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SYNTHESIS AND CHARACTERIZATION OF GRAPHENE OXIDE USING MODIFIED HUMMERS METHOD

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ABSTRACT. Since graphene is expensive and relatively difficult to produce, significant efforts have been made to develop effective and inexpensive methods for creating and utilizing graphene [1], derivatives, or related materials. Graphene oxide (GO) is one such material. It consists of a single layer of atoms produced through the strong oxidation of cheap and abundant graphite. GO is an oxidized form of graphene containing oxygen-functional groups. Due to its ability to disperse in water and other solvents, GO is easy to process and can even serve as a precursor for graphene production. Although GO is not a good conductor, various processes can enhance its properties. GO is typically synthesized as a powder, in dispersion, or substrate coating. There are four primary methods for synthesizing GO: Staudenmaier, Hofmann, Brodie, and Hummers, with many variations available and ongoing improvements aimed at achieving better results and more cost-effective processes. The efficiency of the oxidation process is typically evaluated by the carbon-to-oxygen ratio of GO. In this study, graphene oxide was synthesized from graphite using a modified Hummers method. In this modification, an ice bath and sodium nitrate ($NaNO_3$) were omitted, and the synthesis was conducted at room temperature. FT-IR, RAMAN, XRD, SEM, and EDS analyses were performed for characterization, yielding results consistent with findings reported in the literature.

Keywords. Graphene oxide, graphite powder, modified hummer's method, XRD, characterization.

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1. INTRODUCTION

Graphene oxide (GO) has been studied for over 150 years [1]. Originally called graphite oxide, it was first synthesized in 1859 by Benjamin Brodie, an Oxford University chemist, by exfoliating graphite flakes using potassium chlorate and fuming nitric acid [2]. In 1957, William Hummers and Richard Offeman enhanced this synthesis method, creating what is now known as the Hummers method, which sped up and improved safety by using sulfuric acid, sodium nitrate, and potassium permanganate [1, 2]. The Hummers method is still widely used today, with various modifications to address environmental and safety concerns [2]. A modified Hummers method is often employed to synthesize GO from graphite by treating it with strong oxidants like potassium permanganate, hydrogen peroxide, sulfuric acid, and hydrochloric acid [3]. This chemical process separates graphite into single or multiple atomic layers, expands the interlayer structure, and introduces functional groups [4]. The nature and amount of oxygen-containing functional groups make GO hydrophilic, allowing it to dissolve in water [5]. The synthesis of GO, originally known as graphite oxide, involves reacting graphite with strong oxidants, typically potassium permanganate and sulfuric acid. The resulting product is then rinsed and centrifuged until the pH is neutral, followed by freeze-drying to maintain its solubility [6]. In some cases, manufacturers use dialysis to purify GO. However, contamination with alkali salt byproducts during synthesis makes GO highly flammable due to salt-catalyzed carbon combustion [5, 6]. There are four common methods for synthesizing GO: Staudenmaier, Hofmann, Brodie, and Hummers. Variations of these methods, such as the Tur method (a modified Hummers method), exist [7]. Current research focuses on refining synthesis techniques for improved quality, lower environmental impact, and cost reduction [8, 9]. Due to oxidation, GO's electronic properties differ from those of pristine graphene (e.g., CVD or epitaxial graphene) as structural defects interrupt its sp^2 bonding networks. GO's conductivity can be improved by thinning it or combining it with more conductive materials, making it advantageous for practical applications [10].

