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Article

Oxygen Functional Groups in Graphene Oxide Using Titration Methods: Quantitative Analysis and New Quality Parameters

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Abstract: Graphene oxide (GO), an oxidized form of graphene containing various oxygen functional groups, is recognized for its exceptional properties and one of the most valuable graphene-related 2D materials (GR2Ms). Large-scale industrial production of GO materials from graphite involves various chemical oxidation methods, leading to significant variability in their properties, structures, types and composition of oxygen functional groups, which are critical for their practical applications. The quantification of oxygen functional groups in industrially manufactured GO remains largely unexplored and undisclosed in technical data sheets, creating challenges for end-users. Conventional characterization techniques for graphene materials, including SEM, EDAX and XPS, are limited by their spot-characterization nature and inability to reliably assess the bulk chemical properties of GO materials. To address these challenges, in this paper, we present a demonstration of a simple and industrially affordable analytical method using potentiometric titration to quantify the concentration of oxygen functional groups in GO powders and pastes on a bulk scale. Specifically, Boehm and acid-catalysed titrations were combined and successfully employed to determine the concentrations (mmol/g and mass %) of carboxylic, lactone, hydroxyl, carbonyl, epoxy groups and the total oxygen groups. This method has been validated by quantifying oxygen functional groups in industrially GO samples from three different manufacturers. The results revealed substantial differences in the concentrations of oxygen functional groups and total oxygen level of these GO samples, with carboxylic acid groups ranging from 0.89 ± 0.01 to 1.91 ± 0.08 mmol/g, lactone groups from 0.20 ± 0.01 to 1.76 ± 0.26 mmol/g, phenolic groups from 1.12 ± 0.15 to 2.73 ± 0.05 mmol/g, carbonyl groups from 0.65 ± 0.19 to 2.21 ± 0.26 mmol/g, and epoxy groups from 1.15 ± 0.05 to 1.37 ± 0.05 mmol/g. These variations, likely stemming from different GO manufacturing processes, highlight the importance of accurately determining these parameters. Furthermore, based on these measurements, we introduce, for the first time oxygen group indexes (OGI) as a novel quality parameter for distinguishing the quality of industrially produced GO materials. This study demonstrates how these simple, cost-effective methods, when implemented and adopted can significantly contribute to the chemical characterization and quality control of GR2Ms, addressing a critical gap in the graphene industry.

Keywords: graphene oxide; functional groups; Boehm; titration; characterization

1. Introduction

Graphene Oxide (GO), is an oxidised form of graphene and one of the most important members of graphene-related 2D materials (GR2Ms) that attracted tremendous attention due to its unique physicochemical properties and diverse range of applications including large-scale production of graphene [1–3]. Unlike pristine graphene, GO contains abundant oxygen functional groups, including carboxyl, hydroxyl, epoxy, carbonyl, and lactone groups, distributed across its basal plane and edges [4,5]. These functional groups endow GO with distinctive characteristics such as hydrophilicity, chemical reactivity, and ease of dispersion in aqueous and polar solvents, making it a versatile material for fields such as energy storage, water purification, biomedical applications, catalysis, and functional coatings [6–8]. Several different methods have been reported for preparing GO from graphite using different acids, oxidants, and conditions such as Brodie, Staudenmaier, Hofmann, Hummers, modified Hummers, Tour and others [9–12]. The formation mechanisms of different oxygen functional groups in GO and their precise structure remain uncertain due to the variability of oxidation routes, the non-stoichiometric nature of the oxidized products, and the limitations of current characterization techniques. Several contradictory models have been proposed to elucidate the structural and chemical features of GO with different oxygen groups, including Hofmann−Holst, Ruess, Scholz−Boehm, Nakajima−Matsuo, Lerf−Klinowski, and Szabó−Dékány [13–16]. As a commonly accepted model now, GO is presented as oxidised graphene with a roughly 2:1 C/O ratio and a random distribution of flat aromatic regions with unoxidized benzene rings and wrinkled oxidised regions containing different functional groups as schematically presented in Figure 1. The unoxidized regions at the basal plane retain graphene-like properties with sp^2 carbon, while the oxidized regions are randomly decorated by hydroxyl and epoxy groups linked to sp^3 carbon up and down because of the hydrogen bonds and steric hindrances [17]. The edge of GO is mostly composed of carboxyl and phenolic groups complemented by a small amount of carbonyl, quinone, and lactone groups [18]. However, there is no clear evidence about the spatial arrangement and the distribution relationship between oxygen functional groups along the graphene layer. To have this information and insight into the structure of GO materials and their oxygen functional groups is critically important for their further modification for diverse applications including energy storage, water purification, environmental remediation, biomedical applications, catalysis, composites, sensors and functional and protective coatings.

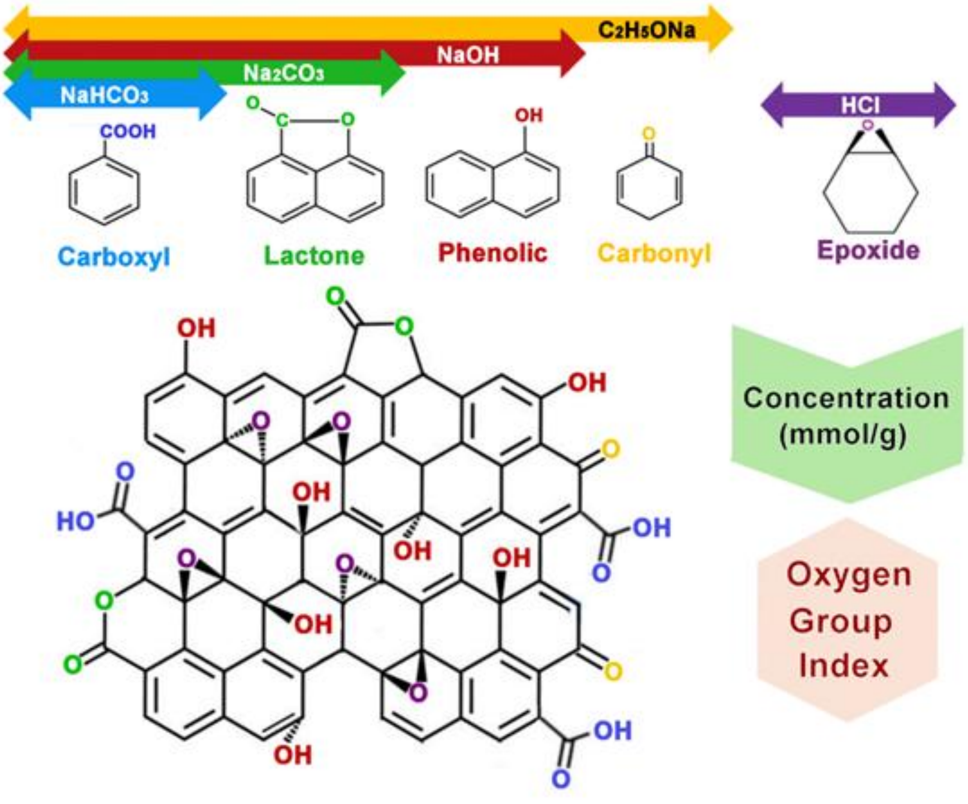

Figure 1. Functional groups in GO and respective titrants (bases) used for titration for their quantitative determination (mmol/g or mass %) and their quality standardization using oxygen group indexes.

Globally, GO is currently produced at a scale of several thousand tons per year. However, even the largest GO manufacturers do not provide fundamental data on the chemical composition of their products, including the oxygen functional group content, in their technical specifications. The main reason for that is lacking access of analytical methods for chemical characterization, usually performed by expensive and specific XPS technique, which is not accessible for industry. This critical lack of essential chemical characterization poses a major barrier to the commercial adoption of GO, restricting end-users from fully leveraging the potential of this exceptional graphene-related material. Accurate disclosure of the chemical properties of GO, particularly the percentage and type of oxygen functional groups, is critical for understanding the reactivity of the material and enables users to tailor GO to specific applications, including the development of composites, membranes, coatings, additives, and functionalized graphene materials. Additionally, oxygen-containing functional groups significantly influence the thermal stability and energetic properties of GO materials. The GO energetic nature makes it prone to explosion and safety risks at elevated temperatures, highlighting the importance of chemical characterization for safe handling, transport, storage, and use. Furthermore, understanding the oxygen group composition that promotes the degradability of GO materials and their products is essential for their biomedical and environmental applications, improving their biocompatibility and contributing to the responsible and sustainable development of graphene technologies. Unfortunately, this information is not provided by GO manufacturers due to the absence of established characterization and quality control methods, making it challenging to ensure the consistent quality and performance of GO materials in the market. Existing characterization techniques for oxygen functional groups, such as X-ray photoelectron spectroscopy (XPS) and Energy-dispersive X-ray spectroscopy (EDS) are surface analysis methods that provide valuable qualitative or semi-quantitative insights. However, they are often limited by high costs, lack of scalability, and the focus on single graphene particles rather than bulk material analysis. These limitations hinder the development of quality standards and the effective comparison of GO materials, particularly for industrial applications where bulk-scale analysis and consistency are essential.

