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# STUDY THE EFFECT OF VARIATION IN VOLTAGE AND ELECTROLYTIC SOLUTION MEDIUM IN THE ELECTROCHEMICAL EXFOLIATION OF GRAPHITE TO GRAPHENE OXIDE

MD. KHORSHED ALAM[1]*, MOHAMMAD AMIRUL HOQUE[2], SABBIR HOSSAIN NAEEM[1], MD. ASADUZZAMAN[1] AND MD. AL-AMIN[1]

*[1]Department of Physics, University of Barishal, Kornokathi, Barishal-8254, Bangladesh*
*[2]Bangladesh Council of Science and Industrial Research (BCSIR), Dhanmondi, Dhaka-1205, Bangladesh*

**Corresponding author e-mail: dmkalam@bu.ac.bd*

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## ABSTRACT

Graphene oxide (GO) and its reduced form have recently attracted significant interest due to their outstanding physical and chemical characteristics, hence finding large applications in fields such as electronics and optoelectronics. The paper portrayed herein presents a simple and cost-effective route to obtaining high-quality GO through electrochemical exfoliation methods. The pre-treated Graphite powder was soaked in concentrated sulfuric acid and nitric acid in a 1:1 ratio, followed by washing and neutralization. Thereafter, it was made into paper with the aid of 1% polyethylene. The sheets were suspended in electrolyte solutions of KCl and NaCl at 10.0V, 12.5V, and 15.0V to make sure the swelling of the sheets occurred. Boiling and centrifugation are done for exfoliation and purification. Characterization of GO was performed using FT-IR, FT-Raman, and UV-vis spectroscopy. It will be verified that the voltage and electrolyte type will tune the properties and characteristics of as-prepared GO effectively. The process results in the scale-up and environmentally friendly process for GO production, making the product efficient for applications in so many high-tech industries.

**Keywords:** *Electrolyte influence, Graphene Oxide (GO), Electrochemical exfoliation, Reduced Graphene Oxide (RGO), Voltage effects*

## 1. INTRODUCTION

Graphene Oxide (GO) and Reduced Graphene Oxide (RGO) continue to be at the forefront of materials science research due to their remarkable physicochemical properties and broad potential applications, particularly in electronics, optoelectronics, and advanced materials. These materials have extraordinary electrical, thermal, and mechanical properties and are therefore generally used for sensors, energy storage devices, and biomedical applications. Issues of concern from an environmental and safety point of view are traditional methods of synthesis, including Hummers' method and its derivatives, through the use of harsh chemicals with complicated procedures. Recent development tries to find more sustainable and effective routes of synthesis. Among the most relevant, especially for its simplicity, cost-effective nature, and reduced environmental impact, is represented by electrochemical exfoliation. The method would exfoliate Graphite into Graphene layers in an electric field together with an electrolyte. In this case, the major parameters involved in such a process are generally the applied voltage and type of electrolytic solution, which has much effect on the quality and yield of the produced GO.

Lots of research effort has been intensified to enhance the understanding of GO synthetic methods and their practical applicability. Dreyer et al. gave a good discussion on the chemical properties and potential uses of GO, urging functionalization to enhance its properties. In 2014, Paton et al. showed a scalable liquid-phase exfoliation technique to produce defect-free layers of Graphene by controlling the exfoliation conditions. Marcano et al. (2010) improved the synthesis process, making GO much better in quality than that synthesized with conventional methods [13]. Pei and Cheng (2012) worked to reduce GO to make it perform better in electronic applications by improving its electrical conductivity [14]. In the process of characterization, Ferrari et al. and Cançado et al. undertook the measurement of defects in Graphene and GO using Raman spectroscopy as an important step in material property tailoring for specific application purposes. Other works showed how thin-film electronics and optoelectronic devices prepared from chemically derived GO. There are also other works on improvement in its properties toward various applications. Zhu et al. [18] have reported on the thermal reduction method to improve GO for supercapacitors. Compton et al. [19] have discussed its electrochemical activity, suggesting a direction for the use in sensors and biosensors. Stankovich et al. [20] characterized GO as filler material in polymer composites that led to the observed enhanced mechanical properties. It has been shown in numerous ways to upgrade energy storage in most applications, primarily within Li-ion batteries towards improved energy density and cycle life. Shao et al. (2015) elaborately developed Graphene-based composites for supercapacitors, showing the functionality of GO in energy storage applications. Wu et al. (2012) showed photocatalytic properties of GO and suggested its applications in environmental fields such as water purification [23], whereas Tang et al. have demonstrated its applications in flexible and wearable electronics [24].

