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A Sustainable and Environmental-Friendly Acid-Reduced Route for Graphene Oxide Synthesis Through Modified Hummers Method

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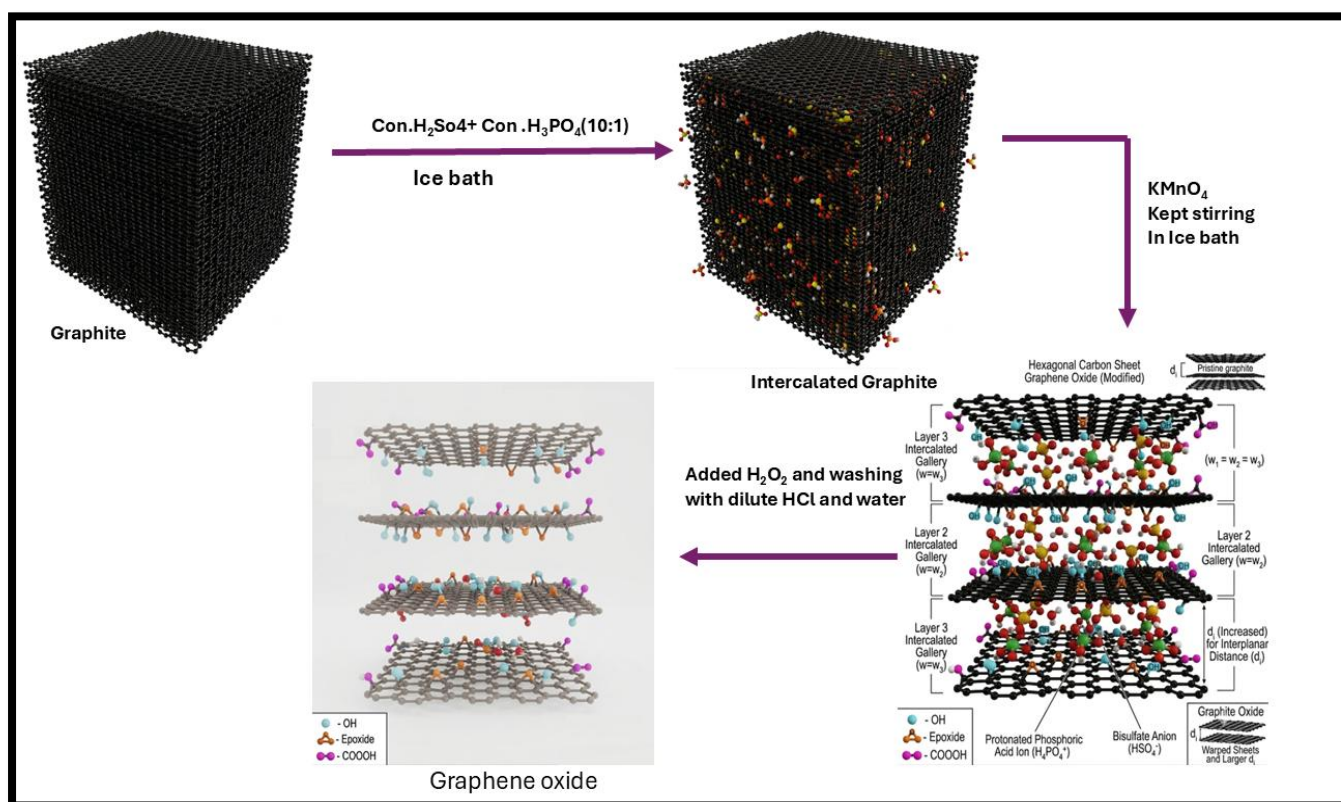
Abstract

The two-dimensional nanomaterial Graphene Oxide (GO) emerged as a cornerstone in materials science whose unique hydrophilic functional groups and tuneable physicochemical properties empower advanced applications in the fields of biomedicine, sensors, energy storage, catalysis etc. However, the Conventional Hummers Method for GO synthesis produces massive volumes of hazardous mineral acids waste, thereby presenting severe ecological hazards, complex wastewater management challenges and heightened operational safety risks related to exothermic reactions. Herein, we report a sustainable and green modification to the Hummers Method by suggesting an acid-sparse method for the preparation of GO, along with an effective oxidization of Graphite that has a reduced acid consumption. The modified protocol uses tailored synthetic parameters and a rate-controlled addition of the oxidant KMnO_4 to facilitate the Synthesis of well-exfoliated GO with reduced use of acids and enhanced operational safety. A comprehensive characterization was done in order to confirm the efficient exfoliation, the successful oxidation

Keywords

Graphene Oxide; Scalable Graphene Oxide Production; Acid-reduced Process; Sustainable Synthesis; Improved Hummers Method

Graphical Abstract



Schematic Diagram of Graphene Oxide preparation

Introduction

Graphene-based materials are amongst the most revolutionary classes of nanomaterials devised in the twenty-first century [1-2]. Since the isolation of Graphene, researchers have investigated a broad spectrum of methodologies for exploiting its exceptional electrical conductivity, mechanical strength, thermal stability, and large surface area [3]. Graphene Oxide is one of the various derivatives which has garnered considerable attention due to its synthetic convenience, rich surface functional groups, and its potential as a precursor for reduced-graphene-oxide and other functional Carbon nanostructures [4-6]. Graphene Oxide is made of atomically thin Carbon sheets decorated with Oxygen-containing functional groups such as hydroxyl, epoxy, carbonyl and carboxyl moieties. These functionalities disrupt the extended π conjugated structure of Graphene and makes it hydrophilic, enabling stable aqueous dispersion. These properties promote the integration of Graphene Oxide into a broad spectrum of

technological implementation including water purification membranes, flexible electronics, supercapacitors, batteries, sensors, catalysts, biomedical devices and polymer composites [6-13].

