

Research Article

Synthesis and Characterization of Reduced Graphene Oxide (rGO) Started from Graphene Oxide (GO) Using the Tour Method with Different Parameters

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A new approach to synthesize graphene is oxidizing graphite powder with a mixture of $\text{H}_2\text{SO}_4/\text{H}_3\text{PO}_4$ acids and potassium permanganate. Parameters such as reaction time, reaction temperature, and amount of concentration were varied to study the degree of oxidation of graphite to graphene oxide. Currently, an improved method for the preparation of graphene oxide was the most common one. A mixture of $\text{H}_2\text{SO}_4/\text{H}_3\text{PO}_4$ (9 : 1 volume ratio) instead of only H_2SO_4 resulted in increased hydrophilic and oxidized GO without the emission of toxic gas, which differs from the traditional Hummers' method. The graphene oxide (GO) was converted to reduced graphene oxide (rGO) by chemical reduction using ascorbic acid as the reducing agent. The GO and rGO were characterized by UV-visible spectroscopy, FTIR spectroscopy, and X-ray diffraction patterns. The result showed that treating graphite powder with potassium permanganate (1 : 9) and a mixture of concentrated $\text{H}_2\text{SO}_4/\text{H}_3\text{PO}_4$ acids at 50°C for 12 hours resulted in a better oxidation degree. The designed synthesis strategy could be easily controlled and is an alternative green approach for the production of graphene oxide and reduced graphene oxide.

1. Introduction

Graphene-based materials are playing a great role in the world in every aspect. Among these, graphene is one of the recent miracle materials. Especially, it has many applications in the electronics world since it is the most thinnest, transparent, strongest, and conductive material [1].

Synthesis of graphene oxide (GO) is achieved by placing graphite in concentrated acid in the presence of an oxidizing agent. The Tour method demonstrated a less hazardous and more efficient method for graphite oxidation. This and its modified versions are presently the most commonly used methods for the oxidation of graphite [2, 3].

In the most successful cases, the chemical reduction of GO was conducted using hydrazine or hydrazine hydrate. However, these are highly poisonous and explosive [4] that precautions must be taken when large quantities are used.

Consequently, new approaches for effectively converting GO to reduced graphene oxide (rGO) under mild conditions needed to be explored [5]. L-Ascorbic acid (L-AA), having a mild reductive ability and nontoxic property, is naturally employed as a reducing agent in living things [6] and has also been used as a primary reductant when graphene oxide was converted to reduced graphene oxide [7, 8]. More significantly, in comparison with the conventional reductants used in GO reduction, such as hydrazine and hydrazine hydrate, L-AA itself and the oxidized products are environmentally friendly [9].

Fernandez-Merino et al. revealed that GO reduced by vitamin C (VC) could achieve a C/O ratio of about 12.5 and a conductivity of 77 S/cm , which are comparable to those produced by hydrazine in a parallel experiment. In addition, VC has a great advantage of its nontoxicity in contrast to hydrazine and a higher chemical stability with water than

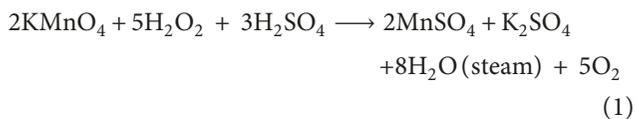
NaBH₄. Furthermore, the reduction in the colloid state does not result in the aggregation of rGO sheets as produced by hydrazine, which is beneficial for further applications [10].

The aim of this study is synthesis of reduced graphene oxide by the Tour method using the cheapest material, that is, graphite, as a starting material using different parameters such as the effect of concentration, temperature, and reaction time in the oxidation level of graphene oxide in a cost-effective way. This was mainly carried out using sulfuric acid, phosphoric acid, and potassium permanganate excluding sodium nitrate that emits toxic gases into the environment. This makes the method environmental friendly. Another most cost-effective and environmental friendly method is the reduction of graphene oxide into rGO (reduced graphene oxide) using ascorbic acid. Even though this method is green, it was not performed with different parameters. In this project, graphene oxide has been produced with different parameters.

Graphene has the potential of replacing the costly and brittle silicon-based electronic devices, but it is in its infant stage. This study is therefore significant for the following: contributes to the study of graphene oxide and reduced graphene oxide, shows how to produce graphene oxide from cheap graphite, shows the effect of parameters on the synthesis of graphene-based material, creates awareness about different applications of graphene-based materials, and provides information for future related studies.

