Malaysian Journal of Microscopy 2025 21(2) 14-22

MALAYSIAN JOURNAL OF MICROSCOPY

Journal homepage: https://malaysianjournalofmicroscopy.org

RESEARCH ARTICLE

NOVEL SYNTHESIS OF GRAPHENE OXIDE (GO) FROM PYROLYZED WASTE TIRE: TUNING THE OXIDATION DEGREE VIA VARYING $KMnO_4$

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Abstract. Graphene oxide (GO) is highly valued for its tunable properties, making them versatile for high-performance applications. Reliance on high-purity graphite flakes as the primary precursor increases production cost and raises environmental concerns, highlighting the need for sustainable alternatives. Converting carbon from waste materials into GO offers a green approach, but remains challenging due to their heterogenous composition and lower graphitic order compared to conventional graphite. Thus, this research presents the synthesis and characterization of GO from recovered carbon black (rCB) of pyrolysis waste tires through modified Hummers' method, with a focus on tuning the concentration of oxidizing agent ($KMnO_4$). The synthesized materials were thoroughly analyzed using X-ray diffraction (XRD), Fourier-transformed infrared spectroscopy (FTIR), and field emission scanning electron microscope-energy dispersive X-ray (FESEM-EDX). Results show that increasing $KMnO_4$ concentration promoted the oxidation and exfoliation of rCB to GO, as shown by the XRD peak shifting to a lower angle ($2\theta = 11.23°$), which was first observed at a $KMnO_4$ concentration of 3 g. FTIR analysis revealed that increasing $KMnO_4$ concentration enhanced the oxidation of rCB, as supported by the increase in the absorption bands at 3395 cm^{-1} (O-H), 1735 cm^{-1} (C=O), and 1215 cm^{-1} (C-O) in GO-5 samples, indicating successful incorporation of oxygen-containing functionalities with higher oxidation levels. FESEM images further supported these findings, showing a morphological transformation from aggregated spherical particles in raw rCB to thin, wrinkled GO sheets at higher $KMnO_4$ concentrations, indicating successful exfoliation and GO formation. These findings demonstrate that controlling the concentration of $KMnO_4$ influences the oxidation degree and structural properties of synthesized GO from carbon source derived from waste and provides a promising pathway for recycling waste into high-value materials.

Keywords: Graphene oxide, waste tire, carbon, Hummers' method.

Article Info

Received 17 August 2025
Accepted 20 September 2025
Published 4 December 2025

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ISSN: 1823-7010, eISSN: 2600-7444

1. INTRODUCTION

Graphene oxide (GO) is a two-dimensional carbon-based nanomaterial derived from the oxidation of graphite. Recently, GO has gained significant attention due to its unique structure, which includes abundant oxygen-containing functional groups, high surface area, and tunable physicochemical properties [1]. These characteristics make GO a promising material for many applications, including membrane technology, energy storage, sensors, and biomedical devices [2,3]. GO was primarily obtained from purified graphite using oxidative processes such as the modified Hummers' method. However, the reliance on graphite poses several limitations. It is non-renewable and relatively expensive as it requires high-purity grades of graphite, thus hindering its large-scale production and resulting in expensive GO prices in the global market.

To overcome this drawback, researchers have focused on alternative carbon sources, particularly those derived from waste materials [4]. One promising source is the recovered carbon black (rCB) obtained from the pyrolysis of waste tires. The accumulation of waste tire generation was estimated to reach 1 billion tons and projected to reach 1.2 billion tons in 2030 [5]. The waste tire generated in massive quantities presents a significant environmental burden due to their non-biodegradability and fire hazards. Pyrolysis converts these tires into valuable byproducts, including carbon-rich rCB containing amorphous and graphitic carbon domains [6]. Using rCB as a precursor to the synthesis of GO helps address waste management challenges and supports the circular economy by transforming low-value waste into high-value nanomaterials.

Despite this appeal, the synthesis of GO from rCB remains a complex task due to the varying graphitic nature of the carbon black compared to natural graphite [7]. These structural differences affect the interaction between oxidizing agents and the carbon source, which in turn influence the oxidation degree, structural integrity, and quality of the resulting GO [8]. Moreover, there is a critical lack of optimization in the synthesis parameters, particularly regarding the concentration of oxidizing agents like potassium permanganate ($KMnO_4$), which play a vital role in determining the extent of oxidation and the functionalization of GO. Previous research has introduced approaches to enhance GO production from waste sources [9], but systematic optimization remains insufficient. It is also worth noting that different carbon sources behave differently during oxidation, further affecting the characteristics of the final GO product [10].

