested and compared against CGO, GO possesses a strong antibacterial ability, whereas modification to CGO does not decrease its antibacterial capabilities whatsoever.

3.7. Data Analysis

The Kruskal-Wallis Test was carried out to determine the significant differences in heavy metal ion adsorption and organic dye adsorption capabilities when comparing Gt, GO (adsorbent) and CGO (adsorbent) simultaneously, as well as antibacterial properties when comparing control, GO and CGO simultaneously. As can be seen from the results (Figure 3.5A), the heavy metal ion adsorption and organic dye adsorption capabilities of Gt, GO and CGO are significantly different ($p < 0.05$). The bacterial growth when comparing control, GO and CGO is also significantly different ($p < 0.05$).

Test	p-value
Zn ²⁺ Adsorption	0.00193
Cu ²⁺ Adsorption	0.00193
MB Adsorption	0.00193
MO Adsorption	0.00193
Antibacterial	0.00885

Figure 3.7A

The Mann Whitney U test was then carried out to determine significant differences in heavy metal ion adsorption, organic dye adsorption and antibacterial capabilities between GO and CGO (Figure 3.5B). For heavy metal ion and organic dye adsorption, the capabilities of GO and CGO are significantly different ($p < 0.05$) in both adsorbent and membrane form. However, the difference in antibacterial capability is not significant.

Test	p-value for CGO (Adsorbent) compared to GO (Adsorbent)	p-value for CGO (Membrane) compared to GO (Membrane)
Zn ²⁺ Adsorption	0.01208	0.01208
Cu ²⁺ Adsorption	0.01208	0.01208
MB Adsorption	0.01208	0.01208
MO Adsorption	0.01208	0.01208
Antibacterial	0.75656	-

Figure 3.7B

The Mann Whitney U test was also carried out to determine the significant differences in heavy metal ion adsorption and organic dye adsorption between virgin GO/CGO membranes and their desorped counterparts (Figure 3.5C). With $p < 0.05$ considered as significant, the differences in adsorption capabilities between virgin and desorped membranes are significant.

Test	p-value for CGO (Membrane) compared to CGO (Desorped Membrane)	p-value for GO (Membrane) compared to GO (Desorped Membrane)
Zn ²⁺ Adsorption	0.01208	0.01208
Cu ²⁺ Adsorption	0.01208	0.01208
MB Adsorption	0.01208	0.01208
MO Adsorption	0.0164	0.01208

Figure 3.7C

4. Conclusion

From the FTIR spectroscopy tests, GO powder was successfully synthesized from graphite powder, and the modification to CGO with oxalic and hydrobromic acid was also

successful. Based on heavy metal ion sorption tests, GO possesses significant heavy metal ion adsorption capabilities while CGO possesses a greater heavy metal ion adsorption capacity than GO, showing the potential utility for modification of GO. Based on the organic dye sorption tests, GO and CGO possess great organic dye adsorption capabilities. GO was the most capable in MB adsorption, while CGO excelled the most in MO adsorption. Based on the antibacterial tests, both GO and CGO had exceptional capabilities in inhibiting the growth of *E. Coli* bacteria, and both were shown to be nearly identical in their abilities to inhibit the growth of *E. Coli* bacteria. The membrane form of GO and CGO do possess poorer adsorbent capabilities than adsorbent form, but are objectively still strong. Even after desorption, GO and CGO membranes are fairly capable in adsorbing heavy metal ions and organic dyes.

4.1. Future Work

Other forms of modified GO, including reduced GO (rGO), can also be tested and compared to determine the most capable membrane form for heavy metal ion adsorption, organic dye adsorption and antibacterial tests. Heavy metal ion adsorption and organic dye adsorption tests can also be carried out at different pH to determine the relationship between adsorption capacity and pH for the various GO membranes.

5. References

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