2. Materials and Methods

2.1. Synthesis of GO Using the Tour Method. Graphene oxide (GO) was prepared by mixing 90 mL of concentrated H₂SO₄ (sulfuric acid) and 10 mL of concentrated H₃PO₄ (*ortho*-phosphoric acid) (9:1(v/v) ratio). The purpose of adding phosphoric acid is to prevent further oxidation [11]. The mixture of the acids is poured into a mixture of 0.50 g graphite powder and 4.5 g of KMnO₄ (potassium permanganate), resulting in 40–50°C temperature changes. The mixture was then heated to 50°C using a temperature-controlled water bath and stirred for 12 hours. As the reaction extended, the mixture turned out to be paste. After 12 hours, the content was cooled to room temperature and then 250 mL of distilled water was added to the mixture to stop the reaction. Then, 10 mL of 30 wt.% H₂O₂ was added to reduce manganese ion to soluble manganese sulfate and manganese oxides. The purpose of H₂O₂ was to reduce residual KMnO₄ to soluble manganese sulfate (MnSO₄) in an acidic medium, as described in the following reaction [12]:



When 30 wt.% H₂O₂ was added, bubbling occurred and a bright yellow color was observed, indicating a high level of oxidation. The solution was then filtered using the filter paper to remove the metal sulfate and a graphite oxide (GTO) filter cake was produced. The cake was washed with 5% HCl aqueous solution until the sulfate ions are removed

completely. The removal of the metal sulfate ions was confirmed using BaCl₂ solution. The washing process was carried out repeatedly by centrifugation at 4000 rpm for 4 hrs, and the supernatant was decanted away. The pH of the collected material was checked using a universal indicator. The collected material (GTO) was stirred in distilled water at 60°C for 12 hours in a water bath. This process is called exfoliation [13].

2.2. Reduction of Graphene Oxide (rGO). Graphene oxide was reduced by using ascorbic acid as a reducing agent. 400 mg of GO powder was dispersed in 400 mL of distilled water (0.1 mg·mL⁻¹). Next, 4 g of ascorbic acid (AA) was added to this solution and stirred with a magnetic stirrer for 30 min at 60°C; the reduced product was centrifuged at 4000 r/s for 40 min to remove the supernatant. Then, excess 30 wt.% H₂O₂ was added to the black paste to oxidize the remaining ascorbic acid by stirring for 30 min at 60°C. After stirring, the resultant black product was collected by centrifugation at 4000 r/s, washed with ethanol and distilled water 3 times, respectively, and dried at 120°C for 24 hours. The preparation process of reduced graphene oxide (rGO) consists of three steps: oxidation or intercalation, exfoliation, and reduction.

- (i) Oxidation or intercalation: graphite oxide (GTO) was prepared from graphite powder by reacting with a strong oxidizing agent called potassium permanganate mixed with H₂SO₄/H₃PO₄ (9:1) (v/v) at 50°C for 12 hours in a water bath. H₂SO₄ is the most common intercalating agent.
- (ii) Exfoliation: in this step, the oxidized form of graphite was dispersed into distilled water to form single-layer graphene oxide (GO), and then it was heated with a magnetic stirrer for 12 hours at 60°C in a water bath. Then, the black paste was collected by filtration followed by centrifugation and dried at 60°C for 24 hours.
- (iii) Reduction: it is the last stage in synthesizing reduced graphene oxide (rGO). In this stage, the GO obtained from stage (ii) was dispersed and ascorbic acid was added as the reducing agent, followed by heating it at 60°C for 30 min. The reduced product was collected using filtration followed by centrifugation. Lastly, the black product was washed with ethanol and distilled water three times, respectively, followed by centrifugation; then, the product was collected and dried in an oven at 120°C for 24 hours.

2.3. Characterization Methods. Visual characterization means directly observing the changes in GO before and after reduction. The color change in the sample was captured by using a video camera. A UV-visible spectrometer (PerkinElmer, USA) was used to evaluate the photocatalytic activities of the sample. The GO and rGO solution samples were scanned at a wavelength ranging from 200 nm to 800 nm. FTIR (PerkinElmer, USA) was employed to analyze the presence/absence of functional groups on GO and rGO

sheets. GO and rGO pellets were prepared using KBr, and the samples were scanned in the range from 400 cm^{-1} to 4000 cm^{-1} to obtain the FTIR spectra. The X-ray diffraction (XRD Philips X-ray diffractometer) patterns of GO powders were recorded with a scanning rate of 1° per minute in a 2θ range from 5° to 80° with Cu-K α radiation ($\lambda = 1.5418\text{ \AA}$) to characterize the interlayer spacing.

3. Results and Discussion

3.1. Visual Observation. The color changes were simply observed when chemicals were added. These color changes were captured by using a video camera. Scheme 1(a) shows that the powder of graphite is dark with a luster. It can be seen from Scheme 1(b) that the graphite oxidized by concentrated $\text{H}_2\text{SO}_4/\text{H}_3\text{PO}_4$ and KMnO_4 is greenish dark paste. For instance, the mixture of sulfuric and *ortho*-phosphoric acids and potassium permanganate in the oxidation process was used to improve the efficiency of the oxidation. This process is still widely investigated for two reasons: (1) it gives an opportunity to produce large-scale graphene or few-layered graphene and (2) it has the potential to provide the samples with a controlled number of graphene layers upon the process parameter optimization [14].