Therefore, the objective of this study is to synthesize GO from carbon derived from pyrolyzed waste tire and investigate the effect of varying $KMnO_4$ concentrations on the structural and morphological properties of the resulting GO. By examining the relationship between the oxidant concentration and the characteristics of GO, this study provides the optimize synthesis conditions suitable for carbon sources derived from waste material and advancing the sustainable and cost-effective production of GO.

2. MATERIALS AND METHODS

2.1 Synthesis of GO Using Modified Hummers' Method

Graphene oxide (GO) was synthesized using recovered carbon black (rCB) derived from waste tires as the precursor, following a modified Hummers' method. The rCB used in this study was obtained from Eco Power Synergy Sdn. Bhd. with contained carbon (C) element ranging from 92% to 98%. Initially, 23 mL of concentrated sulfuric acid (H_2SO_4), 1.0 g of sodium nitrate ($NaNO_3$), and 1 wt.% of rCB were added into a beaker placed in an ice bath. The mixture was stirred for one hour at a temperature below 10 °C. To investigate the effect of oxidant amount, three GO samples were prepared by gradually adding potassium permanganate ($KMnO_4$) in varying amounts of 1 g (GO-1), 3 g (GO-3), and 5 g (GO-5), followed by continuous stirring for another hour while maintaining the temperature below 20 °C.

The resultant mixture was then heated to 35 °C and stirred for 30 minutes. Following this, 46 mL of deionized water was added, and the reaction temperature was increased and maintained at 98 °C for 15 minutes to complete the oxidation process. The heater was turned off, and the mixture was cooled to room temperature.

To facilitate the exfoliation of graphite oxide into graphene oxide, 100 mL of distilled water was added, and the mixture was stirred for an additional hour. To terminate the reaction and eliminate residual oxidants, 10 mL of 30% hydrogen peroxide (H_2O_2) was slowly added and stirred for one hour. The resultant mixture was subjected to six cycles of centrifugation (5 minutes per cycle at 8000 rpm) using a washing solution of hydrochloric acid (HCl) and distilled water in a 1:3 ratio, with a total volume of 1000 mL. Finally, the washed GO solution was dried at 70 °C for 24 hours in an oven.

2.2 *Material Characterizations*

The X-ray diffraction (XRD) patterns were obtained by using Cu Kα radiation (λ=1.5406 Å) with an acceleration voltage of 40 kV and a current of 20 mA, on a Rigaku Rigaku-Denki (Geigerflex). Using the Perkin Elmer Spectrum 400 ATR-FTIR, the FTIR spectra of the rCB and the GO samples were obtained at a resolution of 16 cm^{-1} for a frequency of 20 kHz, ranging from 500 to 4000 cm^{-1}. A field emission scanning electron microscope (Model: JSM-7600F, Brand: Jeol) was used to analyze the surface morphology of the rCB and GO samples at 300 kX. The samples were placed on a holder after being thinly coating with gold-palladium (Au-Pd) to ensure a clear and accurate image.

3. RESULTS AND DISCUSSION

3.1 X-ray Diffraction (XRD) Analysis

The X-ray diffraction (XRD) analysis was performed to evaluate the crystallinity and phase transitions of recovered carbon black (rCB) derived from waste tires and graphene oxide (GO) synthesized at varying potassium permanganate ($KMnO_4$) concentrations. As shown in Figure 1, the XRD pattern of rCB displayed a distinct peak at $2\theta = 25.51°$, corresponding to the (002) plane of graphite, which signifies the stacked arrangement of graphene layers. An additional peak at $2\theta = 44.67°$, attributed to the (101) plane, indicates the presence of in-plane crystalline ordering. Furthermore, a peak at $2\theta = 78.16°$, associated with the (110) plane, was observed, suggesting a relatively high degree of graphitization [11].