Recent studies in improving the sustainability, scalability, and performance of GO production were conducted by Guo et al. (2021) to increase both the yield and quality of GO in an electrochemical exfoliation technique, while reducing the environmental burden of the process by replacing the organic electrolytes commonly used with water-based ones [25]. Liu et al. (2022) reported a scaled-up synthesis of GO by the modified Hummers method combined with electrochemical exfoliation, resulting in electronic-property-enhanced materials [26]. Wang et al. (2023) focused on the optimization of electrochemical parameters to increase the mechanical strength of GO—another crucial need when considering its use in flexible and wearable devices [27]. Apart from this, new applications of GO have also been explored. Sharma et al., 2021, gave a new direction in the application of GO toward the next generation of supercapacitors with better improvement for energy storage performance. Zhou et al., 2022, did research on GO for biomedical purposes, especially on drug delivery systems, by making most of its biocompatibility and high surface area. Li et al., 2023, have demonstrated that functionalized GO can be well used for cleaning up water, specifically in clearing heavy metals and organics from a polluting environment.

Such advances, however, do not include detailed studies that would optimize the parameters of electrochemical exfoliation, especially variable voltage and different electrolytic solutions. It should also be necessary to systematically establish the optimum conditions to gain quality GO in maximized yields and modified properties. With most such steps having already led to significant progress in preparing GO, very little attention has been paid to fine-tuning the electrochemical exfoliation parameters. In particular, there has been a lack of studies into the effects of applied voltage and types of electrolytes on the quality and yield of GO. This study systematically examines the effects of varying voltage and different electrolytic solutions during the electrochemical exfoliation of Graphite into GO. Our objective lays in finding a cost-effective and efficient process to produce good-quality GO with controllable properties, having increased yield. The derived results from this research may improve sustainable industrial scale production of GO for broad applications within different high-tech sectors.

## 2. METHODS AND MATERIALS

### 2.1 Materials

The materials used in this work include PVA, distilled water, concentrated $H_2SO_4$, concentrated $HNO_3$, KCl, NaCl, and Graphite powder. All the chemicals used in this research work were of analytical grade and hence of a high degree of purity that would normally be suitable for accurate scientific work. They were used directly in the experiments without further purification, showing their quality was good enough for research purposes.

### 2.2 Pretreatment of Graphite Powder

The powder had been pretreated to eliminate impurities; thus, this Graphite powder was first cleaned with concentrated sulfuric acid, $H_2SO_4$, and concentrated nitric acid, $HNO_3$, in a 1:1 ratio. This mixture of acids was boiled at about 80°C just to enhance effectiveness in the removal of impurities. The temperature was stirred well with the Graphite and the acid solution for about 30 minutes to give enough time for the action of acids in reacting and dissolving any impurities that may be present in the Graphite. After this, the so-treated Graphite was now ready and prepared for subsequent uses in this research. The Graphite powder, after this treatment, was washed with distilled water until the pH of the wash was about 7.0 Figure 1a illustrates this.

**Fig. 1.** Preparation and molding process of GO. (a) Pretreating Graphite powder with a mixture of Sulfuric acid and Nitric acid, and (b) Forming Graphite sheets using PVA as a binder.

### 2.3 Molding of Graphite Sheets

For the binder, 1% polyvinyl alcohol (PVA) to give moldability was used on the Graphite powder. The pretreated Graphite powder was mixed with the PVA solution and molded by using a hydraulic press loaded with 60.00 KN applied pressure to get the Graphite sheets; subsequently, the molded sheets were dried at 60°C for 24 hours to have total elimination of moisture content, as presented in Figure 1b [32].

