

Eco-Friendly Liquid-Phase Synthesis and Characterization of Graphene Oxide Films for Chemical Sensor Applications

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ABSTRACT: The great demand for sustainable material in Nanotechnological industries has sparked the discovery of wonder material known as graphene and its derivatives (graphene oxide, reduced graphene oxide) because of its unique properties ranging from structural, electrical and optical properties. However, the conventional method has left chemical and high temperatures that are hazardous to human health and the environment. This research deals with an ecofriendly method of synthesis and deposition of graphene oxide (GO) using spin coating, achieving thin films with uniform structural, optical, and electrical properties. The Scanning Electron Microscope analysis at a magnification of 1 μ m, revealed a wrinkled morphology typical of GO, while Raman spectroscopy unveiled G band at $\sim 1600\text{ cm}^{-1}$ and D band at $\sim 1350\text{ cm}^{-1}$ which confirmed the presence of graphitic domains and defects respectively. UV-V spectroscopy analysis reveals the absorption peak at 232 nm while the shoulder appears around 294 nm which verifies the successful oxidation and exfoliation of graphite into GO, with well-defined optical characteristics. Hence, the linear I-V curve indicated consistent electrical behavior across the film. These findings demonstrate the potential of first eco-friendly GO films for sensors via spin coating for applications in advanced materials and nanotechnology.

KEYWORDS: Eco-friendly Synthesis, Graphene Oxide Film, Green Liquid-phase

I. INTRODUCTION

The great demand for sustainable technology has sparked the discovery of novel materials and the modification of existing ones for application in a variety of spheres of human endeavour, particularly nanotechnology. One of the novel materials discovered in 2004 by Manchester University academics, Novoselov and Geim, were awarded the 2010 Nobel Prize in Physics for their pioneering work on ground-breaking research on graphene, Dresselhaus et al., 2010. To address these challenges, this research has focused on developing green synthesis methods that minimize environmental impact while maintaining the desired properties of graphene oxide. One such approach is the liquid-phase exfoliation of graphite using environmentally friendly solvents such as acetone and deionized water, Avornyo & Chrysikopoulos (2024) and Hernandez et al. (2011). This method provides an efficient and scalable route for producing high-quality GO with controlled structural properties. Spin coating is a widely used technique for depositing thin films of GO onto substrates such as glass slides, ensuring uniform distribution and optimal functional properties for sensor applications (Kim et al., 2010). The characterization of these films is crucial for evaluating their structural integrity and performance. Techniques such as scanning electron microscopy (SEM), Raman spectrum, four-point probe measurements, and UV-visible spectroscopy provide comprehensive insights into the morphology, crystallinity, electrical conductivity, and optical properties of the synthesized GO films respectively (Zhou et al., 2011). On the sonification method it has proven to yield better mechanical and conductivity properties. Sonification slows the oxidation of graphite by impeding the intercalation and reduces the functionalization the graphene basal plane. This study aims to explore the eco-friendly synthesis and characterization of graphene oxide thin films for sensor applications. By optimizing the synthesis process and systematically analyzing the material properties, this research contributes to the growing body of knowledge on sustainable nanomaterial production and its practical applications.

II. RELATED WORKS

Graphene, a single layer of graphite with a 2D honeycomb-like lattice (sp^2 hybridization), possesses high-demanding properties like mechanical, electrical, thermal, and optical properties that are critical for nanotechnological industries (Worku & Ayele, 2023). It is a semiconductor with a zero-band gap at the Dirac point, exhibiting very high electrical conductivity. The electronic states of graphene can be well described by the tight-binding Hamiltonian of π electrons in carbon atoms (Rajib et al., 2022). Its application potential spans from environmental remediation (Abbas et al., 2022) to composites (Pei & Cheng, 2012). Since its first isolation via mechanical exfoliation in 2004 (Mbayachi et al., 2021; Singh et al., 2011), it has been recognized as a carbon allotrope with a planar sheet of atoms arranged in hexagonal rings (Veera, 2019; Katsnelson, 2007). The development of sustainable and eco-friendly materials has gained significant attention in recent years, particularly in the field of nanotechnology. Graphene oxide (GO), a derivative of graphene, is an advanced two-dimensional material known for its exceptional electrical, thermal, and mechanical properties. GO has a similar hexagonal carbon structure to graphene but is decorated with hydroxyl ($-OH$), alkoxy ($C-O-C$), carbonyl ($C=O$), carboxylic acid ($-COOH$) and other oxygen-based functional groups (Pendolino & Armata, 2017). These functional groups are not merely structural defects; they are active sites that facilitate further chemical modification and enhance dispersibility in various solvents, which is crucial for processing and composite formation (Smith et al., 2021). One of the vital strengths of the functional groups present in graphene oxide is that their variation can tune the material's electrical, mechanical, thermal, and physical properties (Sharma et al., 2021).