The optical properties of GO make it an optically transparent material. While not fully transparent, GO films less than five layers thick can achieve over 90% optical transparency, with each additional graphene layer reducing transparency by about 2%. Transparent conductive inks and films can be created by combining GO with carbon nanotubes or silver nanowires [11]. Graphene's superior thermal properties have spurred interest in its use for thermal management applications. Single-layer graphene exhibits high thermal conductivity, but the addition of even one more layer significantly reduces this property. The interlayer spacing plays a crucial role in thermal conductivity, with both the number of layers and the space between them affecting the overall heat transfer [12]. Studies show that GO's thermal conductivity is closely related to oxygen group presence. At an oxygen content of 0.5%, GO's thermal conductivity is approximately 50% lower than that of pristine graphene. As the oxygen content increases, thermal conductivity decreases, with the lowest recorded thermal conductivity for GO being about 8.8 W/mK, lower than the theoretical minimum of 11.6 W/mK [13]. This suggests that thermal transport in GO can be tailored by controlling its oxidation and reduction levels [14]. The mechanical properties of GO are substantially lower than those of pristine graphene. A monolayer of GO has an effective Young's modulus ranging from about 207.6 to 23.4 GPa (assuming a thickness of 0.7 nm), significantly lower than pristine graphene's Young's modulus, reported around 1.0 TPa, with an ultimate fracture strength near 1.0 TPa [15, 16]. Graphene oxide (GO) has a wide range of applications, including catalysis, drug delivery, solar energy, batteries, scaffolds, water desalination, and many other fields [17, 19] . Its high solubility and ability to be reduced make GO well-suited for solution processing, making it a highly desirable nanomaterial. GO also addresses the dispersion challenges often seen with other carbon-based materials like carbon nanotubes [18]. GO has been used as a foundational material in electronic device fabrication [20]. Additionally, studies have shown that GO exhibits good biocompatibility, making it promising for drug delivery. Unlike other carbon nanomaterials such as carbon nanotubes (CNTs), GO does not cause oxidative stress as its synthesis does not use metal catalysts, thus avoiding metal impurities. The functional groups on GO's surface allow it to interact effectively with a variety of organic and inorganic molecules through covalent, non-covalent (π-π stacking or hydrophobic interactions), and/or ionic interactions [21]. These properties further enhance its potential for drug delivery applications [22].

2. MATERIALS AND METHODS

2.1 Materials

Natural graphite flakes (−100 mesh, 99.9% Sigma-Aldrich), H_2SO_4 (95-97%, Sigma-Aldrich), H_3PO_4 (85%, Sigma-Aldrich), $KMnO_4$ (>%99, Sigma-Aldrich), H_2O_2 (35%, Sigma-Aldrich), HCl (37%, Sigma-Aldrich) chemicals were used in the experiments. Deionized water was provided using the Millipore Simplicity ultrapure water system.

2.2 Methods

2.2.1 Synthesis of Graphene Oxide by Modified Hummers Method

GO synthesis was performed using the modified Hummers method [6]. An acidic mixture was prepared by combining 108 mL of sulfuric acid (H_2SO_4) with phosphoric acid (H_3PO_4) in a volume ratio of 9:1 at room temperature, utilizing a magnetic stirrer. Subsequently, 1 g of graphite powder was added to this mixture, and the mixing process continued for 5 minutes. Following this, 5.28 g of potassium permanganate ($KMnO_4$) was gradually introduced to the suspension. At this stage, the suspension appeared black, indicating the initiation of the reaction. Next, 2.70 mL of hydrogen peroxide (H_2O_2) was added dropwise to the mixture and thoroughly mixed. This addition triggered an exothermic reaction, after which the mixture was allowed to cool. Once cooled, a mixture of 25% hydrochloric acid (HCl) and deionized water was added to the suspension. The resulting mixture was then filtered using a vacuum filtration system. Filtration was done using nitrocellulose membrane and washed several times with a hydrochloric acid and deionized water solution. Following washing, the mixture was centrifuged using a Mini Centrifuge - D1008 model - at 7,000 rpm. Finally, the prepared samples were dried in an oven at 80 °C for approximately 48 hours, resulting in graphene oxide being obtained in powder form.

2.3. Graphene Oxide Characterization

2.3.1. Fourier Transform Infrared Spectroscopy (FT-IR)

FT-IR is a chemical analytical method that measures the wavenumber against the infrared intensity of light using the mathematical Fourier transform. This method is based on the absorption of infrared radiation by intramolecular bonds, which occurs due to the vibrational and rotational movements of these bonds. Infrared radiation is absorbed as a result of molecular vibrations. By utilizing this technique, one can characterize the bonding of graphene oxide (GO) molecules and identify functional groups within the structures of organic compounds, assess the state of bonds, determine bonding sites, and ascertain whether the structure is aromatic or aliphatic. In this study, a Perkin Elmer Spectrum Two device with a spectral range of 8300 to 350 cm^{-1} was employed for the analysis of GO. Powdered GO was analyzed by pelleting it with potassium bromide (KBr).

2.3.2. Raman Spectroscopy

The foundation of Raman spectroscopy is Raman scattering. This technique is a crucial spectroscopic method used for both qualitative and quantitative analysis of inorganic and organic systems. It is rapid, straightforward, and can be applied without damaging the sample. Raman spectroscopy is complementary to infrared (IR) spectroscopy, allowing for the detection of weak vibrations that may not be observable in IR. In this study, a Renishaw inVia Raman Microscope equipped with 532 nm and 735 nm lasers was employed for the characterization of graphene oxide (GO).