Graphene Oxide is primarily synthesized by the oxidation of Graphite. Among the diverse preparation pathways reported in relevant literature, the most heavily adopted methodology is still the Conventional Hummers Method due to its commercial viability and procedural convenience. The method, first developed in 1958, uses Potassium Permanganate and concentrated Sulfuric Acid to oxidize Graphite to Graphite Oxide, which can then be exfoliated to produce Graphene Oxide sheets. But the original Hummers Method has certain limitations. The process demands massive inputs of aggressive acid media and it produces environmentally toxic chemical footprints. Due to the safety hazards of the process, accumulation of perilous chemical residues and environmental sustainability concerns, many attempts have been made to modify and improve this method. Prominent among these advancements was the advent of the Improved Hummers Method that adopted a mixture of Sulfuric Acid and Phosphoric Acid as the reaction medium. The presence of Phosphoric Acid improved the oxidation efficiency and reduced the formation of toxic gases, even as it refined the product-quality metrics. However, this improved method still relies heavily on concentrated Sulfuric Acid and being so it preserves many of the environmental and economic challenges of large-scale Graphene Oxide production [14-18].

As the industrial synthesis of GO seeks a sustained commercial interest, its sustainable manufacturing is becoming more and more important. Large-scale synthesis relies heavily on high volumes of concentrated Sulfuric Acid, leading to elevated economic expenditures and acidic effluent streams that must be treated before being discharged. The environmental burden of the acid production, transportation, storage, and neutralization emphasizes the need for 'greener' synthesis-approaches. The principles of Green Chemistry drive the minimization of toxic chemical utilization, reduction in waste generation and increase in the efficiency of resources. In order to apply these principles to the synthesis of Graphene Oxide, innovative modifications are needed to reduce the amount of chemicals used, while maintaining the oxidation efficiency. Sulfuric Acid is one of the main contributors to hazardous environmental impact and also the processing costs from among the different components involved in the Improved Hummers Method [17].

Here we propose a reduced-volume Sulfuric Acid strategy for Graphene Oxide synthesis based on the Improved Hummers Method. We propose that fine-tuning the chemical environment and effective oxidation can be achieved with substantially lower volumes of Sulfuric Acid. Such an approach could offer strong potential for preserving material fidelity, while also reducing the environmental impact and improving the process sustainability. This

work focuses on the evaluation of the possibility of a reduced-acid synthesis, the elucidation of the oxidative pathways under altered reaction parameters and the discussion of the implications of the approach for future industrial-scale production of Graphene Oxide.

Materials and Methods

Materials

Graphite flakes were chosen as the Carbon-source since they are highly pure and have a layered crystal structure. Potassium Permanganate (KMnO_4) ($\geq 99\%$) was chosen as the main oxidant for Graphene Oxide production. Sulfuric Acid (H_2SO_4) (95%) and Phosphoric Acid (H_3PO_4) (85wt%) were used to make the mixed-acid solution. Hydrogen Peroxide (H_2O_2) (30wt%) was applied for oxidation-termination and the elimination of Manganese. Hydrochloric Acid (HCl) (37wt%) and distilled water were used for the purification of Graphene Oxide from the reaction medium. All the above-mentioned reagents, which were of analytical grade, were purchased from Sigma Aldrich Chemicals Pvt Ltd., Bangalore. Also, throughout the entire experiment, double-distilled water was made use of.

Reduced-Acid Oxidation Process

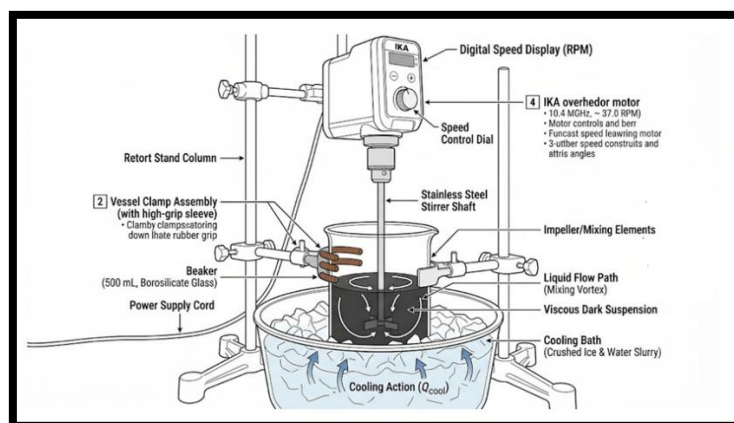


Fig.1 - Schematic picture GO preparation

Graphite flake dispersion was carried out using a Sulfuric-Phosphoric mixed-acids solution in a ratio of 10 : 1 ml (V/V) and kept at stirring for 30 to 60 minutes for uniform distribution of Graphene in the reaction medium. It was then transferred to an ice bath while still being stirred. KMnO_4 was added in small portions and extra care was taken so that any possible explosion or inflammation into fire gets eliminated. The addition of KMnO_4 was continued till the ratio of Graphite to KMnO_4 reached nearly 1:3. The volume of Sulfuric Acid used here was

significantly less than that of the traditional method, but without compromising the oxidant-to-Graphite ratio. Then the reaction mixture was allowed to attain room temperature. Subsequently the reaction mixture was continuously stirred at 60 °C for 2 hours.

During the reaction process, high temperature was maintained to ensure that adequate time was available for oxidation and layer expansion to occur. At this point, Oxygen groups incorporated themselves into the Carbon structure, thus increasing the interlayer distance and loosening the interplanar connections. The stirring was continued for 6-8 hours in order to ensure complete uniform oxidation. The stirring helped with the even distribution of the oxidation species throughout the mixture, even as it facilitated the insertion into the Graphite interlayers. The completion of the Oxidation Reaction was checked and verified by the addition of a small amount of Potassium Permanganate. The reaction medium was then doubled with distilled water with the help of an ice bath till the reaction mixture attained room temperature. It was then kept at stirring for a further 16-24 hours. After the Oxidation was complete, Hydrogen Peroxide was added to stop the reaction and to dissolve all the remaining Manganese salts. Then, the required amount of H_2O_2 solution was added for the termination of the oxidation. After that, washing was conducted using 1:10 HCl solution till pure Graphene Oxide was obtained. Also, washing and centrifuging of the mixture was conducted until neutral pH was reached. Further, the exfoliation of the Graphene Oxide product was accomplished using Ultrasonication before vacuum drying was done to produce Graphene Oxide powder.