To address these limitations, our team is developing and implementing a series of complementary methods for simple and low-cost characterization and quality control of GR2Ms by providing structural, composition and chemical characteristics at bulk or macro scale. These analytical methods, including thermal gravimetric analysis (TGA), titration, UV-VIS spectroscopy, Fourier-transform infrared spectroscopy (FTIR), and Raman spectroscopy, are able to provide this valuable analytical information about GR2Ms in the form of powders, pastes and dispersions, that can be easily adopted by graphene industry. In our recent publications, we demonstrated that the TGA method can effectively differentiate various types of GR2Ms in powder mixtures (GO, rGO, and few-layer graphene (FLG)), including distinguishing non-graphene carbon impurities from graphene. This technique enables the quantitative determination of their concentrations based on distinctive thermal decomposition features, such as the number of decomposition peaks, the temperature at the maximum decomposition rate (T_{max}), and the corresponding mass losses [19–23]. Another complementary method, based on Boehm acid titration (BT), has recently been demonstrated and adopted as an international standard (EIC TS 62607-6-13:2020). This method has proven to be a reliable analytical tool for characterizing the chemical composition of GR2Ms, including the content of oxygen functional groups [24].

The Boehm titration method dating from 1962, is adapted in the industry as a valuable method to provide absolute values of the concentration of oxygen functional groups that avoids the ambiguity and subjectivity of spectroscopic methods such as XPS, EDS and Electron energy loss spectroscopy (EELS) [25–27]. The method is successfully used for the quantification of oxygen functional groups such as carboxyl groups (also in the form of their cyclic anhydrides), lactone groups, hydroxyl groups and reactive carbonyl groups [28–30] on graphite and other carbon materials such as biochar, carbon nanotubes, carbon blacks, and charcoals and recently implemented for GO and reduced graphene oxide [26,31–34]. During the titration process, titrant bases react with ionisable oxygen groups based on their pKa values/basicity, where it is proposed that sodium hydroxide (NaOH, pKa = 14.8) reacts with hydroxyl, lactonic and carboxylic groups while sodium carbonate (Na_2CO_3, pKa = 10.3) will react with hydroxyl, lactonic, sodium bicarbonate ($NaHCO_3$, pKa = 6.4) only with carboxylic groups and sodium ethoxide (C_2O_5ONa, pKa = 15.5), which is stronger than NaOH will react with carbonyl groups addition to the other three (Figure 1). However, the titration method has some limitations as it determines only ionisable oxygen groups and is not able to provide the content of epoxide level, which is one of the most important oxygen functional groups used for functionalization or cross-linking with other materials. Another disadvantage associated with this method is the potential accumulation of CO_2 during the long reaction time, which needs to be eliminated and considered with series of control experiments by using the prepared base solutions for the experiments as soon as possible to minimise reaction time. Epoxy groups cannot be determined by acid titration and to address this limitation, our recent work introduced a new method based on catalytic titration, which enables the quantitative analysis of epoxy groups in GO [32,35]. While the BT method is widely used in industry and recently standardised for quantification of oxygen groups in graphene materials, including GO, surprisingly, this method has not yet adopted in the graphene industry. As a result, GO materials produced in the industry and available on the global market often lack this crucial information, typically labelling their GO products as having 30–50 % oxygen content without specifying the types of functional groups or the exact amount of oxygen groups present in the sample.

The aim of this paper is to demonstrate how titration methods can be successfully used as a low-cost, simple complementary analytical method for the quantitative determination of oxygen functional groups in GO including carboxylic, lactone, hydroxyl, carbonyl, epoxy groups and the total oxygen groups. We combine, two methods including Boehm and catalytic titration for epoxy groups determination as quantitative analysis of oxygen functional groups in GO graphene materials from three world-leading producers. The results, presented in mmol/g and mass % concentration for each functional group as well as their total values, provide a comparative evaluation of these GO materials on the market revealing their differences resulting from various manufacturing processes. This contrasts with the common assumption that all GO materials are similar. Additionally, for the first time, we introduce a set of new quality parameters for functional group termed as Oxygen indexes that include Carboxylic Group Index (CxylGI), Phenolic Group Index (PhGI), and Carbonyl Group Index (CnylGI), Lactone Group Index

(LacGI), Epoxy Group index (EpGI)) and Total Oxygen Group Index (TotOGI). These indexes provide standardized quality values for easily quantifying the functional groups in GO materials, offering a practical tool for industry implementation. This method has the potential to be extended to graphene nanoplatelets, which typically have lower oxygen content than graphene oxide, broadening the applicability of this simple and cost-effective bulk characterization technique across different types of graphene materials [36]. This approach enables industry and end-users to meet the functional group requirements more easily for various applications by working directly with bulk materials from the market, streamlining the development process for graphene-based products and technologies.

2. Methodology

2.1. Chemical and Materials

Industrially produced GO samples in the form of powders labelled GO-1, GO-2, and GO-3 used for this evaluation study were sourced from three different leading GO manufacturers in China and Sri Lanka. Commercial GO samples were received in powder, cake and paste forms, respectively. Each commercial GO sample was initially prepared in powder form to ensure standardization and consistency. GO-1 was used as it is, while GO-2 and GO-3 were air-dried for 18 h and ground using a motor and pestle. Sodium Hydroxide[(NaOH), 98.00%, MW: 40.00 g/mol], Sodium Carbonate [(Na_2CO_3), 99.50%, MW: 84.01 g/mol], Sodium Bicarbonate [($NaHCO_3$), 99.70%, MW:105.99 g/mol], Sodium Ethoxide [(C_2H_5ONa) 95.00%, MW: 68.05 g/mol], were supplied from Sigma-Aldrich. Hydrochloric acid [(HCl), 36.50–32.00%, MW: 36.50 g/mol] from RCL Labscan, milli-Q ultrapure water (18.20 MΩ.cm) and Ethanol [(C_2H_5OH), 100% Absolute, MW: 46.06 g/mol] from Chem-supply, were used for the solution preparation and all the chemicals were used as it is unless otherwise stated.

2.2. Determination of Oxygen Functional Groups by Boehm Titration (BT)

The study employed the BT method to quantify the functional groups of three commercial GO samples located at the basal plane and edges adapted from the EIC standard with more details and schematic illustrations provided in Supporting Information [34].

Initially, stock solutions (1 L) of reaction bases (titrants) were prepared; 0.05 moldm^{-3} $NaHCO_3$, 0.05 moldm^{-3} Na_2CO_3, 0.05 moldm^{-3}, NaOH, 0.10 moldm^{-3} C_2H_5ONa and 0.05 moldm^{-3} HCl. To prepare the samples for titration, 50.00 g of NaOH, $NaHCO_3$, Na_2CO_3 and 40.00 g of C_2H_5ONa solutions were added into four separate High-Density Polyethylene (HDPE) bottles respectively, containing 350 mg of each GO material (three samples from each GO material).

Sample blanks were prepared by mixing 350 mg of each GO material with 50.00 g of Milli-Q ultrapure water, and solution blanks for each base solution were prepared with 40.00 g of C_2H_5ONa and 50.00 g of other three base solutions in HDPE bottles without GO materials. All prepared samples, solution blanks, and sample blanks were kept on a mechanical shaker at 200 rpm for 3 h at room temperature (26 ± 2 °C). After 3 h, samples with NaOH, Na_2CO_3, $NaHCO_3$ and sample blanks were filtered via suction-filtration with Whatman filter paper grade 1 (125 mm diameter) and samples with C_2H_5ONa separated by centrifuging at 4200 rpm for 15 min.

Filtrates were titrated with prepared HCl solution using an automatic potentiometer. For the titration, 10.00 g of titrate solutions of samples treated with NaOH, Na_2CO_3, and $NaHCO_3$ were mixed with 15 mL Mill-Q water. 15.00 g of filtered sample treated with C_2H_5ONa were collected and mixed with 20.00 mL of Mill-Q water before titration. Potentiometric titrations were performed using an automatic potentiometric titrator (916 Ti-Tech, Metrohm Co., Ltd., Herisau, Switzerland) equipped with a calibrated pH electrode (Metrohm Co., Ltd., Herisau, Switzerland) prior to each experiment. The volume of HCl consumed during each titration was recorded for calculations, and each experiment was carried out in triplicates. Average volumes were taken from the readings with less than a 10 % deviation between each run. Quality control measures were implemented to ensure the consistency and accuracy of the results, including the calibration procedures and reagent standardization with sample blanks and solution blanks.