The paste can cling on to the glass rod. In mixing process, potassium permanganate was gradually added with continuous stirring to control the reactivity. If the oxidizing agent is added once, it forms dark brown ash in gaseous form. At the end of 12 hours of boiling at 50°C with a magnetic stirrer, the greenish dark product was converted into paste (Scheme 1(c)). The paste mixed with a 250 mL of water (Scheme 1(d)) and 10 mL of 30 wt.% H_2O_2 became a brown-yellow suspension, which was then converted to pure yellow, as shown in Scheme 1(e). This complete color change indicated the oxidation of graphite to graphite oxide (GTO) at higher level. The model in Scheme 1(f) shows that the GTO suspension with acids and metal ions is directly filtered, and then the surface of the filter is quickly covered with a compact film, which could block the water passing (Scheme 1(g)). The suspension was centrifuged (Scheme 1(h)), and the supernatant was decanted away. Scheme 1(i) indicates that the GTO paste without metal ions and acids is dark brown. Lastly, the GTO solution was yellow, and this was extrafoliated at 60°C for 12 hours. It then turned to light brown color, as shown in Scheme 1(j). Then, the film form of GO was dried in the oven (Scheme 1(k)). As shown in Scheme 1(l), the powder of GO was mixed with 1 : 1 wt. ratio of ascorbic acid and a solution was formed with distilled water, which became dark. The rGO solution was converted to rGO film after drying in the oven for 24 hours (Scheme 1(m)). Lastly, the film was grinded using a mortar to obtain the powder of rGO (Scheme 1(n)).

The suspension with metal ions and acids is washed with 5% HCl solution to remove the metal ions [15]. If the GTO is continually washed with deionized water, the pH will increase. The removal of sulfate ions was detected by barium chloride solution. As the supernatant contains some unwanted ions, the solution formed a white precipitate. To remove the acidity of GTO, it was washed with deionized water until the pH becomes near to 7. The suspension

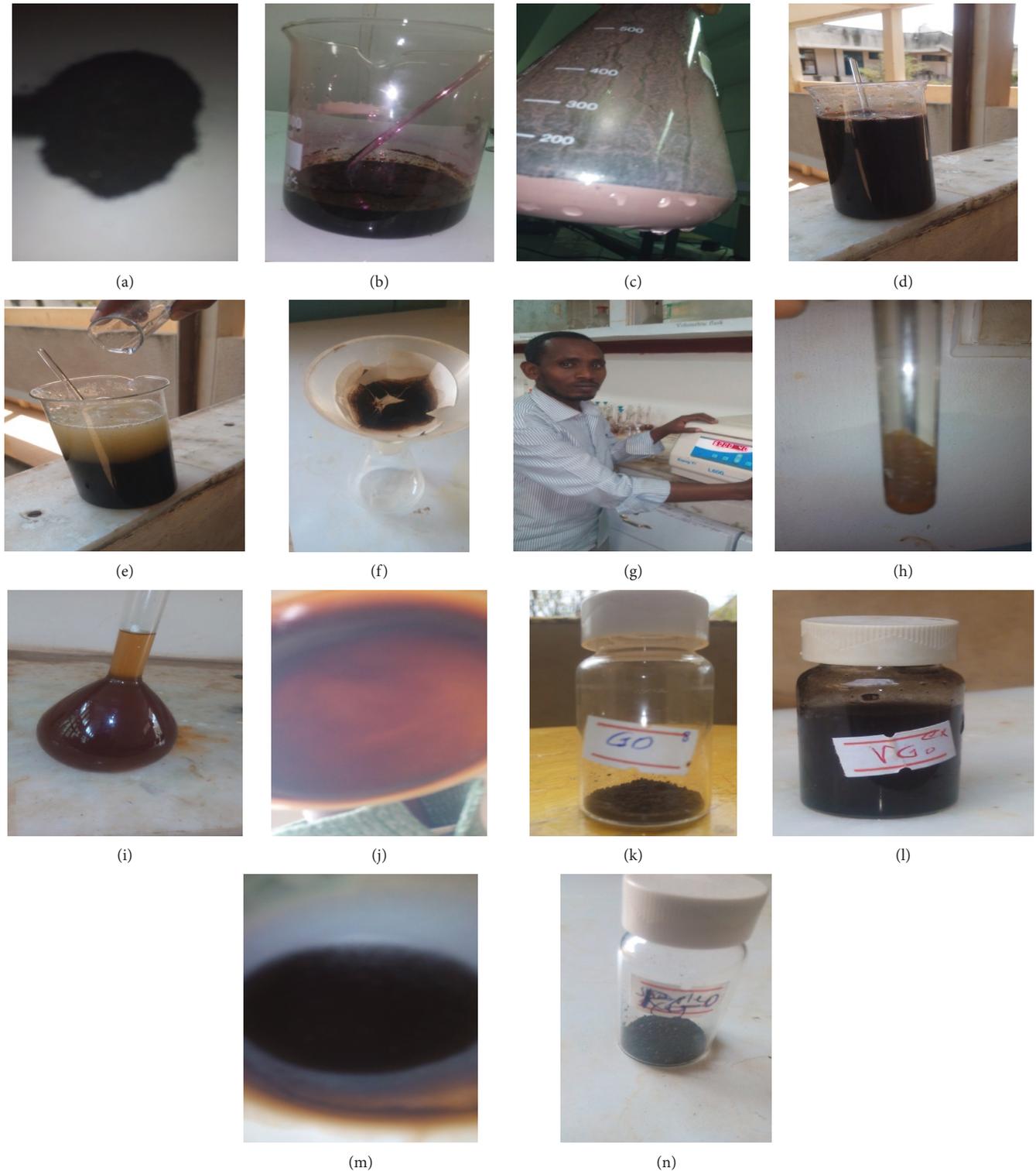
gradually becomes colloid. The chemical reduction of graphene oxide was performed using L-ascorbic acid as the reductant. Before reduction, the graphene oxide dispersion with L-ascorbic acid was light brown, as shown in Scheme 2(a). The mixture was then heated at 60°C for 40 min, and black graphene dispersion was observed, as shown in Scheme 2(b). The clear color change of the dispersion before and after the chemical reaction is apparent, evidencing that graphene oxide is indeed reduced by L-ascorbic acid into graphene [16]. The chemical reduction in a colloidal state, e.g., ascorbic acid (AA) reduction, usually results in a black precipitation from the original yellow-brown suspension, which is probably a result of an increase in the hydrophobicity of the rGO [17].