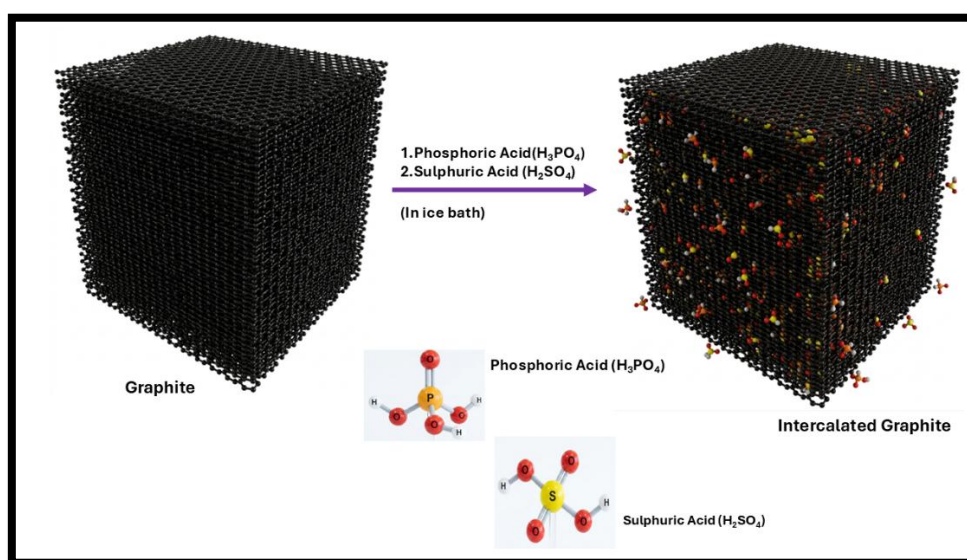


Fig. 2 - Intercalation of Graphite

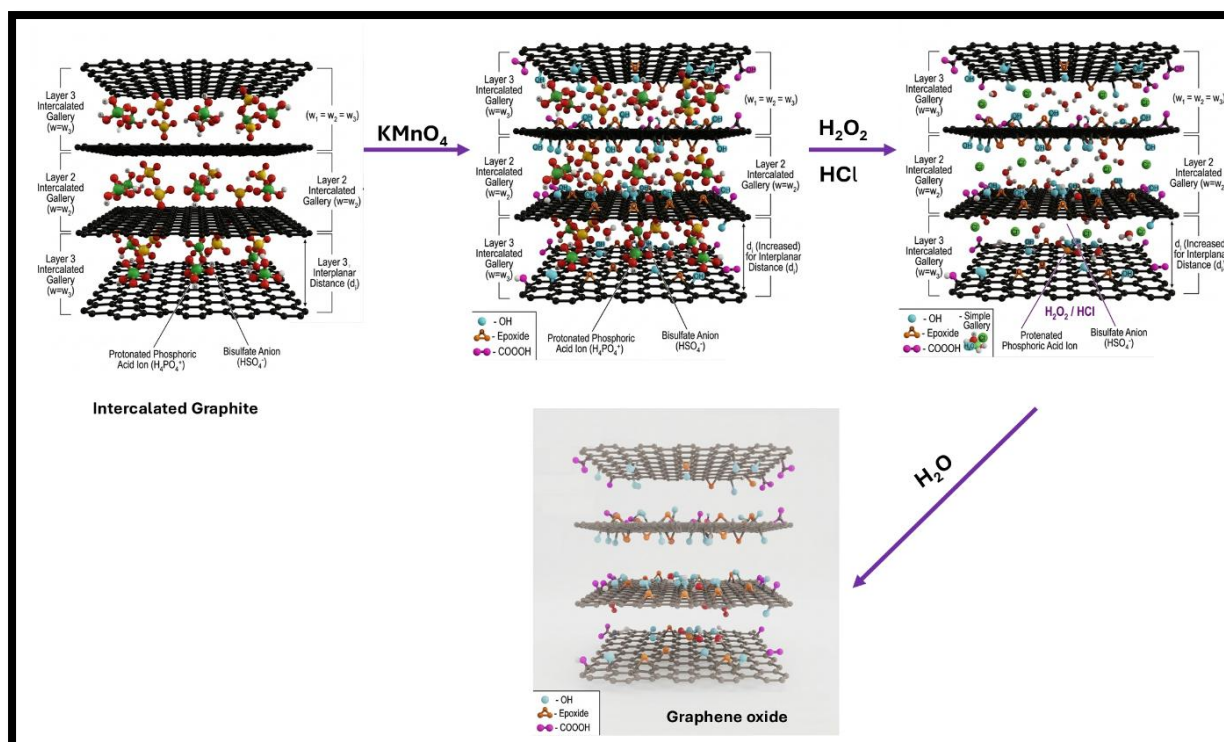


Fig. 3 - Oxidation of Intercalated Graphite into Graphene Oxide

Characterization of synthesized GO samples

X-ray diffraction (XRD) patterns of the synthesised GO samples were measured with a MiniFlex II Benchtop XRD, X-Ray diffract meter using Cu-K α radiation source ($\lambda = 1.54184$ nm). Field emission scanning electron microscopy (SEM- Jeol 6390LA/ OXFORD XMN N [Accelerating voltage: 0.5 to 30 Kv, Filament: Tungsten]) were used to study the morphology of the prepared GO samples. Conofocal Raman Microscope with AFM (Spectral range 0 to 4000 cm^{-1}), Spectral Resolution 600gr/mm grating 5cm-1, Laser wavelength 532 nm, number of scans 10 (Witech alpha 300RA model) was the instrument used to record the Raman Spectrum in order to measure lattice defects. Fourier transform infrared (FT-IR) spectra were measured using a FT-IR spectrometer SHIMADZU IR Prestige-21 (Crystal – ZnSe) Scan Range: 600-4000 cm^{-1} . UV – Visible spectra were collected at 20 °C in the wavelength range of 190 to 1100 nm nm using a Shimadzu UV-1800 spectrophotometer.