### 2.4 Electrochemical Exfoliation

The dried Graphite sheets were subjected to electrochemical exfoliation in different electrolytic solutions of KCl and NaCl. The exfoliation process was conducted at three fixed voltages: 10.0V, 12.5V, and 15.0V. A two-electrode system was used, where the Graphite sheet used as the anode and a Platinum wire was used as the cathode. Exfoliation was carried out at room temperature for 2 hours, as shown in Figure 2a [33].

**Fig. 2:** Electrochemical exfoliation and purification of GO. (a) Electrochemical exfoliation setup in which the Graphite sheet acts as an anode and the Platinum wire serves as a cathode. (b) The process involves purifying exfoliated GO by boiling, washing, and centrifuging it.

### 2.5 Purification

The exfoliated GO underwent a purification process that began by boiling the suspension in distilled water for 30 minutes. This step ensured that any remaining impurities or contaminants were loosened from the GO structure. After boiling, the suspension was carefully rinsed with distilled water to remove any dissolved contaminants. The next step involved centrifuging the suspension at a speed of 5000 rpm for 15 minutes. This helped to separate the solid GO from the liquid supernatant. The washing and centrifugation steps were repeated multiple times until the pH of the supernatant—indicating the liquid portion after centrifugation—reached a neutral value of approximately 7.0. Reaching this pH level was critical to ensure that all residual acids or unwanted materials had been effectively washed away. Figure 2b illustrates the progression of this purification process. This multi-step procedure was essential to achieve a high level of purity in the exfoliated GO, making it suitable for subsequent research or practical applications.

### 2.6 Characterization

The structural and optical characteristics of the obtained GO were thoroughly analyzed using various advanced spectroscopic techniques. First, Fourier Transform Infrared (FT-IR) spectroscopy was employed to identify the specific functional groups attached to the GO structure. This method provided crucial insights into the chemical composition, revealing the presence of oxygen-containing groups such as hydroxyl, carboxyl, and epoxy groups, which are typically introduced during the oxidation process. These functional groups play a significant role in determining the chemical reactivity and potential applications of GO.

Next, Fourier Transform Raman spectroscopy was utilized to examine the vibrational modes of the GO. This technique allowed for the investigation of the material's structural integrity, particularly the degree of disorder or defects present within the Graphene layers. Raman spectra provide information on the D and G bands, which correspond to defects and the graphitic structure, respectively. By analyzing the intensity ratio of these bands, researchers could assess the quality and level of oxidation of GO.

Finally, Ultraviolet-visible (UV-vis) spectroscopy was conducted to explore the optical properties of GO. This technique is crucial for determining the material's bandgap, which influences its electronic and optical behavior. By studying the absorbance spectrum, researchers could calculate the bandgap energy, which is important for potential applications in optoelectronic devices. Additionally, UV-vis spectroscopy offered insights into the material's overall transparency and light-absorbing properties across the UV and visible light spectrum.

Through the combination of these methods-FT-IR, FT-Raman, and UV-vis spectroscopy-the structural and optical characteristics of GO were comprehensively understood, providing a solid foundation for its potential applications in various fields such as electronics, sensors, and energy storage.

## 3. RESULTS AND DISCUSSION

### 3.1 Effect of Voltage on Electrochemical Exfoliation

The study explored the influence of varying voltage levels-specifically 10.0V, 12.5V, and 15.0V-on the electrochemical exfoliation process used to convert Graphite into GO. The results demonstrated a clear correlation between the applied voltage and the rate of exfoliation, showing that as the voltage increased, so did the exfoliation rate. At the lower end of the voltage spectrum, 10.0V, the exfoliation occurred at a moderate pace. Despite the slower rate, this condition produced a substantial amount of GO characterized by a high level of quality, making it suitable for applications that demand fewer defects and consistent sheet size.

When the voltage was raised to 12.5V, the rate of exfoliation increased significantly, leading to a higher yield of GO. Importantly, even with this higher yield, the quality of GO remained excellent, offering a promising balance between quantity and quality. This suggests that 12.5V might be an optimal voltage for producing high-quality GO efficiently.