GO holds great promise for a variety of applications, including sensors, energy storage devices, and electronic components (Geim & Novoselov, 2007). However, traditional synthesis methods, such as the Hummers' method and its modifications, often involve hazardous chemicals like concentrated sulfuric acid (H_2SO_4), potassium permanganate ($KMnO_4$), and hydrogen peroxide (H_2O_2), raising serious environmental and safety concerns (Chen et al., 2022; Dreyer et al., 2010). The environmental footprint of these conventional routes has catalyzed a paradigm shift towards more sustainable production techniques (Yadav & Kumar, 2023). In response, the field of green chemistry has been actively applied to graphene synthesis. A prominent strategy involves using renewable biomass as a precursor. For instance, Chua et al. (2022) demonstrated the synthesis of graphene oxide from sugarcane bagasse, an agricultural waste product, highlighting a circular economy approach that reduces reliance on purified graphite. Similarly, Lee & Park (2024) reviewed various plant-based extracts (e.g., from leaves and fruits) that can serve as reducing and stabilizing agents for producing GO and reduced GO, minimizing the need for toxic hydrazine. These bio-inspired methods not only mitigate environmental impact but also often result in GO with unique morphological properties suitable for biomedical applications. The push for sustainability extends beyond synthesis to the material's entire life cycle. A recent life-cycle assessment (LCA) study by Fernández et al. (2023) compared the traditional Hummers' method with a green electrochemical exfoliation process. Their findings confirmed that the green route significantly reduces energy consumption, carbon emissions, and toxic waste generation, providing a quantitative argument for its adoption. Furthermore, the functional groups on GO can be leveraged for environmental good, such as in water purification. A study by Gupta et al. (2023) developed a GO-based membrane functionalized with specific ions for the highly efficient and selective removal of heavy metals from industrial wastewater, directly addressing the pollution concerns associated with its own traditional production.

III. RESULTS AND DISCUSSION

A. Study Location and Data Collection

Meteorological data for this study, including temperature, humidity, and solar irradiance, were sourced from the headquarters of the Nigerian Meteorological Agency (NiMet) in Abuja. The city features a tropical wet and dry climate and is situated at an average elevation of approximately 455.89 meters above sea level (Nigerian Meteorological Agency [NiMet], n.d.). Its geographic coordinates are $9.07^\circ N$ latitude and $7.60^\circ E$ longitude, encompassing an area of 1,769 square kilometers. The topography immediately surrounding the city (within a 3.2 km radius) is characterized by relatively modest elevation changes. The maximum elevation shift in this area is 143 meters, with an average altitude of 494.7 meters above sea level (NiMet, n.d.). In terms of land cover, the city is predominantly composed of artificial surfaces (77%) and cropland (22%). On a broader scale, the landscapes within

16.1 km and 80.5 km of the city center are largely a mix of cropland and shrubs, accounting for 37% and 43% of the area, respectively.

Abuja experiences annual temperatures that typically fluctuate between a low of 16.6°C and a high of 33.9°C (NiMet, n.d.). The hottest period lasts roughly 2.5 months, during which average daily high temperatures exceed 32.2°C. A cooler season spans approximately 3.5 months, where average highs remain below 28.3°C. December is the coldest month, with average lows of 16.1°C and highs of 30.6°C.