Results and Discussions

The proper fabrication of Graphene Oxide (GO) was assessed by various structural, chemical, morphological, optical, and thermal characterization techniques, which include XRD and FTIR evaluations; the evidence of Graphite oxidation and GO formation was evaluated by Raman Spectrum where it shows the extent of damages in the crystalline structure of Graphite sheet in highly concentrated acidic oxidation environment. UV-VIS spectroscopy was used to investigate the optical properties resulting from the digital structure. Morphologies, layer structure, thickness, elemental distribution and the size and distribution of the synthesized GO sheets were examined by SEM and EDAX. All the results attained confirmed that Graphite was successfully transformed into Graphene Oxide.

XRD Analysis

X-ray Diffraction (XRD) Analysis confirmed the successful oxidation of Graphite to Graphene Oxide using The MiniFlex II Benchtop XRD System. Fig. 4 represents the XRD plot of Graphene Oxide and Graphite which clearly explains the crystallinity, interlayer spacing, crystal structure, and peak shift; the lattice strain graphene shows a sharp reflection from the plane (002) with 2θ value of 26.5° having an inter-layer spacing of approximately 0.34 nm. Controlled oxidation of Graphite results in the formation of Graphene Oxide, with the disappearance of the above reflection and the appearance of a reflection from a new plane (001) that is having a 2θ value of 10.7° and having an inter-layer spacing of approximately 0.82 nm due to the presence of Oxygen-containing functional groups and water molecules. The Peak Width of the new peak gives an idea about the crystalline size and the extent of disorder introduced into the crystallinity of the existing lattice which came about during the oxidation of Graphite to Graphene Oxide [19-21].

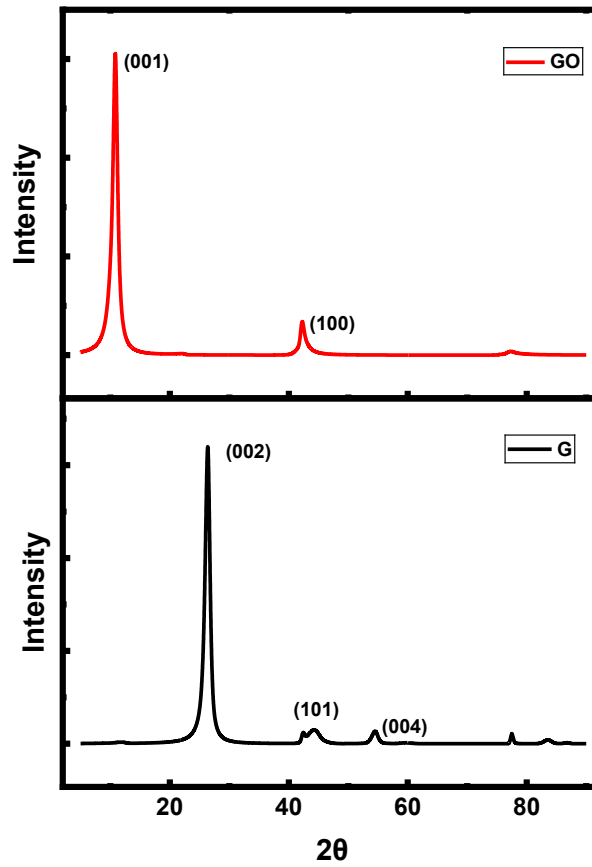


Fig. 4 – XRD plot of Graphite (-G), and Graphene Oxide(-GO)

It is by using Bragg's Law that the Interlayer Spacing (d-spacing) between atomic planes can be calculated:

$$d = \frac{\lambda}{2 \sin \theta}$$

Here λ represents X-ray wavelength.

The crystallite size of GO is calculated from X-ray diffraction (XRD) data using the Scherrer Equation:

$$D = \frac{K\lambda}{\beta \cos \theta}$$

where D = crystallite size (in nanometres or angstroms), K = Shape factor [dimensionless constant, usually close to (0.9 or 0.94) for spherical crystallites], λ = X-ray wavelength [e.g., 1.5406 Å for Cu-K α radiation], β = Line broadening at half the maximum intensity (FWHM), which must be converted into radians[21-22].

Table I- Structural characteristics of Graphene Oxide determined from XRD analysis

Calculated parameters	Formula	Graphene Oxide
Interlayer Spacing (d-spacing)	<i>Bragg's Law:</i> $d = \frac{\lambda}{2 \sin \theta}$	0.82 nm
Crystallite Size (D)	<i>Scherrer Equation:</i> $D = \frac{K\lambda}{\beta \cos \theta}$	7.39nm
Average number of layers	$N = \frac{D}{d} + 1$	10

A value of 7.4 nm means that Graphene Oxide sheets are stacked in domains that average 7.4 nm in thickness. It is the ratio of 'Crystallite Size (D)' to 'd spacing' that gives an idea about the number of layers;

Average number of layers

$$N = \frac{D}{d} + 1;$$

$$\text{If } D \gg d \text{ then } N \approx \frac{D}{d}$$

Adding '1' accounts for the fact that a stacking thickness containing '1' gap actually consists of '2' solid layers. In this case the Average number of layers is ≈ 10 (Ten sheets stacked per Crystallite). This confirms that the material consists of few-layered Graphene Oxide structures rather than thick, unexfoliated Graphite (Table I).

FTIR Analysis

For the detection of chemical structure and the presence of Oxygen and other functional groups FTIR can be used as a powerful tool. SHIMADZU IR Prestige-21 (Crystal – ZnSe) Scan Range: 600-4000 cm^{-1} was used for the FTIR analysis. As we observe the FTIR spectrum of Graphite (-G) and that of Graphene Oxide (-GO) we can clearly see that the oxidation of Graphite to Graphene Oxide resulted in the formation of new -CO bonds; this is shown in Fig. 5. For Graphite, a peak was observed at 1516 cm^{-1} due to the aromatic C = C stretching vibrations of conjugated sp^2 Carbon network. Also, the peak at 1225.34 cm^{-1} was due to the small amount C-O-C (epoxy) stretching vibrations. The above peak is attributable to minor oxidation in Graphite, which may be the result of slight oxidation of Graphite during storage or handling as shown in Fig.5.