Titration with Equivalent Point Determination and Functional Group Calculation

The endpoints were calculated from the titration curve using the first derivative of the pH-volume plot with respect to each titration where the first derivative gives the optimum point on the titration curve while the second derivative provides the certainty of the optimum [34].

$$\frac{dpH}{dV} = \frac{\Delta pH}{\Delta V} \tag{1}$$

Equations (2)–(6) were used to calculate the amount functional groups per gram of GO material based on the difference between the HCl volume consumed by the solution blank and the sample solution with respect to the mass of material used. The first derivative of the pH-volume plot corresponding to each titration gives the optimum point on the titration curve while the second derivative provides the certainty of the optimum [34]. Respective concentrations of ηA, ηB, ηC, ηD, and ηE were measured in mmol/g.

$$\eta A = (V_A * C/m_{A0,t} - V_{A1} * C/ m_{A1,t}) * m_{A1}/m_1 \tag{2}$$

$$\eta B = (V_B * C/m_{B0,t} - V_{B1} * C/m_{B1,t}) * m_{B1}/m_2 \tag{3}$$

$$\eta C = (V_C * C/m_{C0,t} - V_{C1} * C/m_{C1,t}) * m_{C1}/m_3 \tag{4}$$

$$\eta D = (V_D * C/m_{D0,t} - V_{D1} * C/m_{D1,t}) * m_{D1}/m_4 \tag{5}$$

$$\eta E = (V_E * C/ - V_{C0} * C/m_{C0,t} * m_{EC,t}/m_{E,t}{}^{)} * m_E/m_5 \tag{6}$$

- A = NaOH, B = Na_2CO_3, C = $NaHCO_3$, D = C_2H_5ONa, and E = Blank
- ηA, ηB, ηC, ηD = number of moles consumed by reaction with GO samples with respect to A, B, C and D bases in mmol/g
- ηE = Alkalinity of GO sample suspension, in mmol/g
- C = Concentration of HCl solution in mol/L
- mA, mB, mC, mD, mE = mass of the solution A, B, C, D and DI water in A1, B1, C1, D1 and E.
- V_A, V_{A1}; $m_{A0,t}$, $m_{A1,t}$ = the volume of the HCl consumed by titration A0 and A1 filtrate; the mass of A0 and A1 filtrate in grams
- V_B, V_{B1}; $m_{B0,t}$, $m_{B1,t}$ = the volume of the HCl consumed by titration B0 and B1 filtrate; the mass of B0 and A1 filtrate in grams
- V_C, V_{C1}; $m_{C0,t}$, m_{C1} =the volume of the HCl consumed by titration C0 and C1 filtrate; the mass of C0 and C1 filtrate in grams
- V_D, V_{D1}; $m_{D0,t}$, m_{D1} =the volume of the HCl consumed by titration D0 and D1 filtrate; the mass of D0 and D1 filtrate in grams
- m_1, m_2, m_3, m_4, m_5 =mass of dried GO material in A, B, C, and D reaction mixtures and E sample blank in grams

Concentrations of functional groups were calculated by substituting calculated respective mole amounts for each base reaction, and the number of carboxylic, lactone, carbonyl, and phenolic groups were obtained for each GO sample in mmol/g

$$\eta_{\text{carboxyl}} = \eta A + \eta E \tag{7}$$

$$\eta_{\text{lactone}} = \eta B + \eta E - \eta_{\text{carboxyl}} \tag{8}$$

$$\eta_{\text{hydroxyl}} = \eta C + \eta E - \eta_{\text{carboxyl}} - \eta_{\text{lactone}} \tag{9}$$

$$\eta_{\text{carbonyl}} = \eta D + \eta E - \eta_{\text{carboxyl}} - \eta_{\text{lactone}} - \eta_{\text{hydroxyl}} \tag{10}$$

- η_{carboxyl} = The content of surface carboxylic groups on graphene sample in mmol/g
- η_{lactone} = The content of surface lactone groups on graphene sample in mmol/g
- η_{hydroxyl} =The content of phenolic hydroxyl groups on graphene sample in mmol/g
- η_{carbonyl} =The content of surface reactive carbonyl groups on graphene sample in mmol/g

The concentrations of each functional group in mmol/g are converted to mass % using the following equation

$$\text{Mass \%} = ((C_{FG} * MW_{FG})/1) * 100\% \tag{11}$$

- C_{FG} = Concentration of Functional groups on Graphene Sample in mmol/g
- MW_{FG} = Molecular weight of the functional group in g/mol

2.3. Determination of Epoxide Functional Groups by Catalytic Titration Method

The titration method to determine the epoxide contents in GO was performed using our developed method which was recently published [37]. In a glass bottle, 50.00 g of $MgCl_2·6H_2O$ was mixed with 0.80 mL concentrated HCl and 15.00 mL milli-Q water. The mixture was vigorously shaken for a few minutes and kept for 2 h to settle the solution at ambient temperature (26 ± 2 °C). 10.00 mL of the above-mentioned solution was separated and mixed with precisely measured (50.00 mg) GO powder was shaken on an orbital shaker at 250 rpm for 2 h, and filtered through filter paper to obtain a clear solution that is used for potentiometric titration with 0.05 mol/L NaOH solution.

Calculation of Epoxide Concentration (mmol/g) of GO Materials

The volumes of 0.05 mol/L NaOH solution consumed during titrations were recorded and determined by changing the colour of the phenolphthalein indicator (colourless to pink) and confirmed by the first derivative of the pH-vol plot, where the first derivative (Equation (1) gives the optimum equivalent point of the titration curve [34].

Equation (12) was used to calculate the number of epoxide groups per gram of GO material based on the NaOH volume recorded at the endpoint of each titration.

$$\text{Epoxide Concentration} = ((V_{B1} + V_{B2} - V_S) * M)/m \qquad (12)$$

- V_{B1} = Volume of NaOH consumed by Blank 01 (Solution Blank)
- V_{B2} = Volume of NaOH consumed by Blank 02 (Sample Blank)
- Vs = Volume of NaOH consumed by the sample
- M = Concentration of NaOH solution
- m = Mass of the sample in grams

The concentration of the NaOH solution was determined based on the recorded readings in the titration with HCl solution (known concentration), which was calibrated using oven-dried anhydrous Na_2CO_3 at 150 °C for 4 h.

2.4. Determination of Total Oxygen Functional Groups

The concentrations of total oxygen functional groups in mmol/g were calculated by adding the concentrations of each functional group together, and mass % was calculated using the following Equation (13).

$$\text{Mass \%} = (\sum (C_{FG} * MW_{FG})/1) * 100\% \qquad (13)$$

- C_{FG} = Concentration of Functional groups on graphene sample in mmol/g
- MW_{FG} = Molecular Weight of the functional group in g/mol

2.5. Introducing Oxygen Group Indexes (OGIs) as New Quality Parameters for GO Materials

We introduced Oxygen Group Indexes (OGIs) for characterized GO materials as practical parameters that can be used to distinguish properties of different GO produced by different methods and have different concentrations of functionalized groups. This approach is adapted from international standards (ISO 3001:1999) typically used in the chemical industry to quantify critically important components in materials [38].

FGI is defined as the number of moles of functional groups contained in 1 kg of material that can be calculated by the following general equation (14):

$$\text{FGI} = \text{Moles of Functional Group per gram} * 1000 \qquad (14)$$

Five quality indexes, including carboxylic index, lactone index, phenol index, and carbonyl and epoxy index, were introduced for all the measured oxygen-containing functional groups of -COOH, COO-, -OH, and CO- with specific equations

- CxylGI = Moles of Carboxyl Groups per gram * 1000
- LacGI = Moles of Lactone Groups per gram * 1000
- PhGI = Moles of Phenol Groups per gram * 1000
- CnylGI = Moles of Carbonyl Groups per gram * 1000
- EpGI = Moles of Epoxide Groups per gram * 1000
- TotOGI = Moles of Oxygen Groups per gram * 1000

2.6. Other Characterization Methods

Three commercial GO materials used in the study (GO-1, GO-2, and GO-3) were comprehensively characterized before the study by a series of techniques, including XPS, TGA, XRD, FTIR, PSD, and Raman Spectroscopy, to confirm the identity and properties of the GO materials.

Scanning electron microscopy (SEM): The morphology of the GO was acquired by field emission scanning electron microscopy (FE-SEM, Quanta 450 FEG, FEI, USA) at an operating voltage of 10 kV.

Fourier-Transform Infrared Spectroscopy (FT-IR): Functional groups present in the GO samples were confirmed using Nicolet iS50 ATR-FTIR spectrometer (Nicolet, Thermo Fisher Scientific, USA) in the absorbance spectra by obtaining 32 scans with a resolution of 4 cm^{-1} in the range of 500–4000 cm^{-1}.

Ultraviolet-Vis spectrometer (UV-Vis): The detection of the absorption and conjugation network of GO was confirmed using Cary 60 (Agilent) by scanning the dilute GO dispersions containing in a quartz cuvette of 1 cm path length from 200 to 800 nm.

A powder X-ray diffractometer (XRD): 600 Miniflex, Rigaku, Japan, equipped with a Cu X-ray tube (λ = 1.54 Å, 40 kV and 15 mA) was run at a 10° min^{-1} scan speed in the range of 2θ = 5 to 80° to determine the interlayer spacing of the examined GO materials.

Raman spectrometer: LabRAM HR Evolution, Horiba Jvon Yvon Technology, Japan, with a 532 nm laser (mpc3000) was used to characterize the GO materials tested in this work. The Raman spectra were collected at 500 to 3000 cm^{-1} with an integration time of 10 s for three accumulations using a 100× objective lens, and the laser spot was 721.16 nm.