3.2. UV-Vis Characterization

3.2.1. Spectra of GO at Different Concentrations. During the oxidation of graphite, oxygen attaches to the graphene layers, thereby increasing the polarity of the layers which in turn increases their solubility in water. This results in a change in the color of the solution from yellow to brown; depending on the concentration of GO, brown color can be of different intensities. The GO that is formed has an absorption maximum at 225 nm for a well-oxidized material [18].

As the Beer-Lambert law states that absorbance is directly proportional to concentration, a sample with greater concentration has higher absorption than a sample with smaller concentration. Samples 1, 2, 3, and 4 indicated in Table 1 show absorbance at 226 nm, 225 nm, 224 nm, and 227 nm, respectively. These samples contained graphite to potassium permanganate in the weight ratio of 1 : 7, 1 : 8, 1 : 9, and 1 : 10, respectively, as shown in Figure 1. As the amount of the oxidizing agent increased, the oxidation capacity of graphite oxide also increased. This is due to the nature of potassium permanganate. Potassium permanganate is a well-known strong oxidizing agent. In addition to the nature of the oxidant, the oxidation level of GO can be decreased, e.g., by limited reaction time [2, 19], the amount of oxidant [20], and lower temperature [21]. But as the concentration increased, it could be out of optimum oxidizing capacity due to tearing of the basal layer of the sample. Therefore, Sample 3 with $\lambda_{\text{max}} = 224\text{ nm}$ was more oxidized due to the aforementioned reason.

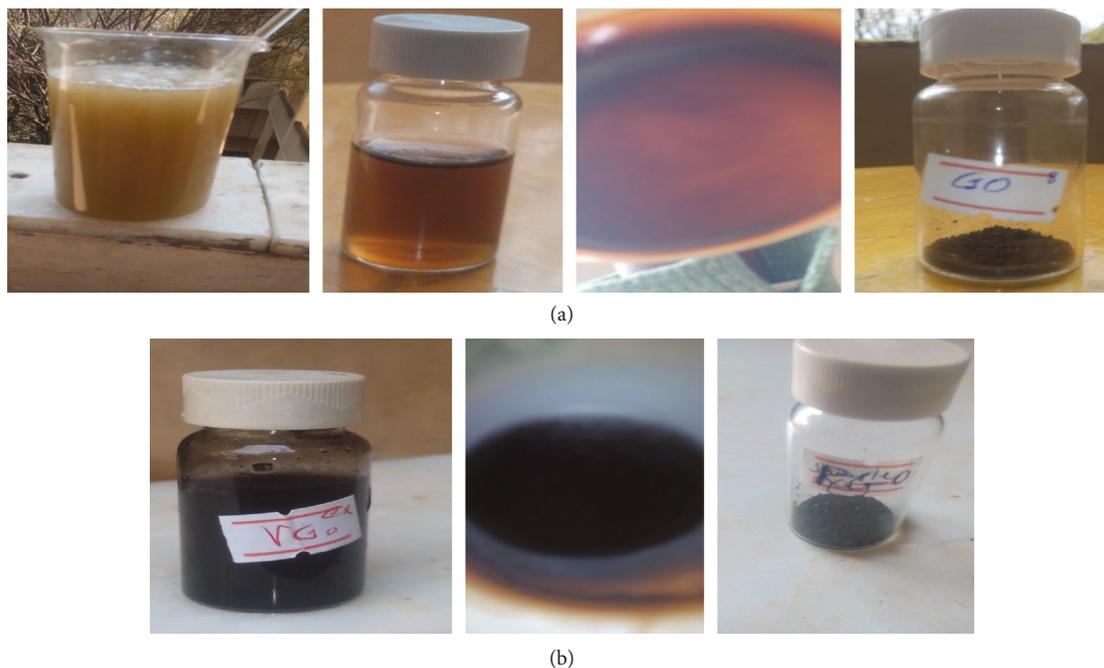
3.2.2. Synthesis of GO at Different Temperatures. The difference in oxidation behavior of graphite is widest at lowest temperature, tending to disappear as the temperature increases. The oxidation of graphite at constant concentration and time is highly temperature dependant. As temperature increased, the rate of oxidation increased rapidly. At 40°C , it shows absorbance at 230 nm; at 50°C , it shows absorbance at 224 nm; and at 60°C , it shows absorbance at 227 nm, which is indicated in Table 2 and Figure 2. According to the above explanation, at 60°C , the rate of oxidation becomes higher. But as the temperature increased in the reaction, carbon monoxide (CO) and carbon dioxide (CO_2) were removed in the gaseous form, and this is due to exfoliation [22]. At 40°C , it shows