Again, a comparative evaluation demonstrates that Graphene Oxide shows quite a number of peaks than what is apparent in Graphite; GO's peak at 3366.39 cm^{-1} is due to the presence of O-H Stretching vibrations, the peak at 1728.12 cm^{-1} is due to C = O stretching vibrations, the peak at 1617.59 cm^{-1} is due to C = C skeletal stretching of sp^2 carbon vibrations and H-O-H bending vibrations of adsorbed water. Further, the peak at 1351 cm^{-1} is due to C-OH bending vibrations, and the peak at 1063.02 cm^{-1} is due to C-O stretching vibration of alkoxy group, and the low-frequency peak at 708.37 cm^{-1} is associated with C-O out-of-plane deformation or skeletal vibrations of oxidized Graphene framework [21-22].

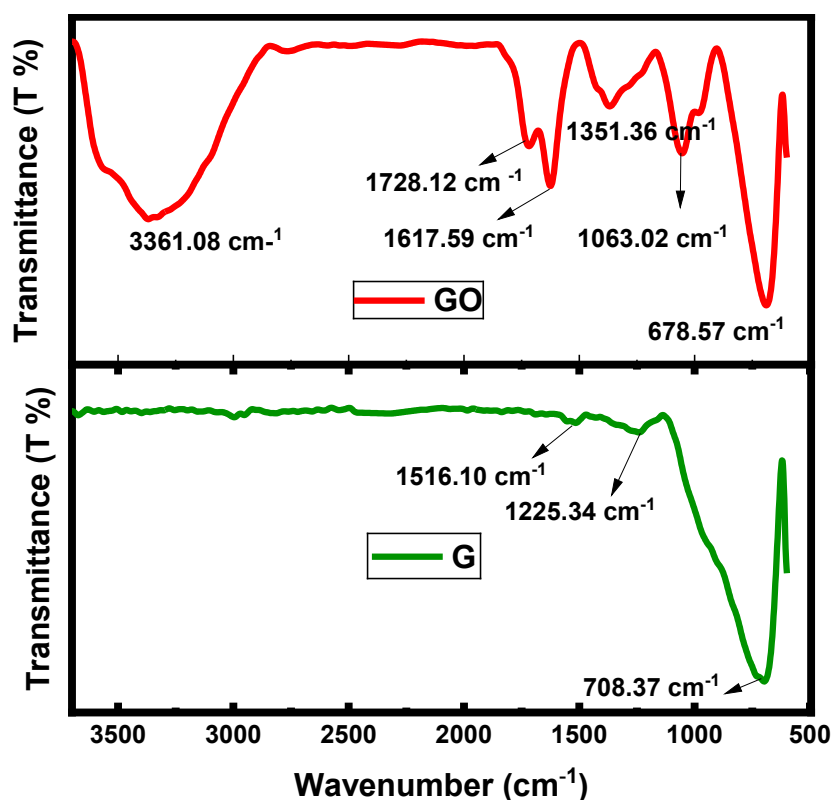


Fig. 5 – FTIR plot of Graphite (-G), and Graphene Oxide(-GO)

Raman Analysis

In graphitized Carbon-based materials we can observe the coexistence of D-peaks, G peaks, their multiplicity peaks, and conjugated C - C bonds which lead to high Raman intensities. Confocal Raman Microscope with AFM (Spectral range 0 to 4000 cm^{-1}), Spectral Resolution 600 gr/mm grating 5 cm^{-1} , Laser wavelength 532 nm , number of scans 10 (Witech alpha 300RA model) was the instrument used to record the Raman Spectrum. As shown in Fig.6, Raman Spectrum exhibits characteristic bands located approximately at 1346.91 cm^{-1} (D band) and 1594.61

cm^{-1} (G band); where the G band corresponds to the first order E_{2g} vibrational model for sp^2 hybridized Carbon atoms and the D band corresponds to A_{1g} vibrational mode; both of them are activated by defects in lattice edges and due to the presence of functional groups.

The appearance and intensification of the D band correspond to the presence of Oxygen (in the form of epoxy, carbonyl, hydroxy and carboxyl) introduced by the oxidation of Graphite and the resultant structural defects in the lattice. This oxidation disturbs the sp^2 hybridized Carbon, resulting in the fragmentation of existing hexagonal graphitic stacking to smaller aromatic graphitic domains with higher degree of disorder. The total density of defects thus increased can be measured by calculating I_D/I_G ($I_D/I_G = 0.98$) ratio of oxidized species (GO) and comparing with the I_D/I_G of Graphite. Here in Fig. 6 we can see the coexistence of pronounced D and G bands which confirms the successful conversion of Graphite to Graphene Oxide with the retention of fundamental hexagonal Carbon-framework structure. For Graphene Oxide, the 2D (2071.05 cm^{-1} and $I_{2D}/I_G = 0.89$) band is weak when compared to Graphite due to extensive oxidation, increased defect density, reduced graphitic crystallinity and multiple or stacked graphene oxide layers. Low I_{2D}/I_G ($I_{2D}/I_G < 5$) indicates multiple layers of Graphene Oxide - that is, few-layered Graphene [14], [23].