Thermogravimetric analysis (TGA): All samples in powder forms were conducted using a Mettler Toledo TGA/DSC 3+ instrument (heating rate at 10 °C/min under air atmosphere and a flow rate of 60 mL/ min).

X-ray photoelectron spectroscopy (XPS): The existence of impurities and the chemical composition of the materials were determined at 225 W, 15 kV, and 15 mA using an AXIS Ultra DLD, Kratos, UK, equipped with a monochromatic Al Kα radiation source (hv = 1486.7 eV). Peak fitting analysis was carried out using Casa XPS™ software on XPS survey spectra recorded at 0.5 eV step size over −10 to 1100 eV at 160 eV pass energy and high-resolution spectra acquired at 0.1 eV and pass energy of 20 eV with the primary peak of adventitious carbon calibrated at 284.8 eV.

3. Results and Discussion

3.1. Characterization of the Structural and Chemical Composition of GO Samples

Comprehensive characterization results of industry-produced GO materials used in this study by XPS, FTIR, Raman, XRD, and TGA are summarized in Figure 2. FTIR spectrum presented in Figure 2a presents three FTIR bands of GO materials, including O−H stretching at around 3400 cm^{-1}, C=O stretching at 1720 cm^{-1}, -OH bending at 1615 cm^{-1}, C-O-C and C-O alkoxy stretching vibrations at 1200–1278 cm^{-1}, and 1050 cm^{-1}, respectively, confirming the presence of typical oxygen functional groups in all three GO materials [39]. UV-vis spectra of the studied aqueous GO dispersions (Figure 2b) show two characteristic GO peak features in all three analyzed samples. A shoulder was detected at about 310 nm, corresponding to an n-π* transition of C=O, while a distinctive absorption peak arising at 230 nm due to the π-π* plasmon peak confirmed the identity of all the GO materials [40].

Raman spectra in Figure 2c show two typical D and G peaks of GO at around 1350 cm^{-1} and 1580 cm^{-1}, respectively, and the intensity ratio of D to G bands (I_D/I_G) was used to estimate the level of disorder present in the materials. The calculated I_D/I_G ratio of representative GO samples is 0.96 ± 0.04, 1.08 ± 0.01, and 1.96 ± 0.01 for GO-1, GO-2, and GO-3, respectively, implying that it is highly defective after the extensive oxidation and exfoliation process.

The XRD diffractograms (Figure 2d) of GO samples show the presence of a prominent peak at 10.2° (GO-1), 11.9° (GO-2), and 9.8° (GO-3) as expected, suggesting an increased interlayer spacing between the graphene layers compared to graphite (2θ = 26.6 °) due to the insertion of oxygen functional groups during the oxidation and exfoliation process. The peak around 43° can be attributed to the (100) plane of GO-1, reflecting its in-plane graphene structure, while the weak signal at approximately 27° in the GO-3 sample suggests the presence of graphitic domains, likely resulting from the restacking of GO sheets, which enhances layered ordering [41].

TG-DTG profiles of the three tested GO, as illustrated in Figure 2e exhibit nearly similar thermal oxidation patterns with four oxidative degradation steps observed. The values for the temperature of maximum mass change rate, T_{max}, determined from the first derivative of TG plot (DTG) verified the specific components decomposed at their respective T_{max}: <100 °C (moisture), 190–196 °C (oxygen group), ~230 °C (S groups) and 486–578 °C (Carbon). The presence of oxygen groups in the three GO samples was also supported by TGA.

XPS plots of GO-1, GO-2, and GO-3 samples, as depicted in Figure 2f-g verify the preceding characterization results with 67–70 at % carbon and 28–31 at % of oxygen found in all the examined GO. The presence of oxygen functional groups was proven by a high amount of oxygen species, including C-OH, C-O-C, C=O and O-C=O, as detected in C1s high-resolution scans of the GO materials.

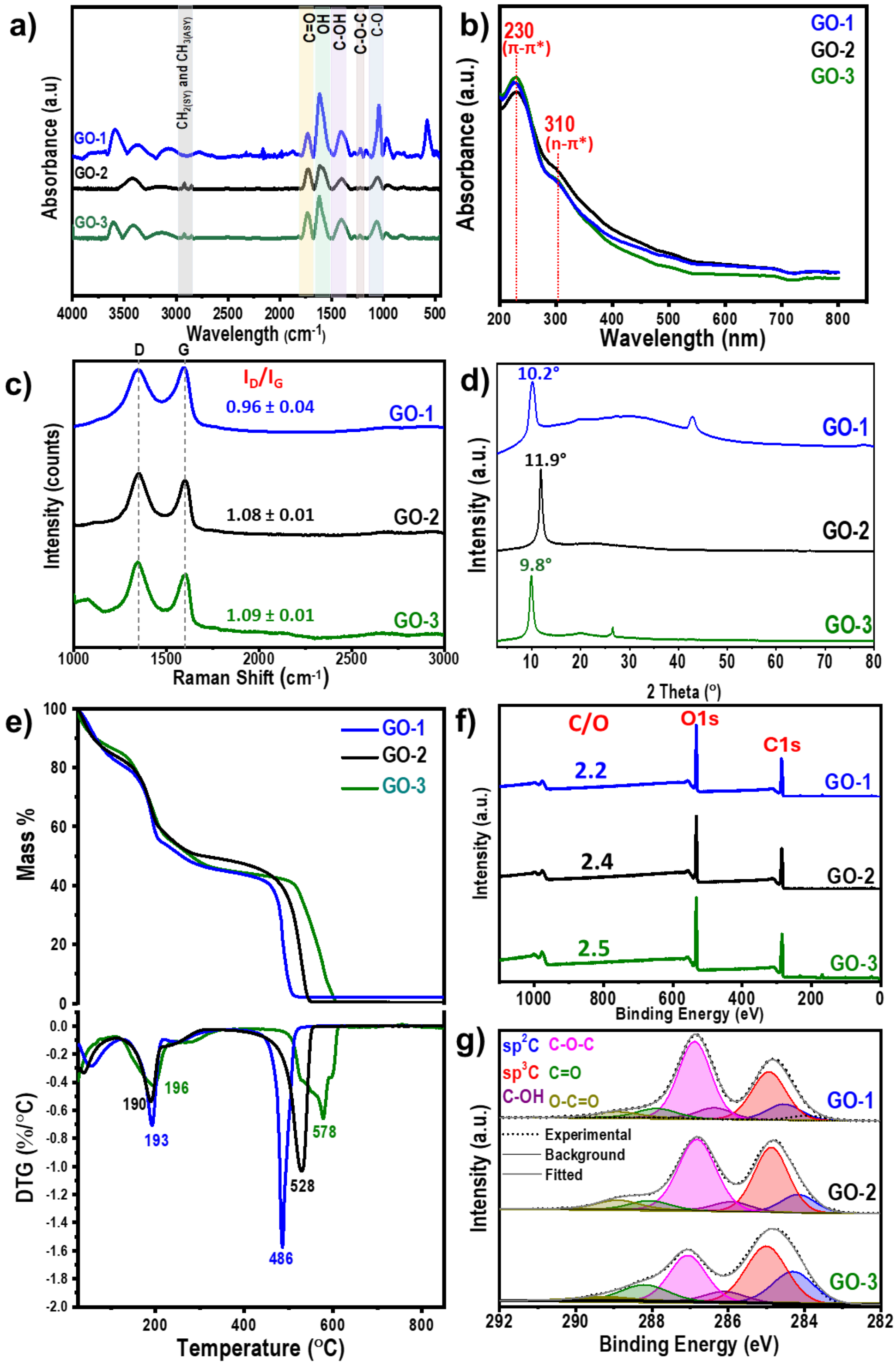

Figure 2. Characterizations of industrially produced GO materials used in this study (GO-1, GO-2 and GO-3): plots of (**a**) FTIR, (**b**) UV-Vis (**c**) Raman, (**d**) XRD, (**e**) TG and DTG, (**f**) XPS survey scan and (**g**) C1s high-resolution scan.

3.2. Quantitative Analysis of Oxygen Functional Groups in GO Using Boehm Titration

To better understand the BT method of GO, a more detailed description of the process is provided in this section. The titration consists of two stages: the reaction of a base titrant with GO and the back titration of the remaining base solution with an HCl solution. In the first stage, the functional groups on GO react with the base solutions to neutralize the respective acidic group according to the pKa/basicity of the base solution. The unreacted base is then quantified through acid-base titration using HCl as the titrant. Acidic and ionised oxygen groups present in GO are often denoted as Brønsted acids (H^+ donors) [42–44] such as carboxyl groups and phenols. Conversely, lactones do not have protons that can be eliminated and the conversion of lactones involves a base-induced hydrolysis reaction [32], where water initiates the process. A hydroxide ion from the water molecule attaches to the cleaved bond, and the released proton neutralizes the base. Anhydrides also undergo hydrolysis with water molecules, but no bases are consumed in the subsequent reactions. Oxidation Debris (OD) which are highly oxidized nanometric carbonaceous fragments produced during GO processing also reacts with bases in the reaction. However, since the releasing rate of OD to alkaline solutions from GO is extremely slow, their effect in the quantification of acidic functional groups through BT is minimal and generally considered negligible [45,46].