SCHEME 1: Steps of graphene oxide and reduced graphene oxide synthesis.

absorbance at 230 nm which may contain some unoxidized graphite. Therefore, at 50°C, it shows absorbance at 224 nm, which is selected as the reaction temperature for the degree of oxidation of graphite to graphene oxide (GO). It is known that it detonates when heated to temperatures greater than 55°C [23].

3.2.3. Synthesis of GO at Different Time Periods. Reaction time is an essential factor for oxidation and reduction. It is found that the carbon content increased with increasing annealing time. Samples 8, 9, and 10, as shown in Table 3 and Figure 3, show absorbance at 231 nm at 10 hours, 223 nm at 12 hours, and 227 nm at 14 hours, respectively. As the time



SCHEME 2: Visual characterization of (a) GTO solution and GO solution, film, and powder and (b) rGO solution, film, and powder. Solutions in (a) and (b) were formed at the 1 : 1 wt./v ratio of the sample with distilled water as the solvent, and their images were taken using a video camera.

TABLE 1: Synthesis of GO at different mixture weight ratios of graphite and potassium permanganate.

Sample	Graphite (g)	KMnO ₄ (g)	H ₂ SO ₄ /H ₃ PO ₄ ratio (v/v)	H ₂ O ₂ (mL)	Temperature (°C)	Time (hours)
1	0.5	3.5	9:1	10	50	12
2	0.5	4.0	9:1	10	50	12
3	0.5	4.5	9:1	10	50	12
4	0.5	5.0	9:1	10	50	12

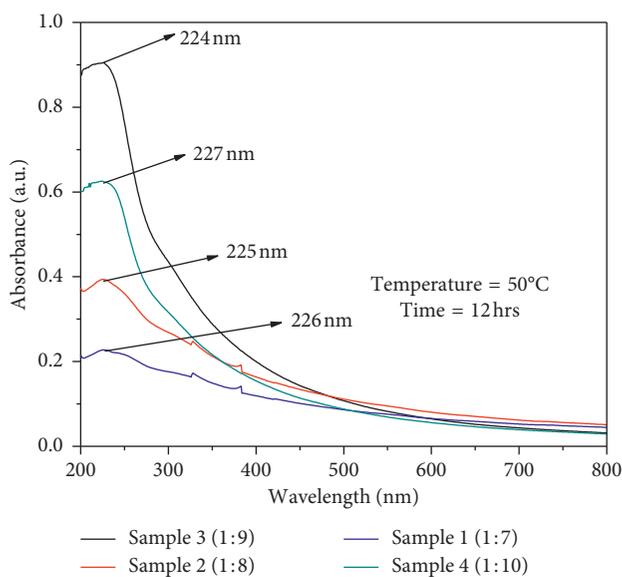


FIGURE 1: UV-Vis spectra at different concentrations.

rate increased, the oxidation capacity also increased. But as the reaction time increased, it became overoxidized [23]. This overoxidation may cause tearing of the basal layer. In a

short period, the oxidizing capacity decreased [24]. Therefore, Sample 9 underwent better oxidation of graphite oxide to graphene oxide, which was observed at 12 hours of oxidation time that showed absorbance at 223 nm. The effect of sonication process was also examined, and it was found to enhance the exfoliation to bilayer graphene. It suggests an enhanced exfoliation of graphite when oxidation was performed with ultrasonication treatment and when time and temperature were increased, also confirming the enhanced oxidation effect [24].