The city's clearest weather begins around November 7th and lasts for about 3.6 months, with January standing out as the clearest month. In contrast, a cloudier period starts around February 24th and persists for about 8.4 months, reaching its peak in May. The wet season extends for 6.2 months and carries a greater than 42% probability of precipitation on any given day. This is counterbalanced by a 5.8-month dry season; December has the fewest number of rainy days. August is the wettest month, while December is the driest. Daylight hours in Abuja remain relatively consistent year-round, averaging about 12 hours with only minor variation. Conversely, humidity levels shift significantly with the seasons. A muggy and often uncomfortable period lasts for about nine months, from late February to late November. During this time, conditions are humid or oppressive for at least 27% of the time. August is the most humid month, with nearly all 31 days classified as muggy or worse (NiMet, n.d.).

B. MATERIALS

The following materials were used in this study:

- (a). Preparation of Solvent Mixture: acetone and DI water were mixed in a 7:3 ratio (acetone: water), that is 70% acetone and 30% DI water. This mixture can effectively disperse graphite while balancing exfoliation and stability. The solvent mixture was stirred thoroughly to ensure a homogeneous solution.
- (b). Graphite Dispersion: The graphite powder was added to the acetone/DI water mixture at a concentration of around 5 mg/mL. Hence, the mixture was stirred with a magnetic stirrer for 15 minutes to pre-disperse the graphite particles.
- (c). Sonication Process: The mixture was transferred to a suitable container that fits the ultrasonicator. The solution was then ultrasonicated in an ultrasonic bath for 2 hours, for moderate yield and good quality of graphene. It was ensured that the temperature of the mixture did not exceed 40°C during sonication using an ice bath, as higher temperatures can degrade the graphene quality.
- (d). Centrifugation: After sonication, the mixture was centrifuged at 3000 RPM for about 40 minutes. This was done to separate larger graphite particles and unexfoliated flakes from the exfoliated graphene. The above condition was to divide the mixture into two (2) layers: supernatant and sediment. The supernatant (liquid part) will contain the exfoliated graphene, while the sediment will contain non-exfoliated graphite.
- (e). Collection of Graphene Oxide: To collect the graphene oxide, the supernatant was carefully decanted into a clean container without disturbing the sediment. This supernatant contains the few-layer or single-layer graphene oxide.
- (f). Washing and Purification: The deionized water was gradually added to the supernatant to remove any residual acetone or impurities.
- (g). Cleaning the Glass Slide: The glass slides were washed thoroughly with soap and water to remove any grease or dirt that may alter the quality of the graphene. Hence, the slides residue rinsed with acetone to remove organic residue contents. Subsequently, it was rinsed with deionized (DI) water to ensure all cleaning agents were removed. Finally, the glass slide was dried by placing it on a hot plate at a bit temperature (~60°C) to remove any remaining moisture.
- (h). Spin-Coating: The cleaned glass slide was secured onto the spin-coater stage using a vacuum and a pipette was used to deposit a small droplet (approximately 200 µL) of the graphene dispersion onto the center of the glass slide. However, the spin-coater was started at a low speed (600 RPM) for 15 seconds to spread the liquid evenly. It was further increased the spin speed to a higher value (4000 RPM) and maintained this speed for about 50 seconds. The high-speed rotation helped to create a uniform thin film as the solvent evaporated rapidly.

- (i). Annealing the film: After spin-coating, the glass slide was placed on a hot plate set to around 70°C for 7 minutes to ensure complete evaporation of the solvent and good film adhesion on the substrate (glass slides). Figure 1 demonstrate the schematic flow of the deposition process of the work.