However, when the voltage was further increased to 15.0V, the exfoliation process became much faster, but this came at a cost. The higher exfoliation rate led to over-exfoliation, which in turn reduced the quality of the GO produced. The over-exfoliation resulted in smaller GO sheets with an increased number of defects, which could compromise the material's performance in certain applications. This observation was clearly depicted in Figure 3.1, where the relationship between the applied voltage, exfoliation rate, and the quality of the resulting GO was illustrated.

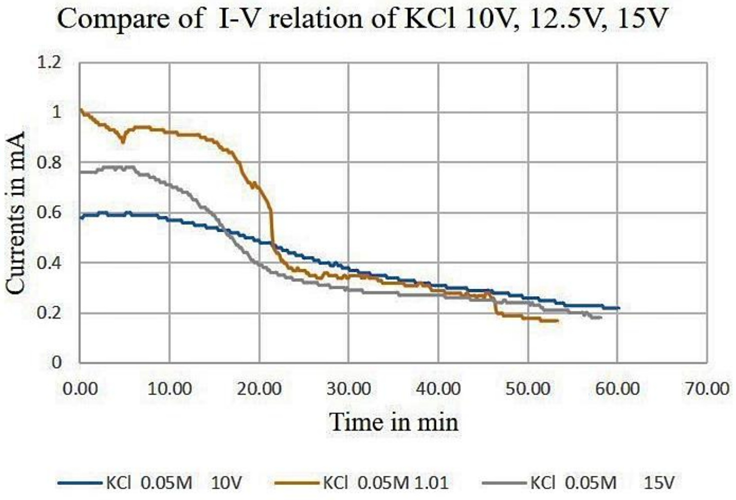

**Fig. 3:** Effect of Voltage on the Exfoliation Rate and Quality of GO.

**Table 1:** Exfoliation yield and GO quality at different voltages and electrolytes

| **Condition** | **10.0V** | | **12.5V** | | **15.0V** | |
|---|---|---|---|---|---|---|
| | **KCl** | **NaCl** | **KCl** | **NaCl** | **KCl** | **NaCl** |
| **Exfoliation Rate** | Moderate | Moderate | High | High | Very High | Very High |
| **GO Quality** | High | Moderate | Very High | Moderate | Moderate | Low |

### 3.2 Effect of Electrolytic Solution on Electrochemical Exfoliation

The study also explored the effect of different electrolytic solutions (KCl and NaCl) on the electrochemical exfoliation process. The type of electrolyte used significantly influenced the exfoliation rate and the quality of GO produced. KCl was more effective than NaCl in producing higher quality GO with fewer defects (Fig. 4). The difference in performance is attributed to the varying ionic strengths and mobilities of $K^+$ and $Na^+$ ions, which affect the efficiency of the exfoliation process. KCl, with its higher ionic strength, facilitated the ion intercalation and exfoliation process much better and ultimately yielded good-quality GO. NaCl, on the other hand, is of a lower ionic strength than KCl; thus, it was relatively less effective, leading to a slow exfoliation process and defective GO.