3.2.4. Reduction of Graphene Oxide. Graphene oxide (GO) that was obtained from the oxidation of 0.5 g graphite with 4.5 g of potassium permanganate and (90 : 10) mL v/v ratio of sulfuric and phosphoric acids was reduced by 4.0 g ascorbic acid to get reduced graphene oxide (Scheme 3), as shown in Figure 4. In chemical reduction process, AA (ascorbic acid) works both as a reducing agent and a protecting agent, which makes the process economical, nontoxic, and environmental friendly. From Figure 4, it is observed that graphene oxide (GO) shows a maximum absorption peak at ~ 226 nm attributable to π - π^* transition of the atomic C=C bonds and a shoulder peak at ~ 300 nm due to n - π^* transitions of C=O

TABLE 2: Synthesis of GO at different reaction temperatures.

Sample	Graphite (g)	KMnO ₄ (g)	H ₂ SO ₄ /H ₃ PO ₄ ratio (v/v)	H ₂ O ₂ (mL)	Temperature (°C)	Time (hours)
6	0.5	3.5	9:1	10	40	12
7	0.5	3.5	9:1	10	50	12
8	0.5	3.5	9:1	10	60	12

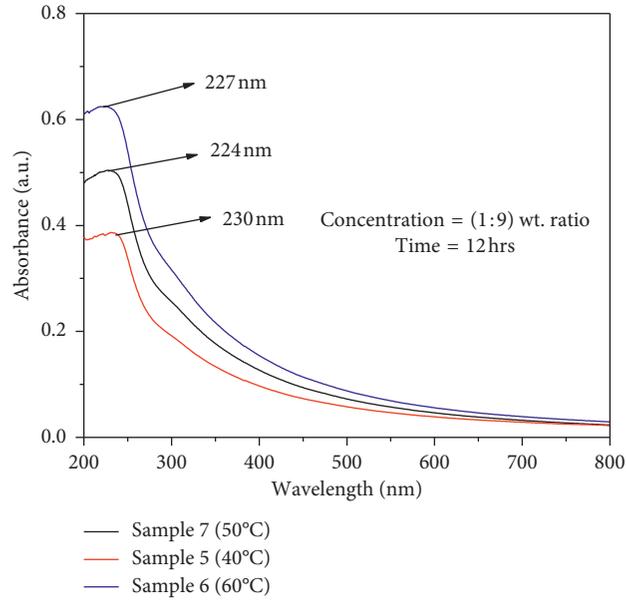


FIGURE 2: UV-Vis spectra at different temperatures.

TABLE 3: Synthesis of GO at different reaction time periods.

Sample	Graphite (g)	KMnO ₄ (g)	H ₂ SO ₄ /H ₃ PO ₄ ratio (v/v)	H ₂ O ₂ (mL)	Temperature (°C)	Time (hours)
6	0.5	3.5	9:1	10	50	10
7	0.5	3.5	9:1	10	50	12
8	0.5	3.5	9:1	10	50	14

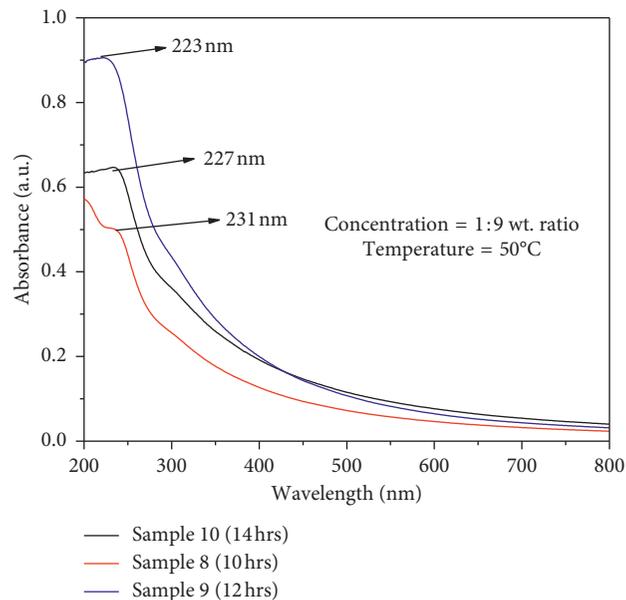
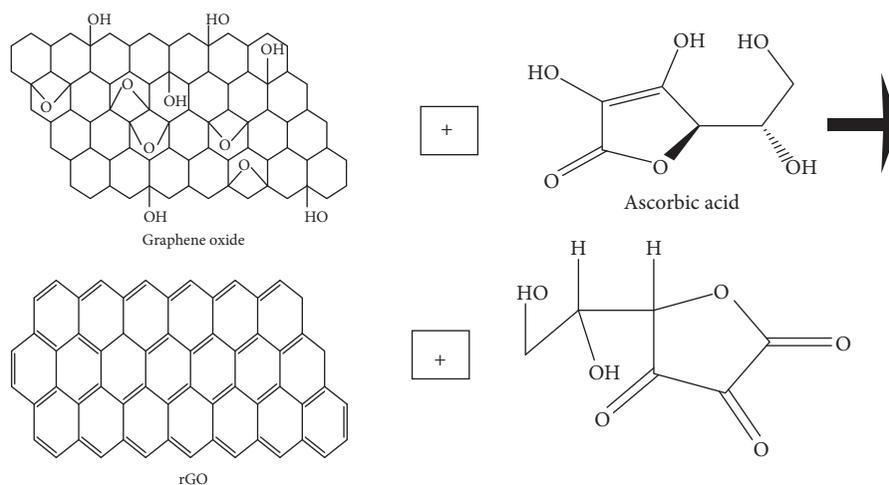