2.3.3. X-Ray Diffraction Analysis (XRD)

XRD method is based on the principle that each crystal phase diffracts X-rays in a characteristic manner, depending on its unique atomic arrangement. In standard qualitative XRD analyses, samples are examined within a 2-70 degree range using nickel-filtered copper X-ray tube devices, and the resulting X-ray diffractograms are interpreted according to ASTM standards. In this study, a Panalytical Empyrian device was utilized for the analysis of graphene oxide (GO). This device enabled the observation of phase changes in the crystal structure under atmospheric, vacuum, and inert gas conditions at temperatures up to 1200 °C.

2.3.4. SEM and EDS Analysis

In this study, a JEOL JSM 5600 device was utilized for the analysis of the morphology, chemical composition, and topography of graphene oxide (GO). The topography, morphology, shape, size, and compositional structures were examined. Additionally, gold coating was applied using a Leica EM ACE600 coater prior to scanning electron microscopy (SEM) imaging, thus preparing the sample for SEM analysis. Energy-dispersive X-ray spectroscopy (EDS) was performed following the SEM imaging to determine the components present in the structure.

3. RESULT AND DISCUSSION

The degree of oxidation in graphene oxide (GO) varies based on the method employed, reaction conditions, and the properties of the graphite used. GO contains various amounts of carbon, oxygen, and hydrogen and functional groups such as epoxy, hydroxyl, carbonyl, and carboxyl [23]. Compared to known production methods reported in the literature, the Hummers method releases fewer toxic and hazardous gases and allows for the synthesis of larger quantities of GO at a lower cost [24]. In this study, the reaction began with 1 g of graphite powder, resulting in the production of 3.37 g of GO powder. This increase in weight indicates the formation of functional groups within the graphite structure.

3.1. FT-IR

Graphene oxide (GO), with its numerous functional groups, exhibits rich and complex FT-IR spectra [2]. The FT-IR spectra for GO are shown in Figure 1. Previous studies have reported varying interpretations of the FT-IR spectra of GO [6, 26, 27]. Functional groups such as O-H, C-OH, COOH, and C-O were identified in the FT-IR spectrum of GO [25]. The peaks between 2500 cm^{-1} and 3500 cm^{-1} correspond to the stretching modes of carboxyl O-H bonds, which arise from alcohols and water molecules incorporated into the GO structure; the peak around 3300 cm^{-1} reflects this [28]. Significant peaks at 2960 cm^{-1} and 2874 cm^{-1} are attributed to CH_2 symmetric and asymmetric bands, indicating the presence of unoxidized C=C bonds in graphite, which appear in the band at 1585 cm^{-1} [26, 29]. Additionally, peaks near 1250 cm^{-1} indicate the absorption of epoxy rings. Other notable absorption bands in the fingerprint region are observed at approximately 1351 cm^{-1} and 1452 cm^{-1}, which correspond to the bending modes of alcohol groups and the deformation modes of O-H in hydrogen bonds to heteroatoms of hydroxyl, epoxy, or sulfate groups, respectively. The most intense band in the fingerprint region is reported between 1039 cm^{-1} and 1060 cm^{-1}, with its shift attributed to structural differences in GO samples [26, 27, 30].

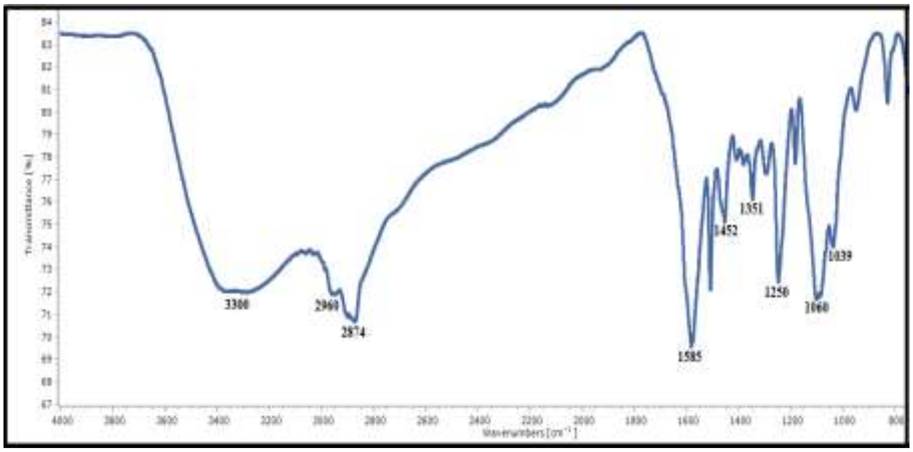

Figure 1. FT-IR spectra of GO.