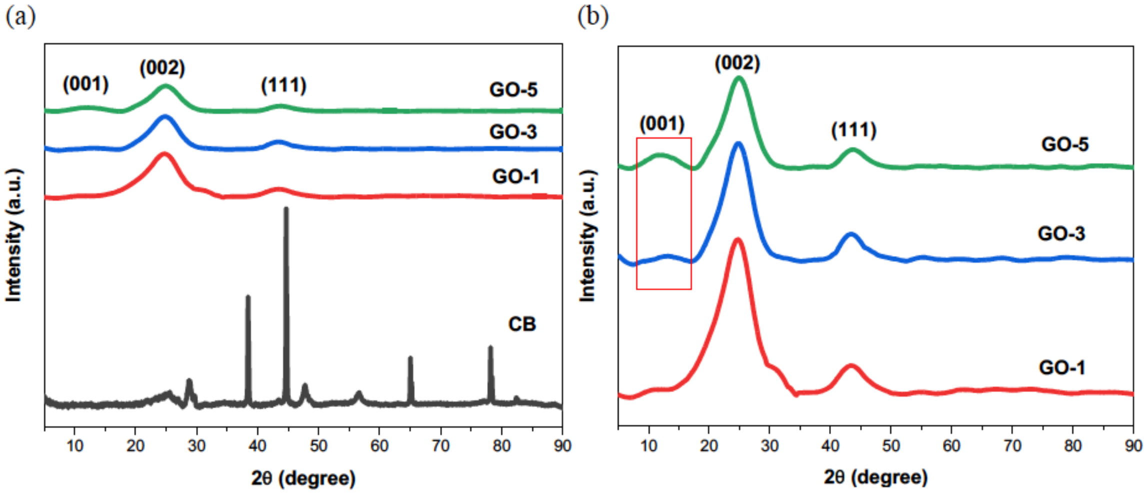


Figure 1: XRD patterns (a) CB, GO-1, GO-3, and GO-5, and (b) magnified XRD results showing the presence of new peak at ~10° indicates formation of GO

Notably, GO samples synthesized with higher $KMnO_4$ concentrations, namely GO-3 and GO-5, exhibited new diffraction peaks at $2\theta = 11.23°$ and $2\theta = 10.65°$, respectively, corresponding to increased interlayer spacings (d-spacing) of 0.79 nm and 0.83 nm. These d-spacing values are consistent with previously reported data on GO synthesized from graphite, confirming successful oxidation of rCB and introducing oxygen-containing functional groups [12,13]. However, the appearance of a dominant peak around $2\theta = 24.57°$ in all GO samples suggests partial restacking of the GO layers. This restacking likely results from incomplete exfoliation, which reduces the interlayer spacing and shifts the GO diffraction peak toward higher angles [14]. The presence and evolution of these peaks further support the hypothesis that increasing $KMnO_4$ concentration enhances the oxidation extent, leading to more effective exfoliation of GO sheets and expansion of the interlayer distance.

3.2 Chemical Group Identification via FTIR

The Fourier-transform infrared (FTIR) analysis of GO samples synthesized with different concentrations of $KMnO_4$ reveals notable differences in the functional groups present. Figure 2 shows the FTIR spectra representing the functional groups of three different GO samples. The broad peak around 3407-3392 cm^{-1} corresponds to hydroxyl (O–H) stretching vibrations. The prominent peak observed in the sample synthesized with 1 g of $KMnO_4$ (GO-1) suggests a high density of hydroxyl groups. This indicates that at lower $KMnO_4$ concentrations, significant incorporation of hydroxyl functionalities occurs, a feature characteristic of partially oxidized GO.

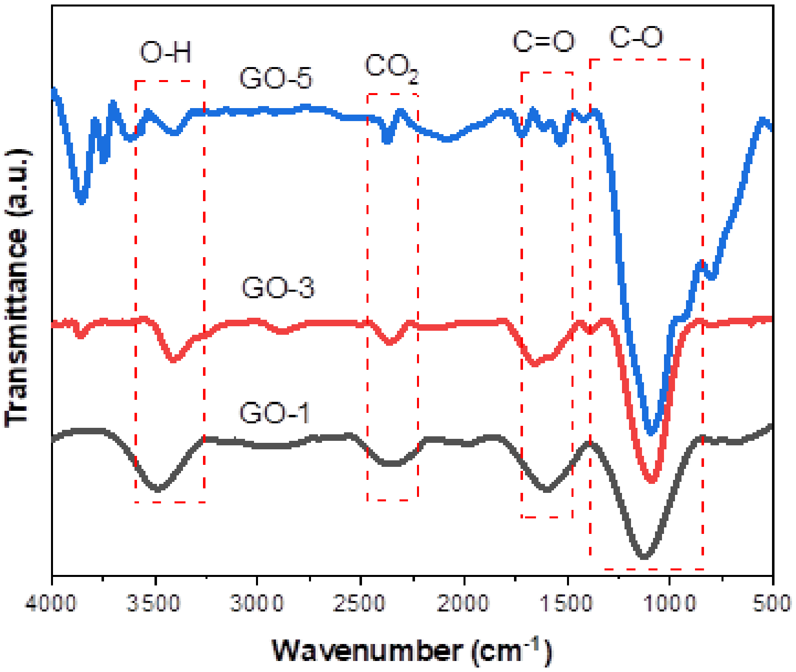


Figure 2: FTIR spectra of GO-1, GO-3, and GO-5.