FIGURE 3: UV-Vis spectra at different time periods.



SCHEME 3: Schematic representation of reduced graphene oxide.

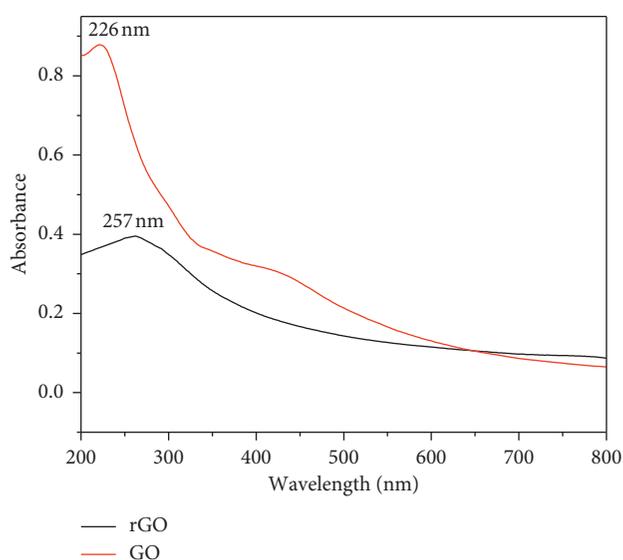


FIGURE 4: UV-Vis spectra of GO and rGO.

bonds [25]. The successful synthesis of reduced graphene oxide (rGO) was confirmed by UV-visible spectroscopy (Figure 4). The UV-visible spectrum of graphene oxide (the black curve in Figure 4) shows an absorption peak at 226 nm.

After reduction by using AA, the peak of reduced graphene (the red curve) was observed at 257 nm. The absorption of reduced graphene (red curve) shifts from 226 to 256 nm, suggesting that the electronic conjugation within graphene sheets is restored after the reduction [26].

3.3. FTIR Characterization. In order to examine the efficiency of introduction of oxygen-containing functional groups into carbon lattice, FTIR spectroscopy was used. At each spectrum, several typical modes corresponding to oxygen-containing functional groups were detected. Here, stretching vibration modes of C–O bonds arise at $1,078\text{ cm}^{-1}$. The peaks between $1,190\text{ cm}^{-1}$ and $1,382\text{ cm}^{-1}$

are related to C–OH stretching vibration [25]. Deformation vibration modes of O–H groups were observed at $1,431\text{ cm}^{-1}$. The bands appeared at $1,880\text{ cm}^{-1}$ correspond to adsorbed water molecules, indicating hydrophilic properties of the graphene oxide. At approximately $1,622\text{ cm}^{-1}$, C=O stretching vibration of carboxyl groups was detected. The peaks at around $3,400\text{ cm}^{-1}$ arise from O–H stretching vibration. The peak arising from C–H stretching vibration at around $2,854\text{ cm}^{-1}$ was observed.

When reducing graphene oxide to graphene, the functional groups disappeared. This is clearly reflected in the FTIR spectrum (Figure 5) for not having bands corresponding to any functional groups. The presence of oxygen-containing functional groups, such as C=O and C–O, further confirmed that the graphite indeed was oxidized into GO and was consistent with the literature [27]. The presence of C=C groups showed that even graphite oxidized into GO; in the main structure of the layer, graphite was still retained.

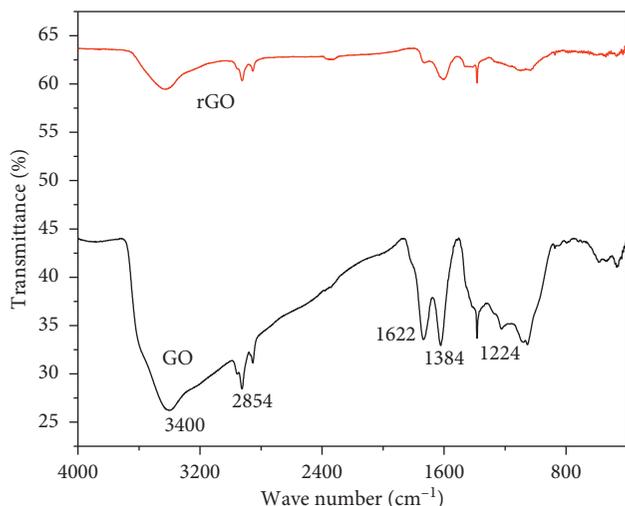


FIGURE 5: FTIR spectra of GO and rGO.