3.2. Raman Spectroscopy

Raman analysis is a rapid, non-destructive technique used to examine structural changes in materials. In the Raman spectrum, the G and D peaks hold significant meaning. The G peak corresponds to the regular structures within the carbon framework, while the D peak indicates irregularities and defects in the carbon structure, which arise from oxidation [33]. Raman analysis of graphene oxide (GO) synthesized via the modified Hummers method was conducted. The spectra of the samples are presented in Figure 2. In the Raman spectrum, the D peak of GO appears at 1176 cm^{-1}, while the G peak is observed at 1588 cm^{-1}, consistent with the literature. The intensity of the D peak reflects the presence of irregular structures, whereas the intensity of the G peak indicates the presence of regular structures [31, 32]. The intensity ratio between the G and D bands provides an indication of the degree of graphitization. As shown in Figure 2, the intensity ratio (I_D/I_G) for GO is 0.74, likely due to the increased disordered structure caused by oxygen functional groups introduced during the oxidation of graphite [34, 35].

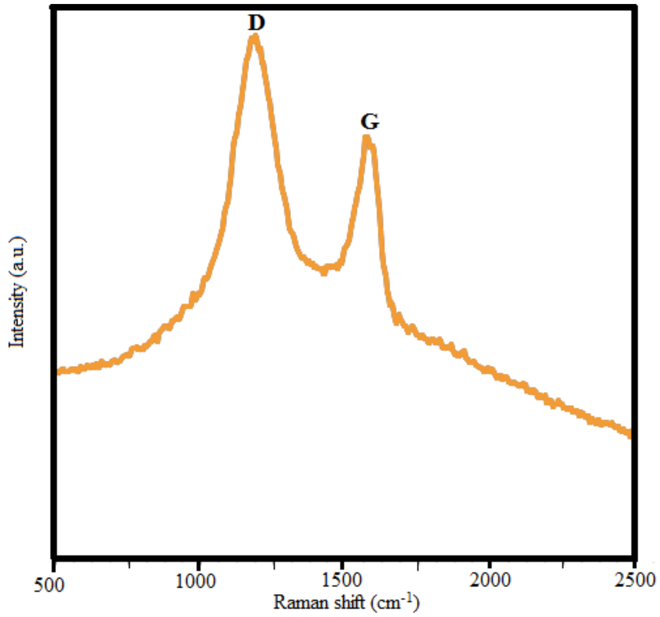

Figure 2. Raman spectra of GO.

3.3. X-Ray Diffraction Analysis (XRD)

XRD analyses for graphite and graphene oxide (GO) are presented in Figure 3. A sharp characteristic peak is observed at 2θ = 26.66° in the XRD pattern of the graphite sample, while a sharp characteristic peak at 2θ = 10.97° is noted for GO. The XRD values for graphite and GO are in close agreement with those reported in the literature [36, 37].

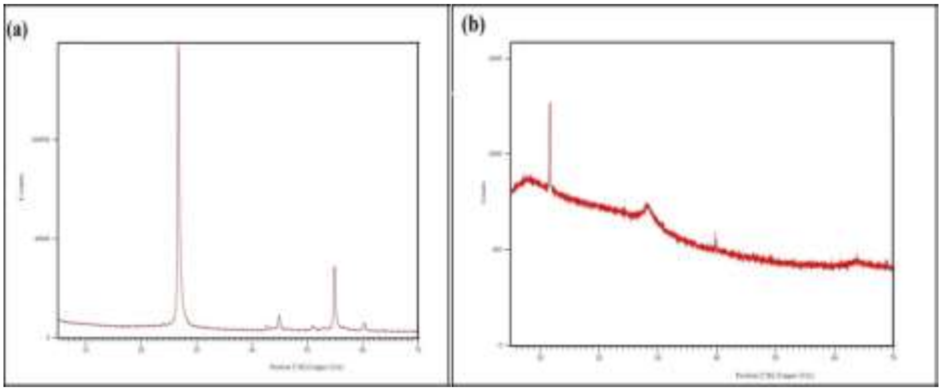

Figure 3. X-ray diffraction patterns of graphite (a) and GO (b).