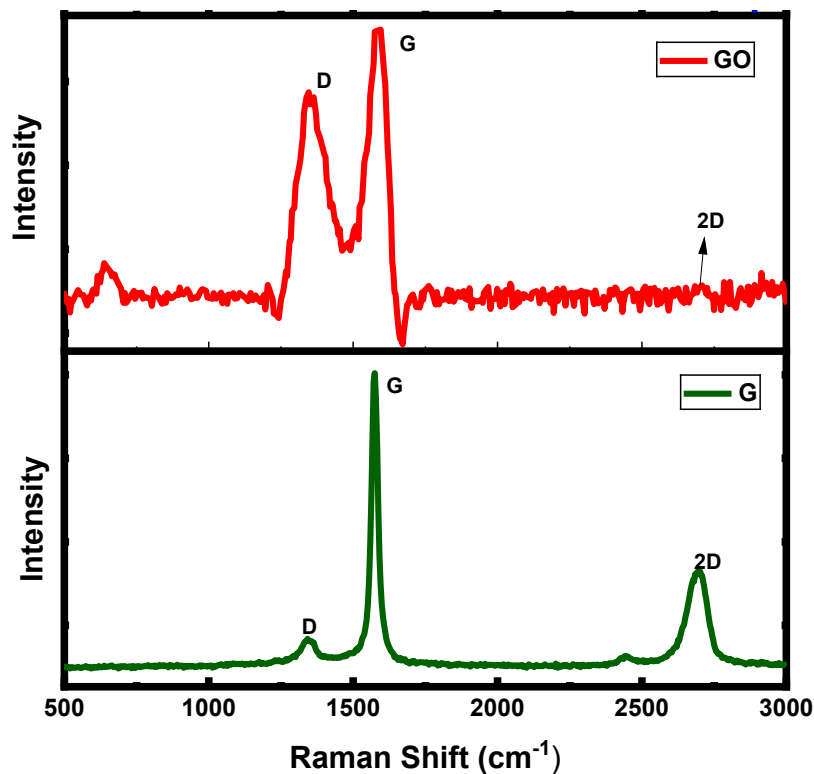


Fig. 6 – Raman Plot of Graphite (-G) and Graphene oxide (-GO)

UV-Vis analysis

UV absorption spectra were recorded using Shimadzu 1800 UV Spectrometer. An Absorption Maximum ($\lambda_{\max}$) at ~ 236 nm was observed: This prominent characteristic peak corresponds to the $\pi \rightarrow \pi^*$ electronic transitions of aromatic C=C bonds within the surviving sp^2 Carbon hybrid domains of the graphitic framework. A Shoulder / Extended Absorption Tail (~ 302 nm) was also observed: The broad tail extending toward the visible region is attributed to the $n \rightarrow \pi^*$ electronic transitions of Carbonyl (C=O) and other Oxygen-containing functional groups[23-26].

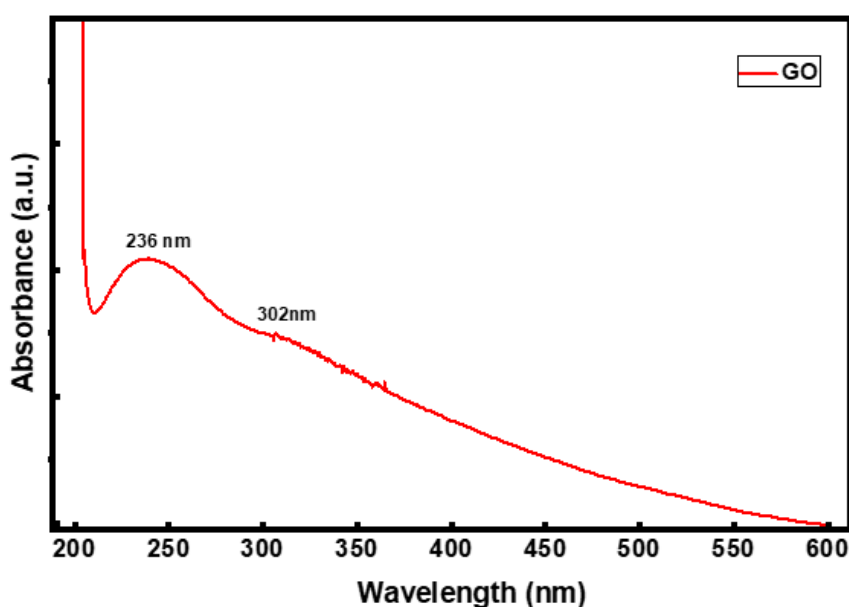


Fig. 7 – UV plot of Graphene Oxide (GO)

SEM-EDAX Analysis

The synthesized Graphene Oxide (GO) was characterized by Scanning Electron Microscopy (SEM- Jeol 6390LA/ OXFORD XMN N [Accelerating voltage: 0.5 to 30 Kv, Filament: Tungsten]) at different magnifications (Fig. 7) to study the morphology of GO. The SEM image of low magnification ($500\times$) shows a heterogeneous collection of irregularly shaped agglomerates scattered throughout the sample. Fig. 7 reveals that the morphology of the particles is loosely packed with several voids in between the particles; this implies the formation of porous aggregates due to strong Van der Waals interactions and Hydrogen bonding between Oxygen-containing functional groups during the drying process.

At a higher magnification (10,000 $\times$), the microstructure of GO sheets appears as thin crumpled lamellar structures due to the overlapping and folded nanosheets forming a three-dimensional network; this arises because of oxidation-induced disassembly of the sp^2 Carbon lattice, together with the insertion of Oxygen-containing functional groups causing enhanced lattice distortion. Thus, a successful exfoliation of Graphite to produce few-layered Graphene Oxide sheets is confirmed by the curled edges of the sheets and the multilayer stacking, along with a partial restacking which occurs during solvent evaporation. This leaves an accessible surface area for further functionalization or composite formation. Moreover, the interconnected porous structure observed in the low-magnification image indicates the presence of diffusion pathways that may improve adsorption, catalytic activity, or electrochemical performance depending on the applications and further functionalization processes.

In general, the SEM observations confirm the typical morphology of Graphene Oxide in the synthesized material with crumpled, layered and wrinkled nanosheets with partial agglomeration due to the successful oxidation and exfoliation of Graphite; these are found to be in good agreement with previously reported morphologies of chemically synthesized Graphene Oxide[21].

EDAX analysis was performed to determine the elemental composition of the synthesized Graphene Oxide (GO); it was revealed that Carbon and Oxygen are the predominant elements, 55.79 wt % (62.87 at %) and 43.59 wt % (36.87 at %) respectively (Table II). The high Oxygen content confirms the successful oxidation of Graphite through the introduction of Oxygen-containing functional groups during synthesis. A C/O atomic ratio of approximately 1.70, which is characteristic of highly oxidized Graphene Oxide, indicates the extensive oxidation of the graphitic framework. A trace Sulphur content (0.62 wt %; 0.26 at %) was also detected; this can be attributed to a residual Sulphate species from the oxidation process. The low Sulphur concentration suggests effective purification of the product[26].