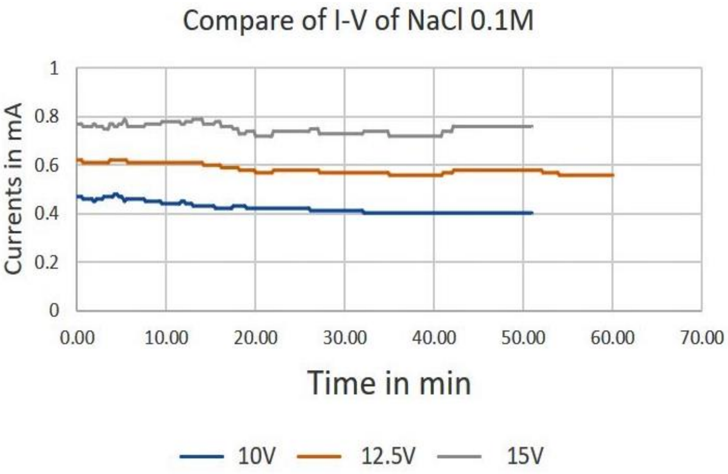

**Fig. 4**: Effect of Electrolyte Type on the Quality and Yield of GO.

### 3.3 Characterization of GO

GO samples produced under different experimental conditions were carefully analyzed using a range of spectroscopic techniques. FT-IR (Fourier-transform infrared spectroscopy) was employed to investigate the functional groups present on GO surface, providing insights into the degree of oxidation and the types of oxygen-containing groups introduced during the exfoliation process. FT-Raman spectroscopy was used to assess the structural integrity of GO, examining the degree of disorder, defects, and overall crystallinity. In addition, UV-vis spectroscopy was utilized to study the optical properties of GO samples, offering valuable information regarding the electronic transitions and the overall absorption characteristics. These complementary techniques provided a comprehensive characterization of GO, helping to assess the impact of the experimental conditions on both the chemical composition and structural quality of the material.

### 3.3.1 FT-IR Spectroscopy:

The FTIR spectra of GO samples as shown in Fig. 5, revealed several characteristic peaks that provide critical insights into the chemical structure and successful modification of the material. Specifically, the peak observed at 1720 $cm^{-1}$ corresponds to C=O stretching vibrations, indicating the presence of carbonyl or carboxyl groups. The peak at 1220 $cm^{-1}$ is associated with C–O stretching, which is characteristic of epoxy or alkoxy groups. Additionally, a broad peak at 3410 $cm^{-1}$ signifies O–H stretching, pointing to the hydroxyl functional groups typically attached to the surface of GO.

These distinct peaks are crucial because they confirm the successful oxidation of Graphite into GO. The emergence of oxygen-containing functional groups, as evidenced by these peaks, is a clear indicator that the material's chemical structure has been altered during the oxidation process. This transformation is essential for the functionalization of Graphene, as these oxygen groups allow for further chemical reactions and applications in various fields.

Furthermore, the quality of GO samples is reflected in the sharpness and definition of these peaks. Higher-quality samples tend to exhibit more pronounced and well-defined peaks, suggesting a more homogeneous and controlled oxidation process. This distinction is important in ensuring consistency in GO production, especially for applications where material properties such as conductivity, surface chemistry, and mechanical strength are critical. Therefore, the FTIR analysis not only validates the oxidation process but also serves as a tool for assessing the quality of the resulting GO.

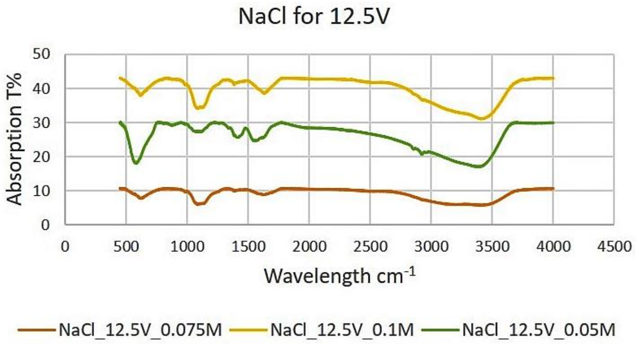

**Fig. 5:** FT-IR Spectra of GO Showing Functional Groups in GO.

### 3.3.2 FT-Raman Spectroscopy

The FT-Raman spectra of GO samples as shown in Fig. 6, exhibit two prominent features known as the D and G bands. These bands are detected at approximately 1350 $\text{cm}^{-1}$ and 1580 $\text{cm}^{-1}$, respectively. The D band, centered around 1350 $\text{cm}^{-1}$, is commonly associated with the presence of defects and structural disorder within the material. This band typically arises due to disruptions in the carbon lattice, which can result from imperfections, edge effects, or even oxygen-containing functional groups in the case of GO. Essentially, the higher the intensity of the D band, the greater the degree of structural irregularities or defects in the sample.