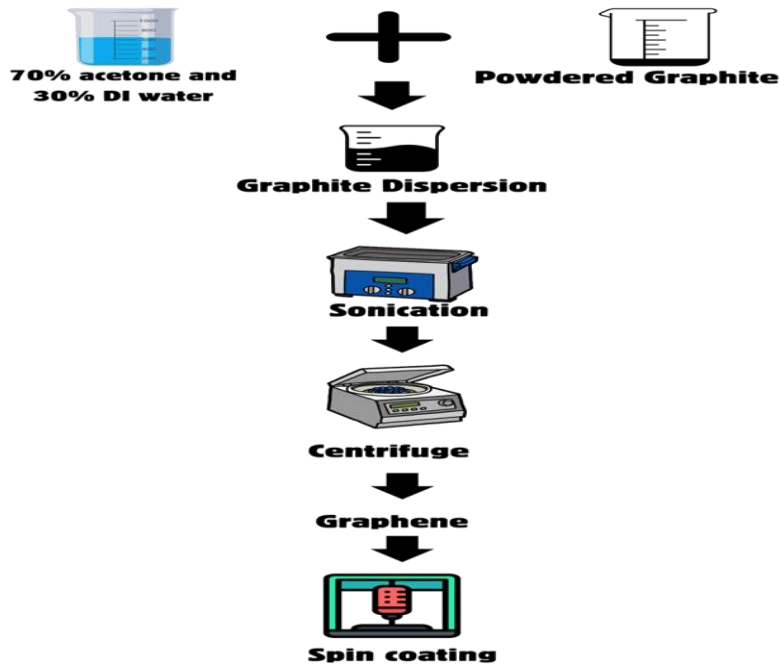


Figure 1: Schematic flow chart of the synthesis and deposition process

IV. RESULTS AND DISCUSSION

At a magnification of 1 μm , plate 1 shows the SEM image provides a detailed view of the graphene oxide (GO) film structure, showcasing its microstructural arrangement at a fine scale. The image reveals a characteristic layered and wrinkled morphology, which is a well-known feature of graphene oxide films. These wrinkles are attributed to the drying and settling processes during spin coating, as well as the intrinsic structure of GO sheets, as previously reported in the literature (Park et al., 2009). Additionally, the continuity of the GO layer, as observed in the image, indicates a well-controlled spin-coating process, which is crucial for achieving uniform film deposition (Li et al., 2011). This morphology is particularly advantageous for applications in sensors, transparent conductive films, and electronic devices, where the unique properties of GO can be effectively utilized (Kim et al., 2010).