3.2.1. Quantitative Analysis of Carboxyl Functional Groups in GO

Titration for the quantitative determination of the concentration of the carboxylic group in industrial GO materials is performed by titration using $NaHCO_3$ with the reaction outlined below. In an aqueous solution mixed with GO material, $NaHCO_3$ hydrolyses into hydroxide ions and leads to the deoxygenation of GO sheets. Carboxylic groups on edges are easily deprotonated with $NaHCO_3$ by substituting H^+ of the group with Na^+ as follows.

$$\mathrm{NaHCO_3 + GO\text{-}COOH \longrightarrow GO\text{-}COO^{-}Na^{+} + HCO_3\,(H_2O{+}CO_2) + Excess\ NaHCO_3} \tag{15}$$

$$\mathrm{NaHCO_3 + HCl \longrightarrow H_2CO_3 + Na^{+}Cl^{-}} \tag{16}$$

Excess $NaHCO_3$ titrated with 0.05 mol/L HCl and corresponding calculation directly provide the carboxyl group content through the above-mentioned Equation (7). Titration for quantitative determination of the concentration of carboxylic group in industrial GO materials obtained by titration of $NaHCO_3$ is presented in Figure 3. By referring to the pH-Vol graph on these figures, the endpoint of the titration was determined from the first derivative of the pH-Vol curve where the solution pH was near 3.7. With the addition of HCl, the pH curve decreases, and two inflection points can be noticed near 4 and 8, which confirms the second derivation of the pH-vol curve where the intersects cross zero. The slight inflection recorded near pH 8 could be attributed to the dissociation of CO_2 from the atmosphere as the solution itself only presents HCO_3^- ions. This phenomenon can be expressed as follows [47],

$$\mathrm{CO_2 + H_2O \longrightarrow H_2CO_3} \tag{17}$$

$$\mathrm{H_2CO_3 + HCl \longrightarrow HCO_3^{-} + H^{+}} \tag{18}$$

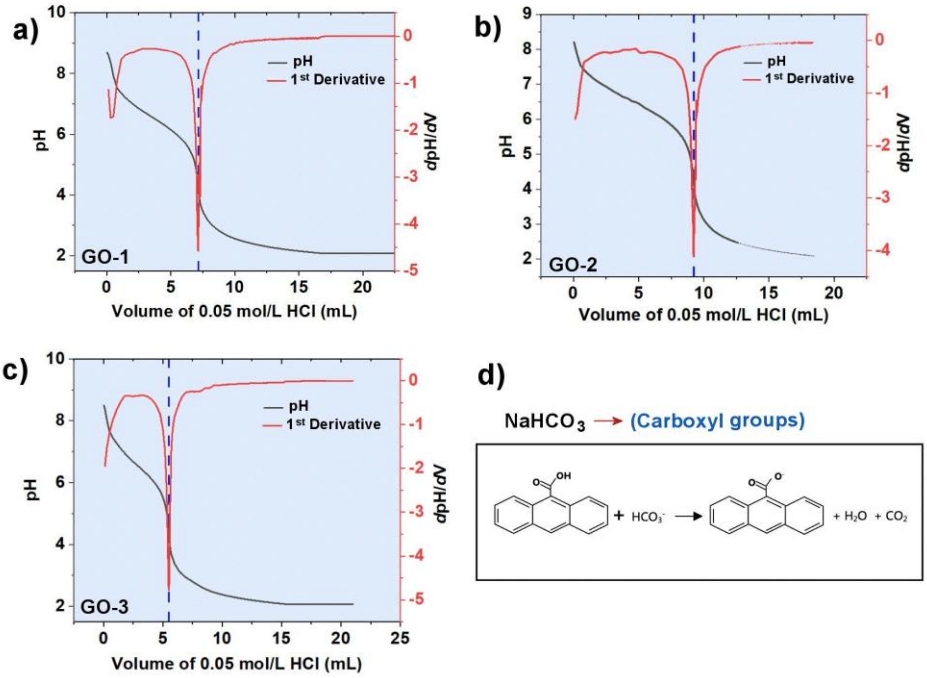

Figure 3. Boehm titration results for quantitative analysis of carboxyl functional groups showing pH-Vol curve of a titration between remaining $NaHCO_3$ and HCl after the base reaction with a) GO-1 b) GO-2, c) GO-3 and d) reaction mechanism.

The carboxyl groups concentration (mmol/g and mass %) for the three GO samples is presented in Table 1 with COOH concentration recorded in the range of 0.89 ± 0.01–1.91 ± 0.08 mmol/g and the mass % from 4.00 ± 0.04 to 8.59 ± 0.36%. Results showed that GO-1 has the lowest, while GO-3 exhibited the highest COOH concentration.

Table 1. The quantitative analysis of carboxyl functional groups in industry produced GO sample obtained by $NaHCO_3$ titration.

	GO-1	**GO-2**	**GO-3**
Carboxyl Group Concentration (±SD mmol/g)	0.89 ± 0.01	1.64 ± 0.13	1.91 ± 0.08
Carboxyl Group (±SD mass %)	4.00 ± 0.04	7.37 ± 0.60	8.59 ± 0.36

3.2.2. Quantitative Analysis of Lactonic Functional Group in GO

Titration for quantitative determination of the concentration of lactonic functional group in industrial GO materials was performed by titration using Na_2CO_3 as a titrant. The titrant, Na_2CO_3, is a stronger base than $NaHCO_3$ to react with both lactonic and carboxylic groups on GO materials. In the titration of excess Na_2CO_3, HCl is introduced with Na_2CO_3, a double displacement reaction takes place, as in step 1, CO_3^{2-} is converted to HCO_3^- and then HCO_3^- is converted into CO_2. On the basal plane of GO, lactone groups called γ-alkyl butyrolactone, react with Na_2CO_3 aqueous solution as follows, the process called saponification [48].

$$\gamma^- \text{ Lactone} + H_2O + Na_2CO_3 \rightleftharpoons \text{Salt (COO-Na+)} + NaHCO_3 \tag{19}$$

Due to the high value of isobaric potential (>5.0 kcal/mole) of the reaction between $NaHCO_3$ and Lactonic group, $NaHCO_3$ does not react with γ-alkyl butyrolactone in compounds that contain more than 10 carbon atoms, between 20–120 °C. $NaHCO_3$ derives from Na_2CO_3, tends to deprotonate –COOH, as mentioned in Section 3.2.1. The lactone amount can be calculated after substituting the number of carboxylic acid groups within the GO sample from the calculated mol amount equivalent to the second endpoint of the titration curve with respect to the reaction between Na_2CO_3 and HCl.

Titration for quantitative determination of the concentration of carboxylic group in industrial GO materials obtained by titration with Na_2CO_3 is presented in Figure 4. Two endpoints (3.7 and 8.3) can be seen in the first derivative of the pH-Vol curve as the reaction takes place in two main reactions: first CO_3^{2-} is converted to HCO_3^-

and further converted to CO_2 by HCl, where the 2nd endpoint was taken and for the calculations. The determined results for lactonic group concentration (mmol/g and mass %) on GO samples are presented in Table 2. Record shows that the amount of -COO of the tested GO samples is within the range of (0.20 ± 0.01–1.76 ± 0.26) mmol/g and mass percentage between 0.88 ± 0.04–7.73 ± 1.15%.

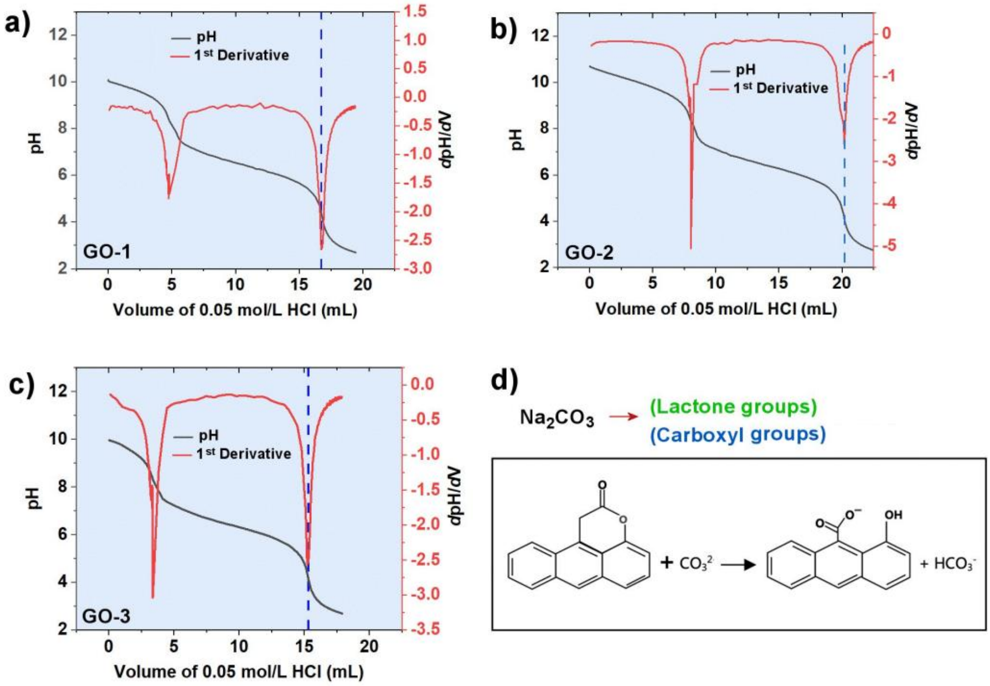

Figure 4. Boehm titration results for quantitative analysis of lactonic functional groups showing pH-Vol curve of a titration between remaining Na_2CO_3 and HCl after the base reaction with a) GO-1 b) GO-2, c) GO-3 and d) reaction mechanism.