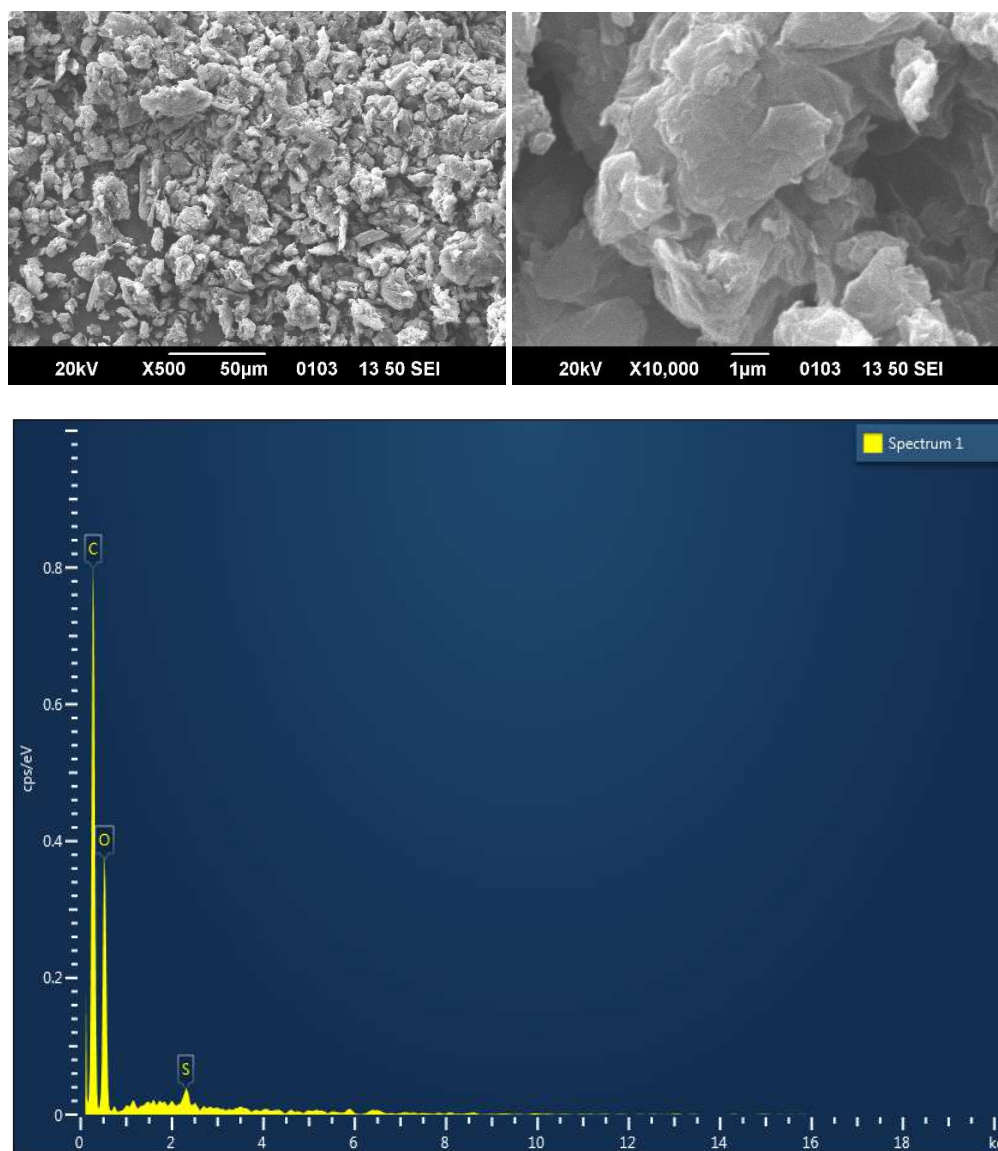


Fig. 8 – FTIR plot of Graphite (-G), and Graphene Oxide (-GO)

Table II- EDAX analysis of GO

Spectrum 1			
Element	Line Type	Wt%	Atomic %
C	K series	55.79	62.87
O	K series	43.59	36.87
S	K series	0.62	0.26
Total:		100	100

Stoichiometry Evaluation, Structural physio-chemical assessment and Sustainability-Analysis of Graphene Oxide synthesis for process-efficiency

The Modified Hummers method was systematically evaluated for the synthesis of Graphene Oxide (GO) using a comprehensive suite of quantitative stoichiometric process, structural physicochemical assessment and sustainability parameters for Process-efficiency. Firstly, the effect of reagent composition on the oxidation process was investigated and calculated by means of the stoichiometric analysis based on the graphite-to-oxidant molar ratio (4.39:1), acid concentration and acid-to-graphite ratio (volume or moles of acid/mass or moles of Graphite = 10). The efficiency of the process was evaluated by the yield of GO, mass expansion, Carbon recovery, oxidation efficiency and oxidant utilization. The resultant amount of GO when compared to that of the initial amount of Graphite used showed a 189% increase in yield and a 89.52% expansion in mass; this reveals a doubling of the amount of product obtained. Secondly, the structural and physio-chemical properties of the synthesized GO were explored by XRD, Raman spectroscopy and FTIR analyzes; here parameters such as interlayer spacing, crystallite size, defect density, degree of oxidation and functional-group distribution were determined. From FTIR, the spectra oxidation index was calculated and found to be 0.78; by the use Raman Spectrum, the (I_D/I_G) ratio was determined and it directly indicated the structural disorder and defect density, which was calculated and found to be 0.98 with a 149.3% increase in interlayer spacing within the crystal lattice. Thirdly, the sustainability of the route was assessed by calculating the decrease of reagents, water and solvent consumption, etc; here the synthesis of GO was achieved by maintaining the acid concentration and acid-to-graphite ratio (volume or moles of acid /mass or moles of Graphite) as 10. This helped to drastically reduce the amount of Hydrochloric Acid required during the washing of GO to remove biproducts like Sulphate ions and Manganese Oxide. These integrated calculations provide a quantitative perspective to evaluate whether the reduced acid and oxidant consumption can be achieved without sacrificing the structural and physicochemical quality of the final GO product (Table III).