3.4. SEM and EDS Analysis

Images of graphite and graphene oxide (GO) were obtained using a scanning electron microscope (SEM). The SEM images for graphite are shown in Figure 4, while those for GO are presented in Figure 5. Additionally, the amounts of carbon and oxygen elements and the carbon/oxygen ratio determined by the energy-dispersive X-ray spectroscopy (EDS) integrated into the SEM are provided in Figure 6 for graphite and Figure 7 for graphene oxide. The SEM images indicate that the graphite structure has fragmented and separated into smaller, more transparent GO structures as a result of the modified Hummers method. The images of GO reveal a layered, wrinkled surface with randomly distributed thin layers that are similar in appearance [38, 39]. EDS analysis showed that the carbon (C) ratio in the graphite structure was 100%, while in GO, it was 71.58%. The oxygen (O) ratio in GO was determined to be 28.42%.

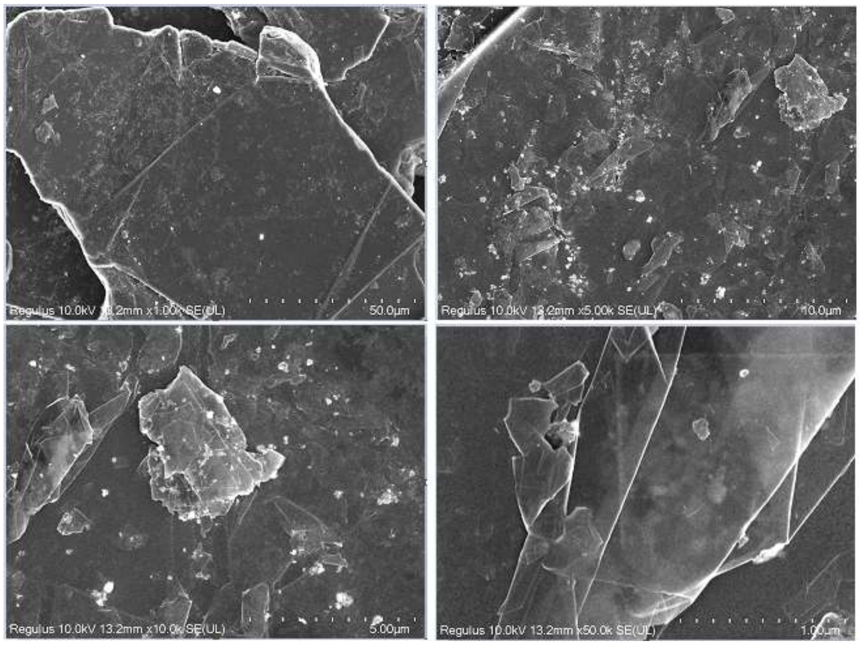

Figure 4. SEM images of graphite powder.

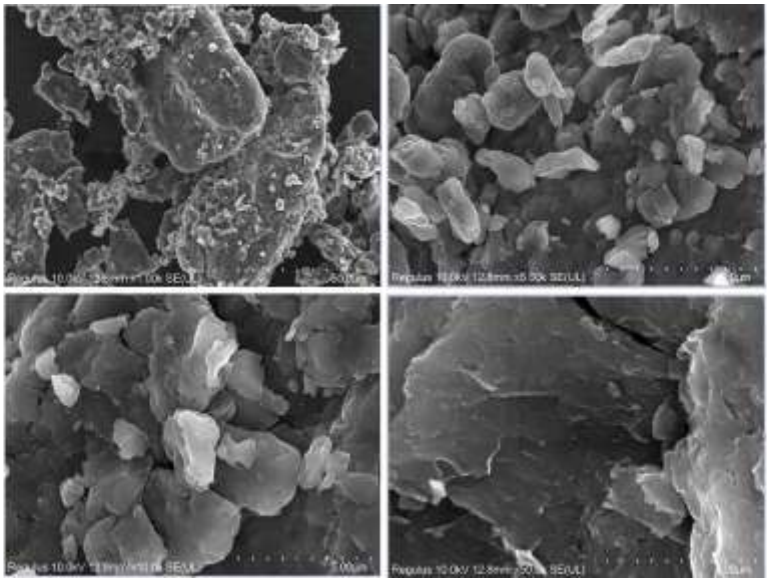

Figure 5. SEM images of GO.