Table 2. The quantitative analysis of carboxyl functional groups in industry produced GO sample obtained by $NaHCO_3$ titration.

	GO-1	**GO-2**	**GO-3**
Lactonic Group Concentration (±SD mmol/g)	0.20 ± 0.01	1.76 ± 0.26	0.41 ± 0.04
Lactonic Group (±SD mass %)	0.88 ± 0.04	7.73 ±1.15	1.82 ± 0.18

3.2.3. Quantitative Analysis of Hydroxyl (Phenolic) Group in GO

Titration for quantitative determination of the concentration of hydroxyl functional group in industrial GO materials was performed by titration using NaOH as a titrant. During the reaction with NaOH, (a stronger base than $NaHCO_3$ and Na_2CO_3), it reacts with lactonic, carboxylic groups and phenolic groups on GO materials.

Alcoholic hydroxyl groups (R-OH) and phenolic hydroxyl groups (Ar-OH) are the two main kinds of hydroxyl groups [49,50] present in GO materials. Because of the varied ways in which they are connected, these two types of hydroxyl groups have different characteristics, including pKa value and hydrophilicity. The phenolic hydroxyl group is ionized and acidic in an aqueous solution, and its hydrophilicity is superior to that of the alcoholic hydroxyl group, even though both hydroxyl groups are extremely hydrophilic [49].

The titration of excess NaOH with HCl is considered a strong base-strong acid titration that gives an equivalent point at the pH of 7. NaOH reacts with carboxylic and phenolic groups, substituting their H^+ of the group with Na^+. For lactonic groups, first, the lactonic groups hydrolyse with water and then deprotonate, which gives rise to the resulting phenol. Phenolic group content can be calculated by substituting the carboxylic and lactone group content from the corresponding mole amount with respect to the consumed NaOH moles.

Titration for quantitative determination of the concentration of phenolic group in industrial GO materials obtained by titration Na_2CO_3 is presented in Figure 5. As the reaction is a strong acid-base reaction, the endpoint of the reaction is recorded at 7, and the effect of CO_2 does not make an alteration to the results. The results for phenolic group concentration (mmol/g and mass %) for GO samples are presented in Table 3. Three GO samples where the -OH group was found to be in the range of 1.12 ± 0.15 to 2.73 ± 0.05 mmol/g and 3.24 ± 0.41 to 7.91 ± 0.13 mass % for the three examined GO samples.

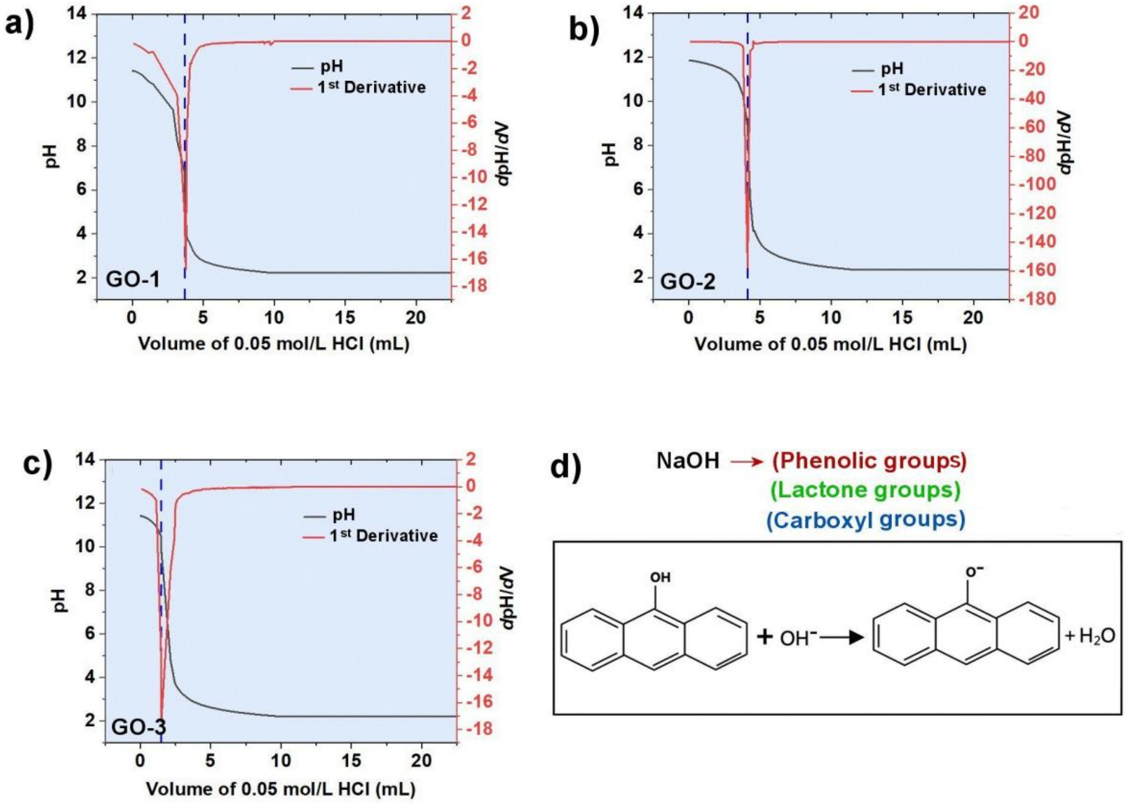

Figure 5. Boehm titration results for quantitative analysis of phenolic (hydroxyl) functional groups showing pH-Vol curve of a titration between remaining NaOH and HCl after the base reaction with a) GO-1 b) GO-2, c) GO-3 and d) reaction mechanism.

Table 3. The quantitative analysis of phenolic (hydroxyl) functional groups in industry produced GO sample obtained by NaOH titration.

	GO-1	**GO-2**	**GO-3**
Hydroxyl Group Concentration (±SD mmol/g)	2.73 ± 0.05	1.15 ± 0.15	1.12 ± 0.15
Hydroxyl Group (±SD mass %)	7.91 ± 0.13	3.33 ± 0.42	3.24 ± 0.41

3.2.4. Quantitative Analysis of Carbonyl Functional Groups in GO

Titration for quantitative determination of the concentration of carbonyl functional group in industrial GO materials is performed by titration using C_2H_5ONa as a titrant. The C_2H_5ONa is the strongest base used in BT, capable of reacting with all four functional groups (carboxyl, lactone, hydroxyl and carbonyl) on GO material. The titration of excess C_2H_5ONa with HCl is considered a strong base-strong acid titration where the end-point pH reports 7. The carbon atom of the carbonyl functional group (C=O) is a Lewis acid, therefore, $-OC_2H_5$. can act as a Lewis base to form the deprotonated hemiacetal. C_2H_5ONa is highly reactive, where titrations were conducted under N_2 purging to prevent CO_2 interferences.

Quantitative determination of the carbonyl group concentration in industrial GO materials obtained by titration C_2H_5ONa is presented in Figure 6, and the carbonyl group concentration in mmol/g and mass % for GO samples are presented in Table 4. for three GO samples at 0.65 ± 0.19 to 2.54 ± 0.08 mmol/g and 1.81 ± 0.52 to 7.12 ± 0.23 mass %, respectively. According to the study by Lerf [51], $NaOC_2O_5$ can break some of the epoxide groups, which might affect the actual concentration of carbonyl groups.