Table III- structural physio-chemical assessment and sustainability-analysis of GO

Category	Evaluation criterion	Definition / Formula	Result	Analytical significance
	Graphite:KMnO ₄ molar ratio	$n_{\text{graphite}}:n_{\text{KMnO}_4}$ n- number of moles	4.39:1	Amount of oxidant used relative to graphite
Reagent & process efficiency	Oxidant-to-graphite ratio	$R = \frac{n(\text{KMnO}_4)}{n(\text{C})}$ N= number of moles of KMnO ₄ /C[graphite]	0.228	Oxidant requirement per mole of carbon

	Acid-to-graphite ratio	$\frac{V_{\text{acid}}}{m_{\text{graphite}}}$ or $\frac{n_{\text{acid}}}{n_{\text{graphite}}}$ V= volume of acid used, m=mass of graphite	10	Amount of acid used per unit graphite
	Acid reduction (%)	$\frac{\text{conventional amount} - \text{modified amount}}{\text{conventional amount}} \times 100$	91.6%	Reduction in acid consumption
Yield & mass efficiency	GO yield (%)	$\frac{m_{\text{dried GO}}}{m_{\text{initial graphite}}} \times 100$ m = mass of GO/Graphite	189.52%	Overall mass yield of dried GO [27]
	Mass increase (%)	$\frac{m_{\text{GO}} - m_{\text{graphite}}}{m_{\text{graphite}}} \times 100$ m = mass of GO/Graphite	89.5	Mass gained due to oxygen-containing groups/intercalation
Degree of oxidation	Oxidation efficiency (%)	$\frac{\text{actual O incorporated}}{\text{theoretical O incorporated}} \times 100$	FTIR oxidation index of 0.78	Efficiency of oxygen incorporation
	FTIR oxidation index (OI)	$OI_{\text{FTIR}} = \frac{A_{\text{O-containing bands}}}{A_{\text{C=C}}}$ A= absorbance of O-containing bands/absorbance of C=C	FTIR oxidation index of 0.78 [28]	Relative degree of oxidation; higher OI = greater oxidation
	Raman defect density	I_D/I_G I_D = intensity of D band I_G = intensity of G band	0.98	Degree of defects/disorder introduced during oxidation
Structural characteristics	Interlayer spacing	d_{hkl} from XRD	0.82	Expansion of graphite layers after oxidation
	Increase in interlayer spacing (%)	$\frac{d_{\text{GO}} - d_{\text{graphite}}}{d_{\text{graphite}}} \times 100$ d = interlayer spacing of GO or Graphite	141%	Extent of layer expansion

Conclusion

In this study, a sustainable acid-reduced modification approach of the Hummers Method for the synthesis of Graphene Oxide (GO), provides a safer and more environmentally benign alternative to the existing method by reducing the consumption of concentrated acids along with the maintaining of efficient oxidation, the minimizing of hazardous chemical usage along with decreased waste generation, and the improvement of the overall sustainability during the synthesis process without compromising the product quality as demanded by the market.

The GO thus synthesized, exhibited the characteristic structural and morphological features expected for oxidized Graphene. SEM analysis showed crumpled, and layered nanosheets with partial agglomeration (due to weak bonds of attractions), resultant due to successful exfoliation of the graphitic precursor. EDAX analysis confirmed Carbon and Oxygen as the dominant constituent elements, with a C/O atomic ratio of approximately 1.70, showing increased oxidation, with trace Sulphur impurities detection, exhibit effective purification. The FTIR spectrum

explicitly confirms the rich presence of active Oxygen functionalities through characteristic stretching and bending vibrations for O–H (3366.39 cm^{-1}), C=O (1728.12 cm^{-1}), C–OH (1351 cm^{-1}), and alkoxy C–O (1063.02 cm^{-1}). This is further corroborated by the characteristic electronic absorption peak at 230 nm in the UV-Vis analysis, indicative of $\pi \rightarrow \pi^*$ transitions of aromatic C=C bonds and $n \rightarrow \pi^*$ transitions of Carbonyl groups. The coexistence of prominent D-band (1346.91 cm^{-1}) and G-band (1594.61 cm^{-1}) with an intensity ratio (I_D/I_G) of 0.98 confirms a high degree of oxidation while successfully retaining the fundamental hexagonal Carbon framework. Furthermore, the weak 2D band (2071.05 cm^{-1} , with $I_{2D}/I_G = 0.89$ and < 5) confirms the successful creation of few-layered Graphene Oxide sheets. The convergence of XRD, FTIR, Raman, SEM-EDAX, and UV-Vis data provides indisputable evidence that the synthesized material is few-layer Graphene Oxide of high oxidation quality, structurally primed for advanced functionalization, composites, and electrochemical applications.

The proposed method offers several advantages over the Conventional Hummers Method, including reduced acid consumption, decreased environmental footprint, improved operational safety, and the potential for cost-effective scale-up; to these are aligned the principles of Green Chemistry that support the development of more sustainable manufacturing processes for Graphene-based materials.

In short, this work shows that the acid-reduced route as suggested above can produce high-quality Graphene Oxide along with reduction in ‘hazard-ness’ of the environmental and safety issues that come with conventional methods. Future research can aim to improve the oxidation conditions, tunability of the extent of oxidation, check the scaled-up production, and a study of how the resulting Graphene Oxide performs in areas like energy storage, environmental cleanup, catalysis, sensors, and polymer nanocomposites. In short, this method offers a practical way to sustainably produce Graphene Oxide on a large scale for various technical applications.

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Author’s Contributions

All authors contributed to the conception and design of the study. Sona Xavier conducted the literature survey, prepared initial manuscript, and contributed to data collection and Snehamol K Shaji led subsequent revisions. Both together involved in synthesis, Characterization, figure preparation and manuscript editing. Reni George, provided supervision, critical review, and final editing. All authors reviewed and approved the final manuscript.

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Competing interests

The authors declare that there are no competing interests regarding the publishing of this paper

Data availability.

The data supporting the findings of this study are available from the corresponding author upon reasonable request.

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