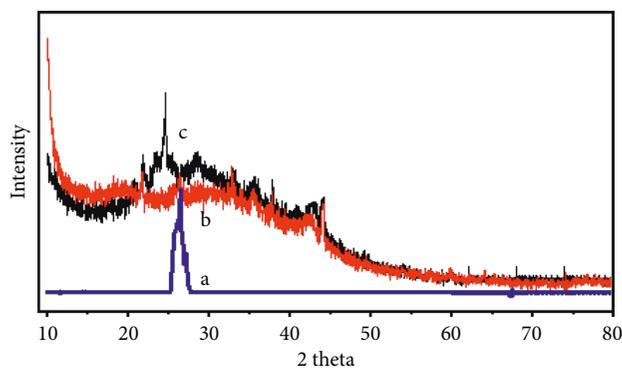


FIGURE 6: X-ray diffraction patterns of (a) graphite powder, (b) GO powder, and (c) rGO powder.

3.4. XRD Characterization. X-ray diffraction patterns for graphite powder, GO powder, and rGO powder were recorded. As shown in Figure 6(a), the diffraction peak for graphite was at $2\theta = 26.4^\circ$ with a corresponding layer to layer distance of 0.337 nm, which is similar to the reported value in the literature [28]. After oxidation of graphite, the diffraction peak for GO was at $2\theta = 10^\circ$ corresponding to a layer to layer distance of 0.88 nm, which is in good agreement with the literature [14, 29]. The patterns showed a larger interlayer spacing of GO than graphite layers due to the formation and introduction of oxygen-containing functional groups between layers [30]. These functional groups facilitated the hydration and exfoliation of GO in water. In addition, the reflection peak appeared around 43° in the XRD pattern of GO (Figure 6(b)), indicating that GO exhibits turbostratic disorder [31], which is believed to be due to incomplete oxidation. The pattern was also not smooth. This might be due to the irregular spacing of GO layers. XRD analysis also confirms the reduction of GO. As seen in Figure 6(c), the diffraction peak of GO ($2\theta = 10^\circ$) shifted to $2\theta = 23.8^\circ$ (diffraction peak of rGO) corresponding to a layer to layer distance of 0.37 nm [32]. This indicates that after reduction, the interlayer distance of rGO

was lower than that of GO due to the removal of oxygen-containing functional groups and agglomeration of reduced graphene oxide sheets. As seen from the figure, the XRD pattern of rGO is not smooth and small peaks were observed. The reason seems that there were residual functional groups on the rGO sheets and affected the structure of the rGO sheet.

4. Conclusion

Synthesis of graphene oxide was achieved by placing graphite in concentrated acid in the presence of an oxidizing agent. The Tour method is a less hazardous and more efficient method for graphite oxidation because it is cost-effective, nontoxic, and environmental friendly. This is the most commonly used methods for the oxidation of graphite. Graphene oxide was reduced by chemical reduction process using AA (ascorbic acid) that works both as a reducing agent and a protecting agent, which makes the process economical, nontoxic, and environmental friendly.

The oxidation level of graphite powder was observed using different parameters such as reaction time, temperature, and quantity of KMnO_4 . Graphene oxide (GO) and reduced graphene oxide (rGO) both were characterized by different characteristic techniques. These techniques include visual observation, UV-Vis characterization, FTIR characterization, and XRD (X-ray diffraction) techniques. UV-Vis spectroscopy showed formation of GO and rGO with maximum absorption peaks at 226 nm and 257 nm, respectively. FTIR spectroscopy spectra showed the introduction of different oxygen-containing functional groups in GO during oxidation and removal of these functional groups after reduction. XRD also showed that the diffraction peak of GO ($2\theta = 10^\circ$) shifted to $2\theta = 23.8^\circ$ (diffraction peak of rGO) corresponding to a layer. This indicated that after reduction, the interlayer distance of rGO was lower than that of GO due to the removal of oxygen-containing functional groups and agglomeration of reduced graphene oxide sheets. The overall results of the study suggests that better oxidation level was obtained by treating 0.5 g graphite with 4.5 g of KMnO_4 (1 : 9 wt/wt ratio) in a 9 : 1 of $\text{H}_2\text{SO}_4/\text{H}_3\text{PO}_4$ (v/v) for 12 hours at 50°C of reaction temperature.

Data Availability

All data generated or analyzed during this study are included within the article.

Conflicts of Interest

The authors declare that they have no conflicts of interests.

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