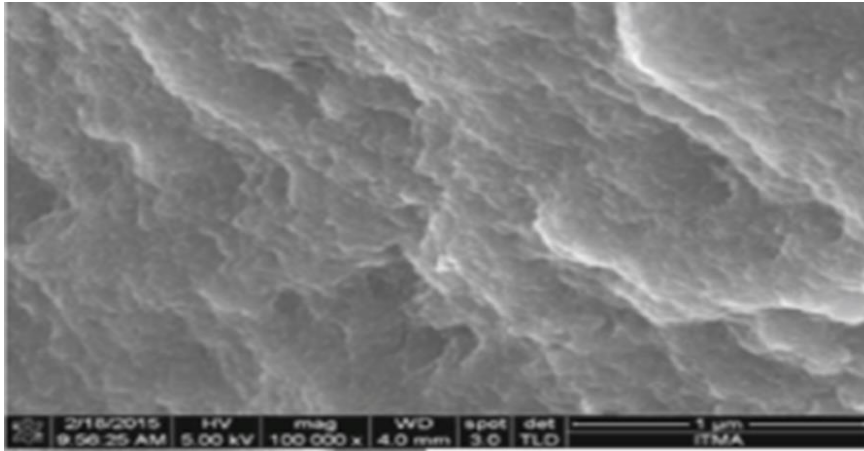


Plate 1: SEM images of graphene Oxide film on a glass slide

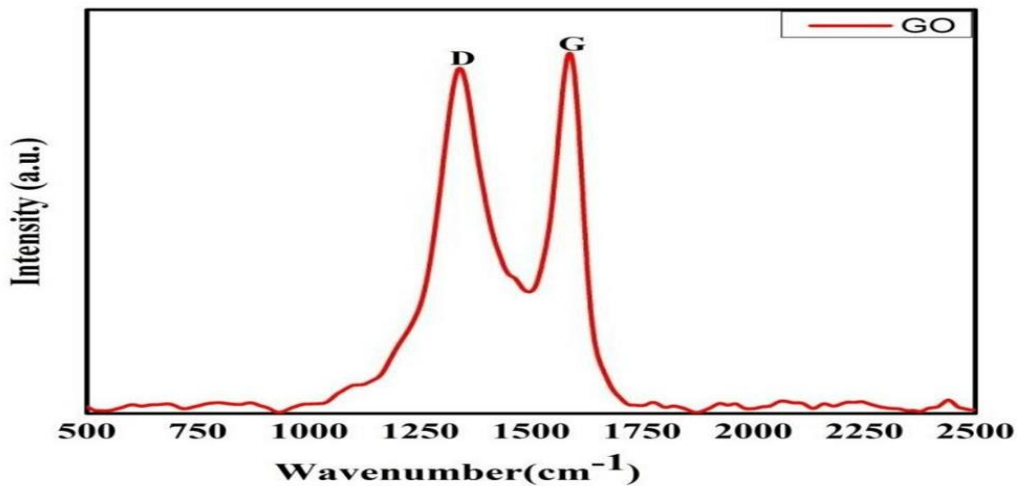


Figure 2: Raman Spectrum for Graphene Oxide deposited on a Glass Slide

Raman spectrum for the deposited film as shown in Figure 2 shows the D band at $\sim 1350 \text{ cm}^{-1}$ is attributed to the presence of defects and disorder in the carbon lattice. These defects arise from structural imperfections, oxygen-containing functional groups introduced during the oxidation process, and edge effects in graphene oxide (Ferrari et al., 2015). The G band at $\sim 1600 \text{ cm}^{-1}$ corresponds to the in-plane vibrations of sp^2 -hybridized carbon atoms within the graphene lattice. This peak is indicative of graphitic domains and is observed in both pristine graphene and graphene oxide (Dresselhaus et al., 2010). The distinct D and G peaks in the spectrum indicate a uniform deposition of graphene oxide on the glass substrate. The well-defined nature of these bands confirms that the spin-coating process has effectively distributed the GO layers across the surface (Zhou et al., 2011).