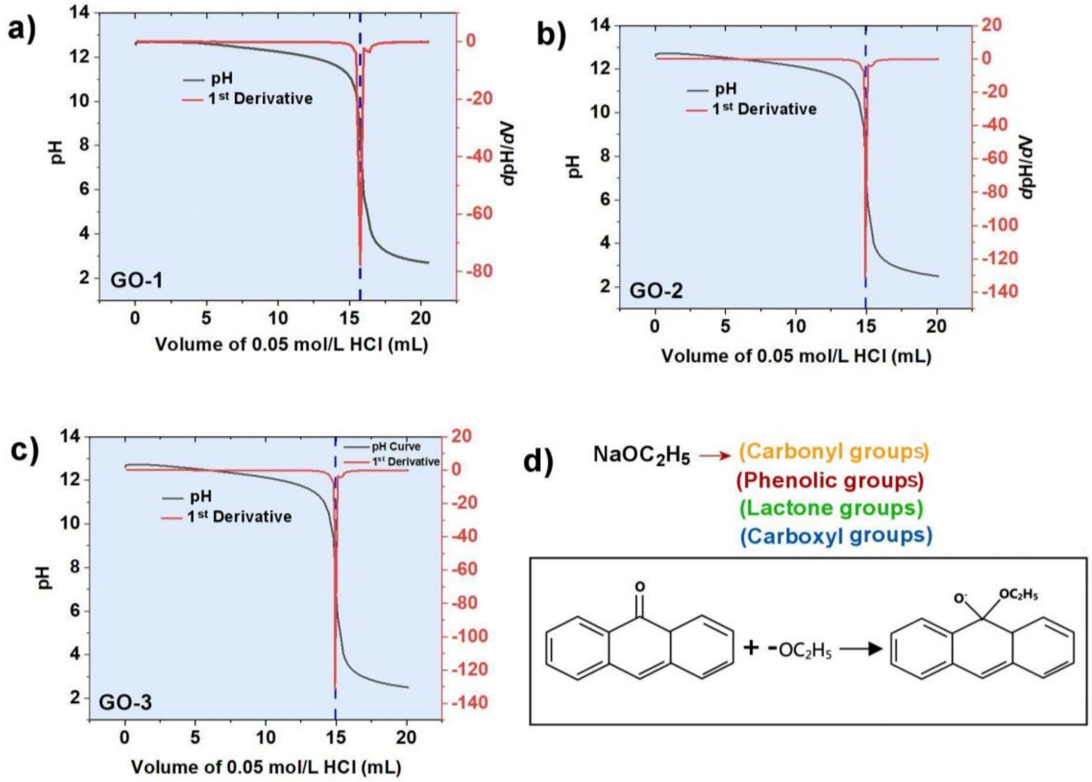

Figure 6. Boehm titration results for quantitative analysis of carbonyl functional groups showing pH-Vol curve of a titration between remaining $Na_2OC_2H_5$ and HCl after the base reaction with a) GO-1 b) GO-2, c) GO-3 and d) reaction mechanism.

Table 4. The quantitative analysis of carbonyl functional groups in industry produced GO sample obtained by C_2H_5ONa titration.

	GO-1	GO-2	GO-3
Carbonyl group concentration (±SD mmol/g)	2.21 ± 0.26	2.54 ± 0.08	0.65 ± 0.19
Carbonyl group (±SD mass %)	6.18 ± 0.72	7.12 ± 0.23	1.81 ± 0.52

3.3. Quantitative Analysis of Epoxy Functional Groups in GO Using Catalyst-Assisted Acid Titration

The process is based on HCl acid titration with $MgCl_2$ catalyst-assisted reaction, which is commonly utilized for water-soluble polymers and has been modified to determine the epoxy group concentration in GO materials [33]. In order to open an epoxide ring, nucleophilic addition and stereochemical inversion are required. Lewis acid often polarizes the epoxy group's oxirane C–C bond, which causes these two carbon atoms to become electrophilic [52]. Thus, GO epoxy ring opening happens when Lewis acid ($MgCl_2$) couples with oxirane oxygen because it is readily available for nucleophilic attack on epoxide oxygen.

The concentration of HCl employed for the reaction with epoxy groups will be ascertained by treating excess HCl with 0.05 mol/L NaOH solution following the reaction and analyzing the produced titration curves. The epoxide group content (mmol/g) was calculated based on the amount of NaOH solution utilized to titrate excess HCl following Equation (12) mentioned above, and the results of the titration are presented in Figure 7 and Table 5 below. The results for Epoxide group concentration (mmol/g and mass %) for GO samples were found to be in the range of 1.15 ± 0.05 to 1.37 ± 0.05 mmol/g and 4.58 ± 0.19 to 5.50 ± 0.20 mass % for the three examined GO samples.

Table 5. The quantitative analysis of epoxide groups in industry produced GO sample obtained by $MgCl_2$-HCl titration.

	GO-1	GO-2	GO-3
Epoxide Group Concentration (±SD mmol/g)	1.35 ± 0.02	1.15 ± 0.05	1.37 ± 0.05
Epoxide Group (±SD Mass %)	5.42 ± 0.07	4.58 ± 0.19	5.50 ± 0.20

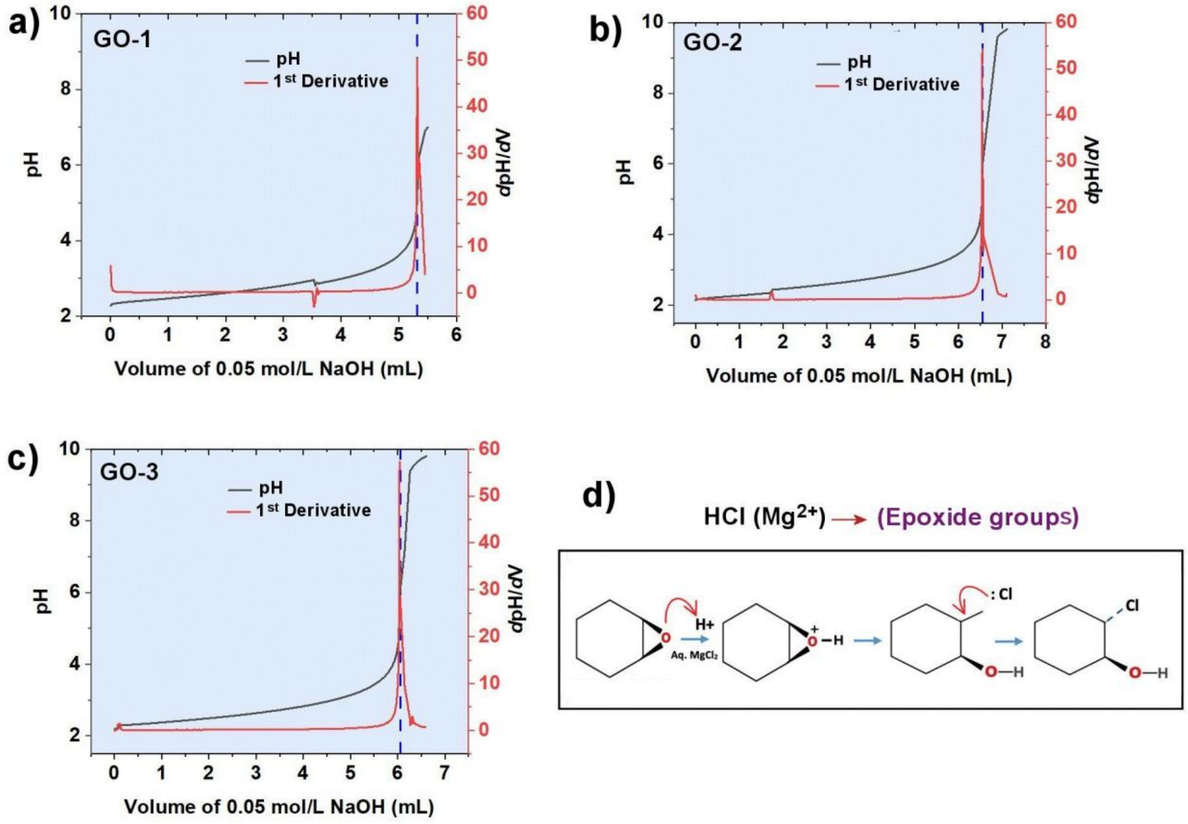

Figure 7. Boehm titration results for quantitative analysis of carbonyl functional groups showing pH-Vol curve of a titration between remaining HCl and NaOH after the acid reaction with a) GO-1 b) GO-2, c) GO-3 and d) reaction mechanism

3.4. Comparative Analysis of Oxygen Functional Groups in Industrially Produced GO Materials

GO is industrially produced from graphite using various oxidation methods and conditions such as time, temperature, exfoliation process), each differing in final GO products, level of oxygen groups, impurities, efficiency and environmental impact. The Hummers' method is the most widely used due to its relatively safe and scalable process, involving potassium permanganate ($KMnO_4$) and concentrated sulfuric acid (H_2SO_4). This method provides moderate to high oxidation levels and, with modification is used in majority of industrial processes. In contrast, the Brodie's method, which uses fuming nitric acid (HNO_3) and potassium chlorate ($KClO_3$), achieves a higher oxidation degree and more controlled structure, but poses significant safety risks due to the potential for explosive reactions and chlorine dioxide release and is less likely used. However, there is no manufacturer of GO materials on the global market that provides GO product specification about level of oxygen groups regardless their materials. The absence of such analytical information not only creates challenges for end users, making it difficult to select the most suitable GO for their product development, but also hinders their ability to monitor batch-to-batch variations effectively.

Based on the titration results, the summary of concentration of all oxygen functional group concentrations is presented in Figure 8 and Table 6, which includes the calculated total concentration of the oxygen functional group. The total oxygen group concentrations are in the range from 0.24 mmol/g to 0.30 mmol/g or 20.84 mass % to 30.41 mass % for three different GO materials. These results clearly confirm significant variations of specific and total oxygen functional groups (specifically GO-2) depending on the manufacturing and oxidation process, making GO with different levels of oxidation. The level of oxidation is determined by conditions such as the oxidation agent, amount of oxidation agent, temperature, time and the graphite precursor as well [53]. These three GO manufacturers did not disclose their GO manufacturing process and conditions, and we are not able to relate the presented results with their production methods.