On the other hand, the G band, located near 1580 $\text{cm}^{-1}$, corresponds to the in-plane vibrations of $sp^2$-hybridized carbon atoms. This band represents the ordered regions of the Graphene structure, where carbon atoms are arranged in a hexagonal lattice with minimal disruptions. A strong G band indicates a well-ordered carbon framework, with fewer structural disturbances and more extensive graphitic domains.

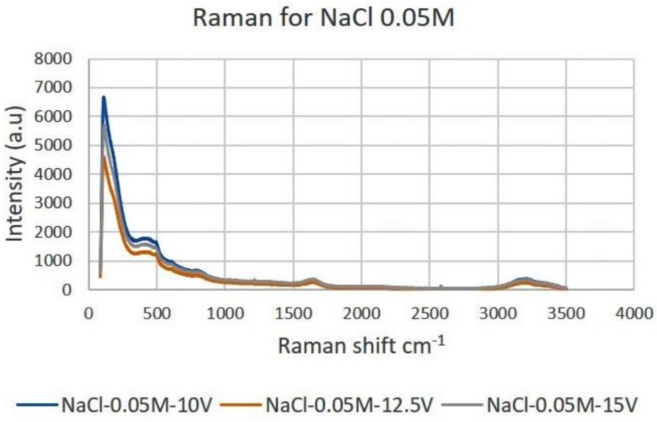

**Fig. 6:** FT-Raman Spectra of GO Highlighting D and G Bands to Show Defect Density.

One of the key parameters used to evaluate the structural integrity of GO is the ratio of the intensities of these two bands, expressed as ID/IG. This ratio provides a quantitative measure of the defect density and the degree of disorder within the sample. A higher ID/IG ratio indicates a higher concentration of defects and greater structural disorder, while a lower ID/IG ratio suggests fewer defects and higher structural quality.

In this particular study, samples prepared under optimal experimental conditions, specifically at 12.5V using KCl as the electrolyte, exhibited a lower ID/IG ratio. This finding implies that these samples had fewer defects and a higher degree of structural integrity compared to those prepared under different conditions. The lower defect density indicates that the preparation method was effective in minimizing imperfections in GO structure, potentially enhancing the material's performance in various applications.

### 3.3.3 UV-vis Spectroscopy:

The UV-vis spectroscopy of GO shown in Fig. 7 provides valuable insights into the material's electronic structure and confirms its formation. In the UV-vis spectra of GO samples, a prominent absorption peak is typically observed at approximately 230 nm. This peak is attributed to the $\pi \rightarrow \pi^*$ electronic transitions of the C=C bonds present in the $sp^2$-hybridized carbon domains of GO structure.

The $\pi \rightarrow \pi^*$ transition refers to the excitation of electrons from a lower-energy $\pi$-bonding molecular orbital to a higher-energy $\pi^*$-antibonding orbital. These transitions are characteristic of conjugated systems like GO, where alternating single and double bonds create regions of electron delocalization. The presence of this absorption peak is a clear indication of the successful formation of GO, as it reflects the specific electronic properties associated with the $sp^2$ carbon structure.

In addition to confirming the material's identity, the absorption peak at 230 nm also offers insights into the electronic properties of GO. Specifically, the position and intensity of this peak provide information about the extent of conjugation and the degree of oxidation in the material. A well-defined, sharp peak suggests a higher degree of conjugation within the $sp^2$ domains, indicating a higher quality GO sample with fewer disruptions in its structure.

The experimental conditions used during the preparation of GO can influence the absorption peak. Variations in synthesis parameters, such as the choice of oxidizing agents, reaction time, and temperature, can affect the degree of oxidation and structural integrity of the resulting GO. As a result, the intensity and position of the absorption peak may shift slightly depending on these conditions.

Higher quality GO samples, which have fewer defects and a more uniform structure, tend to exhibit more defined absorption peaks in the UV-vis spectra. In contrast, samples with a higher degree of oxidation or structural disorder may show broader or less distinct peaks. The variation in peak intensity and sharpness between samples reflects the differences in their electronic structures and, consequently, their overall quality.