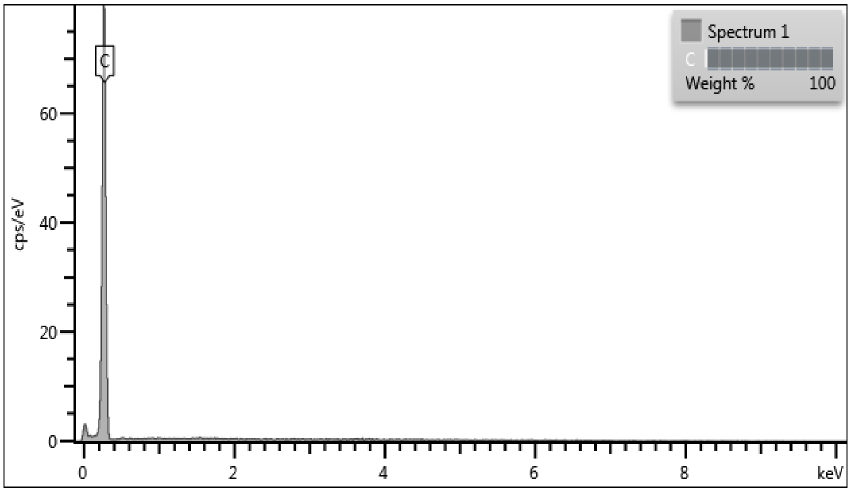

Figure 6. EDS analysis of graphite powder.

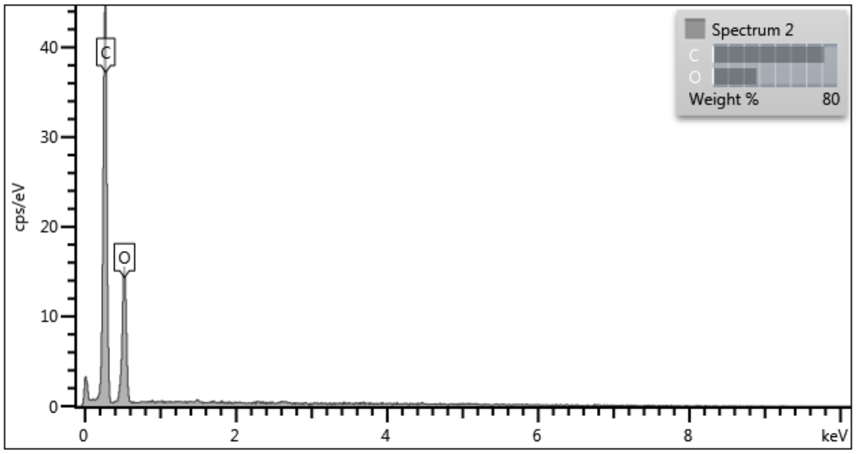

Figure 7. EDS analysis of GO.

4. CONCLUSION

In this study, graphene oxide (GO) was synthesized from graphite powder using a modified Hummers method. Unlike the traditional modified Hummers method, this study did not employ an ice bath and sodium nitrate ($NaNO_3$), suggesting that $NaNO_3$ does not have a significant impact on the process. The results indicate that $NaNO_3$ is not essential for synthesizing graphene oxide. There are several important advantages to not using $NaNO_3$ in graphene oxide synthesis, the most important of which is its positive aspects in terms of environmental impact. $NaNO_3$ can release nitrogen dioxide (NO_2) and other nitrogen-based compounds as by-products, which are harmful to the environment and contribute to air pollution. Excluding $NaNO_3$ minimizes the generation of these toxic gases, making the process more eco-friendly. Another advantage of not using $NaNO_3$ is that it is a safer experiment. The presence of $NaNO_3$ can increase the risk of uncontrolled exothermic reactions, as it reacts with sulfuric acid and potassium permanganate to produce potentially hazardous intermediates. By avoiding $NaNO_3$, the reaction is generally safer, with fewer toxic fumes released, making it easier to handle in laboratory settings. Another advantage of not using $NaNO_3$ is the reduced cost of experimentation. Simplifying the process by reducing the number of chemicals can also lower operational costs. Consequently, GO synthesized without $NaNO_3$ exhibited the same characteristic properties, confirmed through FT-IR, Raman spectroscopy, XRD, SEM, and EDS analyses.

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Author Contribution Statement Mahdi Ahanibarough—Conducted the experiments, created the graphs, performed the analysis, interpreted the results, and drafted the manuscript. Sabriye Perçin Özkorucuklu—Responsible for planning, reviewing, and editing the manuscript.

Declaration of Competing Interest The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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