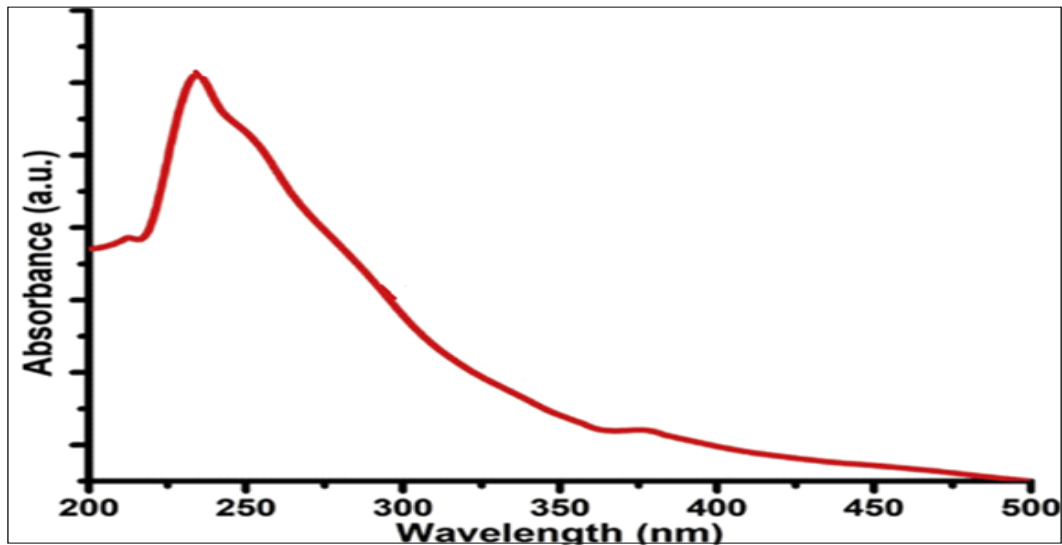


Figure 3: UV-Vis Spectroscopy for Graphene Oxide deposited on Glass Slide

The prominent absorption peak observed at 232 nm in the UV-Vis spectrum corresponds to the π - π^* transitions of aromatic C=C bonds in graphene oxide (GO) as displayed Figure 3. This peak is a characteristic feature of the conjugated electronic structure within the graphitic domains of GO, reflecting the retention of sp^2 hybridized carbon atoms in the material's framework (Zhang et al., 2010). The shoulder appearing around 294 nm is attributed to n - π^* transitions involving carbonyl (C=O) or other oxygen-containing functional groups. These functional groups are introduced during the oxidation process used to convert graphite into graphene oxide. Their presence disrupts the extended π -conjugation of the graphitic structure, introducing localized defect states within the material (Dreyer et al., 2010). The distinct and well-defined peaks in the spectrum indicate that the spin-coating process successfully produced a uniform and consistent deposition of graphene oxide on the glass substrate. This outcome underscores the effectiveness of spin coating in creating thin films with homogenous optical and structural properties suitable for further applications (Hassanzadeh et al., 2014).

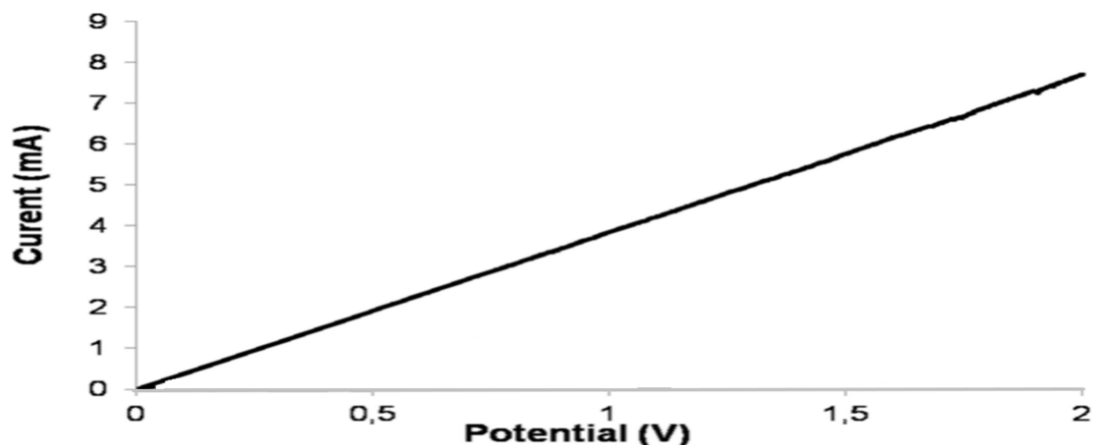


Figure 4: I-V Curve showing Sheet Resistance for Graphene Oxide deposited on a Glass Slide

Figure 4 shows the I-V curve exhibits a linear relationship between current (mA) and potential (V), indicative of ohmic behaviour in graphene oxide (GO) thin films deposited on a glass slide. Such behaviour is characteristic of GO due to its insulating nature, which results from oxygen-containing functional groups that disrupt the conductive π -conjugated network within the material (Pei and Cheng, 2012). The linearity and smoothness of the I-V curve suggest that the spin-coating process effectively produced a uniform and defect-free GO film. Uniform deposition

is essential for ensuring consistent electrical properties across the surface, which is crucial for applications requiring reliable conductivity and resistance (Dubey et al., 2017).

V. CONCLUSION

The work optimized the synthesis and deposition of graphene oxide using spin coating, achieving thin films with uniform structural, optical, and electrical properties. The SEM analysis revealed a wrinkled morphology typical of GO, while Raman spectroscopy confirmed the presence of graphitic domains and defects. The UV-Vis spectroscopy further verified the successful oxidation and exfoliation of graphite into GO, with well-defined optical characteristics. The linear I-V curve indicated consistent electrical behaviour across the film. These findings demonstrate the potential of spin-coated GO films for applications in sensors due to the presence of its functional group like epoxides hydroxyls and oxygen, hence can be a good gas sensor and its stability is still an open-ended research gap to be explored by physicists, chemists and biologists. As the new world revolves around climate and eco-friendly materials graphene nano-sensors have a futuristic application in industrial production as well as environmental remediation due to their benefits in absorption, sensitivity and selectivity.

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