This information is critical for characterizing GO, as it allows researchers to assess the material's electronic properties and structural integrity. The UV-vis spectra, along with other characterization techniques, help to determine the suitability of GO for various applications, such as in electronics, sensors, and composite materials.

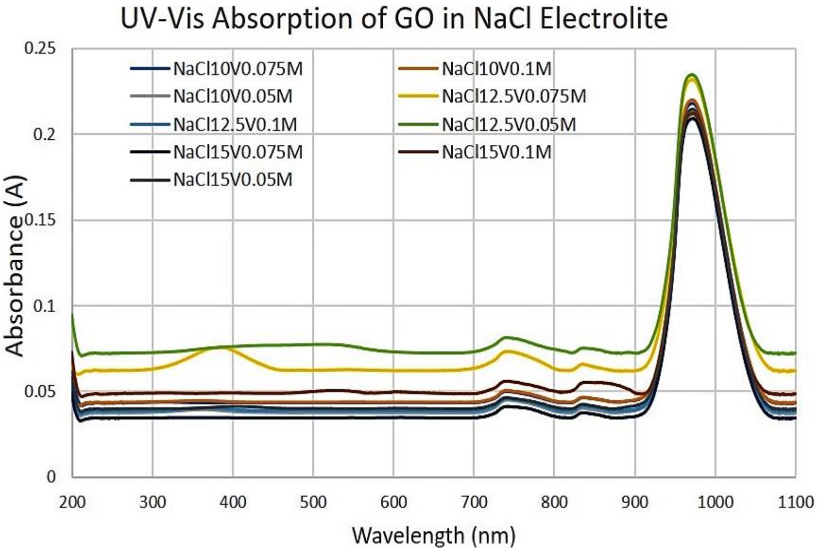

**Fig. 7:** UV-vis spectra of GO displaying the absorption peak characteristic of GO.

### 3.4 Discussion

Results from the present work have shown the importance of applied voltage and type of electrolytic solution to the electrochemical exfoliation of Graphite for the production of GO. It was shown that the effect of applied voltage affects the rate of exfoliation and quality in the produced GO. High voltages, 12.5V, gave a high yield of quality GO. However, voltages too high, such as 15.0V, resulted in over-exfoliation into smaller sizes and sheets of more defective GO.

The nature of the electrolytic solution is another important factor that modifies the exfoliation process. KCl worked better than NaCl to provide high-quality GO with fewer defects. Such effectiveness can be attributed to the fact that more ionic strength of K+ ions, as compared to Na+ ions, increases the mobility and enhances the efficiency of exfoliation.

Characterization by FT-Raman, FT-IR, and UV-vis spectroscopy techniques after synthesizing GO confirmed the presence of functional groups containing oxygen and characteristic peaks, which indicated that the syntheses were successful in nature. The presence of peaks corresponding to C=O, C–O, and O–H groups was identified in the FT-IR spectra, whereas in FT-Raman spectra, the presence of characteristic D and G bands of GO was recorded. In the UV-Vis spectrum, a peak at 230 nm confirmed the formation of GO.

These characterizations provide the necessary insight for the optimization of electrochemical exfoliation parameters by presenting an efficient and cost-effective route toward high-quality GO production. Quality and yield of GO could be controlled by careful tuning of voltage and electrolyte, possibly enabling more sustainable routes toward its industrial-scale production in cases when GO is used in everything from electronics and optoelectronics to advanced materials.

## 4. CONCLUSIONS

This present work demonstrates how quality and yield in GO, produced by electrochemical exfoliation, can be effectively controlled by changing the different applied voltages and types of electrolytic solutions. The best condition was a voltage of 12.5 V in KCl solution, giving very high-quality GO with fewer defects and greater sheet size. Characterization by FT-IR, FT-Raman, and UV-vis spectroscopy confirmed the synthesis of GO containing oxygen functional groups with characteristic peaks.

This approach is appropriate for large-scale and cost-effective production routes toward GO for industrial applications in electronics, optoelectronics, and other advanced materials. These may open up possibilities to optimize the exfoliation process through voltage and varying types of electrolytes, which can lead to greener and more efficient large-scale production of high-quality GO.

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