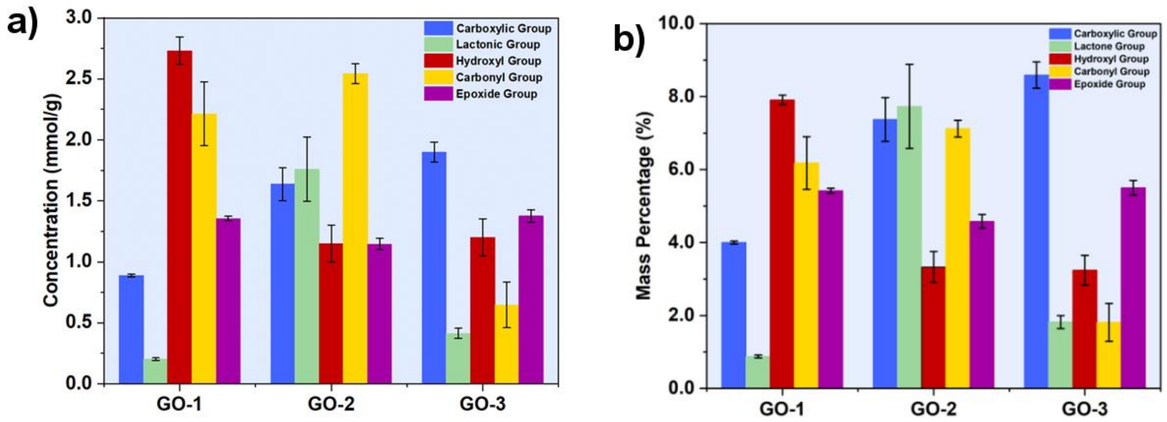

Figure 8. Calculated oxygen functional group concentrations in commercial GO samples (GO-1, GO-2 and GO-3) presented as (**a**) mmol/g and their (**b**) corresponding mass % concentrations.

Table 6. Calculated functional group concentrations and mass % in commercial GO samples based on titration results XPS (SD: Standard Deviation).

	Carboxyl Group Con. ±SD (mmol/g) ±SD (Mass %)	Lactone group Con. ±SD (mmol/g) ±SD (Mass %)	Phenolic Group Con. ±SD (mmol/g) ±SD (Mass %)	Carbonyl Group Con. ±SD (mmol/g) ±SD (Mass %)	Epoxide Group Con. ±SD (mmol/g) ±SD (Mass %)	Total oxygen Con. (mmol/g) Mass %
GO-1	0.89 ± 0.01 4.00 ± 0.04	0.20 ± 0.01 0.88 ± 0.04	2.73 ± 0.05 7.91 ± 0.13	2.21 ± 0.26 6.18 ± 0.72	1.35 ± 0.02 5.42 ± 0.07	**0.24** **24.23**
GO-2	1.64 ± 0.13 7.37 ± 0.60	1.76 ± 0.26 7.73 ± 1.15	1.15 ± 0.15 3.33 ± 0.42	2.54 ± 0.08 7.12 ± 0.23	1.15 ± 0.05 4.58 ± 0.19	**0.30** **30.41**
GO-3	1.91 ± 0.08 8.59 ± 0.36	0.41 ± 0.04 1.82 ± 0.18	1.12 ± 0.15 3.24 ± 0.41	0.65 ± 0.19 1.81 ± 0.52	1.37 ± 0.05 5.50 ± 0.20	**0.21** **20.84**

To gain further insights into the quantitative analysis of oxygen functional groups (mass %) obtained by Bohem and catalytic acid titration, we conducted a comparative chemical analysis obtained with XPS (Figure 2f-g). The quantitative XPS results are generated by the deconvolution of the spectra from C1s peak with several different peaks that enable to provide quantitative information about sp^2 vacancy carbon, C–O (phenols, ethers), C=O (carbonyl, quinones) and O–C=O (carboxyl groups, lactones), which are presented in Table 7. The mass % of functional groups is calculated from elemental or atomic % mass quantification of chemical components. These preliminary XPS values are not in well agreement with titration results and must be taken with the precaution for several reasons. Firstly, they are not specifically related to individual functional groups; they may present these elements as other components, such as moisture and impurities because the deconvolution process has uncertainties depending on the selection of fitting parameters. More studies are needed to correlate these results between different quantitative methods to better understand their differences, limitations and discrepancies.

Table 7. Calculated functional group concentrations in commercial GO samples based on titration results compared with XPS.

Sample	Method	Mass %					
		COOH	**O-C=O**	**C-OH**	**C=O**	**C-O-C**	**Total O group**
GO-1	**Titration**	4.00	0.80	7.90	6.10	5.40	**24.2**
	XPS	3.02		3.14	2.82	28.20	**37.2**
GO-2	**Titration**	7.30	7.70	3.80	7.10	4.50	**30.4**
	XPS	4.18		2.50	2.37	26.04	**35.1**
GO-3	**Titration**	8.50	1.80	3.20	1.80	5.50	**20.8**
	XPS	2.34		3.87	6.54	20.98	**33.7**

3.5 Introducing New Quality Parameters: Oxygen Group Indexes (OGI)

Addressing the concentrations of functional groups in GO materials is crucial for their practical applications, as it enables the tuning of material properties to meet specific requirements. In addition to concentration measurements, there is a need for their transformation into quality classifications and the index values that are commonly used in ISO standard [38]. For this purpose, we introduced for the first time a new quality parameter for GO termed Oxygen Group Indexes "OGI" defined as the number of moles of specific functional groups present in 1 kg of GO material. For each functional group, we defined oxygen group indexes including carboxyl (CxylGI), lactone (LacGI), phenol (PhGI), carbonyl (CnylGI), epoxy (EpGI), and total TotOGI. Based on the titration results obtained, we calculated OGI values for each commercial GO material, as presented in Table 8. These newly introduced OGIs for specific functional groups are valuable parameters that provide critical chemical characteristics and quality indicators for GO materials. These indexes enable manufacturers and end-users to better understand the properties of the GO they produce and optimised their practical applications. By correlating OGIs with key material attributes such as reactivity, dispersion stability, electronic behaviour, and interactions with other substances, manufacturers can fine-tune GO for specific applications, while end-users can make informed purchasing decisions to meet their product development requirements. For instance, carboxyl groups present in GO can serve as anchor points for binding GO to polymer matrices, enhancing the mechanical properties of composite materials. Access to precise OGI data allows for more accurate chemical modification and functionalization of GO, facilitating its role as a versatile building block for composites and hybrid materials.

Table 8. Calculated oxygen functional group indexes (OGI) of commercial GO samples based on titration results.

	CxylGI	**LacGI**	**PhGI**	**CnylGI**	**EpGI**	**TotOGI**
GO-1	0.889	0.202	2.731	2.213	1.355	7.390
GO-2	1.637	1.759	1.150	2.543	1.146	8.235
GO-3	1.909	0.414	1.120	0.647	1.376	5.466

Quantifying and understanding the oxygenated functional groups in GO materials is a critical step for a wide range of advanced applications including composites, energy storage, environmental remediation, sensors, and coating and their industrial adoption. These indexes as new quality parameters will streamline the differentiation of GO materials produced via various synthesis methods, thereby clarifying their chemical compositions in a more accessible manner. This quality approach not only enhances understanding of the functional group profiles of GO materials but also provides essential information for researchers and manufacturers in selecting suitable materials for their specific applications. The introduction of these new quality parameters will provide very useful practical metrics for GO industrial applications, particularly for their composite applications where information about specific functional groups such as epoxide groups is critical.

4. Conclusions

In summary, this work successfully demonstrates the implementation and validation of combined titration methods to provide a comprehensive quantitative determination of oxygen functional groups in GO materials. The method is based on Bohem and catalyst-assisted titration water that can be used for GO materials in different forms such as powders, pastes and dispersions. The presented results using series of industrially produced GO materials showed that concentration (mmol/g and mass %) of series of oxygen functional groups including of carboxylic, lactone, hydroxyl, carbonyl, epoxy groups and the total oxygen groups, can be successfully determined by this simple and low-cost method. The results provide a comprehensive understanding of the oxygen functionalities present on the basal plane and edges of GO, which are integral to its performance in various applications. The introduction of new quality parameters such as oxygen group indexes (OGIs) for each functional group and total oxygen group, further enables a more precise evaluation of GO's functional group content, paving the way for more targeted and efficient use in future research and industrial processes. Hence, this study contributes to the growing body of knowledge on GO quality control and characterization, offering valuable insights and new analytical and quality control tool for graphene industry and graphene researchers that are currently lacking.

Supplementary Materials: The Supporting Information presenting more details about presented analytical methods is available free of charge at Journal website.

Author Contributions: Gimhani Danushika: Writing – original draft, Validation, Methodology, Formal analysis. Pei Lay Yap: Validation, Supervision, Writing – review & editing, Investigation, Data Analysis, Methodology, Conceptualization; Dusan Losic: Writing – original draft, Writing – review & editing, Resources, Supervision, Methodology, Conceptualization

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