All three samples exhibit a peak at approximately 2394 - 2381 cm^{-1}, attributed to CO_2. The consistent appearance of this peak across all samples suggests that CO_2 formation is a common byproduct of the oxidation process. These peaks arise due to the physical adsorption of atmospheric CO_2 onto the highly oxygenated and hydrophilic surface of GO, which occurs during the post-synthesis steps such as washing, drying, or storage [15]. The peak at 1746- 1735 cm^{-1}, corresponding to the stretching vibration of carbonyl (C=O) groups, is less pronounced in GO-1. This indicates that lower $KMnO_4$ concentrations may not sufficiently promote the formation of carbonyl functionalities. Conversely, the appearance of this peak in the GO-3 and GO-5 samples confirms that higher $KMnO_4$

concentrations enhance the oxidation degree, enabling successful incorporation of carbonyl groups into the GO structure.

Additionally, epoxy (C–O) stretching vibrations appear at approximately 1210 cm^{-1} for GO-1, 1193 cm^{-1} for GO-3, and 1215 cm^{-1} for GO-5. Among these, GO-5 exhibits the highest intensity for epoxy groups, further confirming the increased incorporation of oxygen functionalities at higher oxidant concentrations. The FTIR peaks and their corresponding functional groups are summarized in Table 1

Table 1: Wavenumber and functional group for three GO samples with different concentrations of $KMnO_4$.

Functional group	GO-1 (Wavenumber, cm^{-1})	GO-3 (Wavenumber, cm^{-1})	GO-5 (Wavenumber, cm^{-1})	Reference (Wavenumber, cm^{-1}) (Sujiano et al., 2020)
O-H	3392	3407	3395	3435
CO_2	2381	2384	2394	2363
C=O	1740	1746	1735	1726
C-O	1210	1193	1215	1111

3.3 Surface Morphology and Elemental Composition Analysis via FESEM-EDX

Field emission scanning electron microscopy (FESEM) was employed to investigate the surface morphology and structural transformation of GO synthesized from recovered carbon black (rCB) using varying concentrations of $KMnO_4$, as presented in Figure 3. The FESEM image of the raw rCB sample (Figure 3(a)) displays aggregated, spherical-shaped particles, consistent with the morphology reported by Paul et al [16]. This structure is characteristic of unmodified carbon black obtained from pyrolyzed waste tires, indicating minimal oxidation or layer separation.

In Figure 3(b), the GO-1 sample synthesized with 1 g of $KMnO_4$ shows a morphology that remains largely spherical and agglomerated, closely resembling the rCB structure. This suggests that a $KMnO_4$ concentration of 1 g was insufficient to initiate significant oxidation or exfoliation of rCB into GO. With increasing oxidant concentrations, noticeable morphological changes were observed. The GO-3 (Figure 3(c)) and GO-5 (Figure 3(d)) samples exhibit a more densely packed and agglomerated structure, indicating the progression of oxidation. These morphological changes are consistent with the XRD results, where the emergence of new diffraction peaks at lower angles ($2\theta = 11.23°$ and $10.65°$) confirmed an increase in interlayer spacing, reflecting the incorporation of oxygen functional groups and partial exfoliation of the GO sheets. The particles appear less spherical and exhibit sheet-like deformation, representing the onset of exfoliation and formation of layered GO structures. This observation aligns with Dziejarski et al. [11], who reported that during oxidation, carbon black particles begin to coalesce and form elongated, chain-like networks and prolate spheroids, as local graphitic domains reorient into more planar and partially ordered arrangements. GO-5, a well-oxidized and partially exfoliated structure with thin, wrinkled sheets, was observed, resulting in sheet fragmentation. The presence of folded edges further confirms the effective disruption of the carbon framework and the introduction of oxygen-containing groups, facilitating the delamination of GO layers. These morphological changes provide strong evidence that higher $KMnO_4$ concentrations enhance oxidation and exfoliation, leading to the structural transformation of rCB into GO nanosheets.

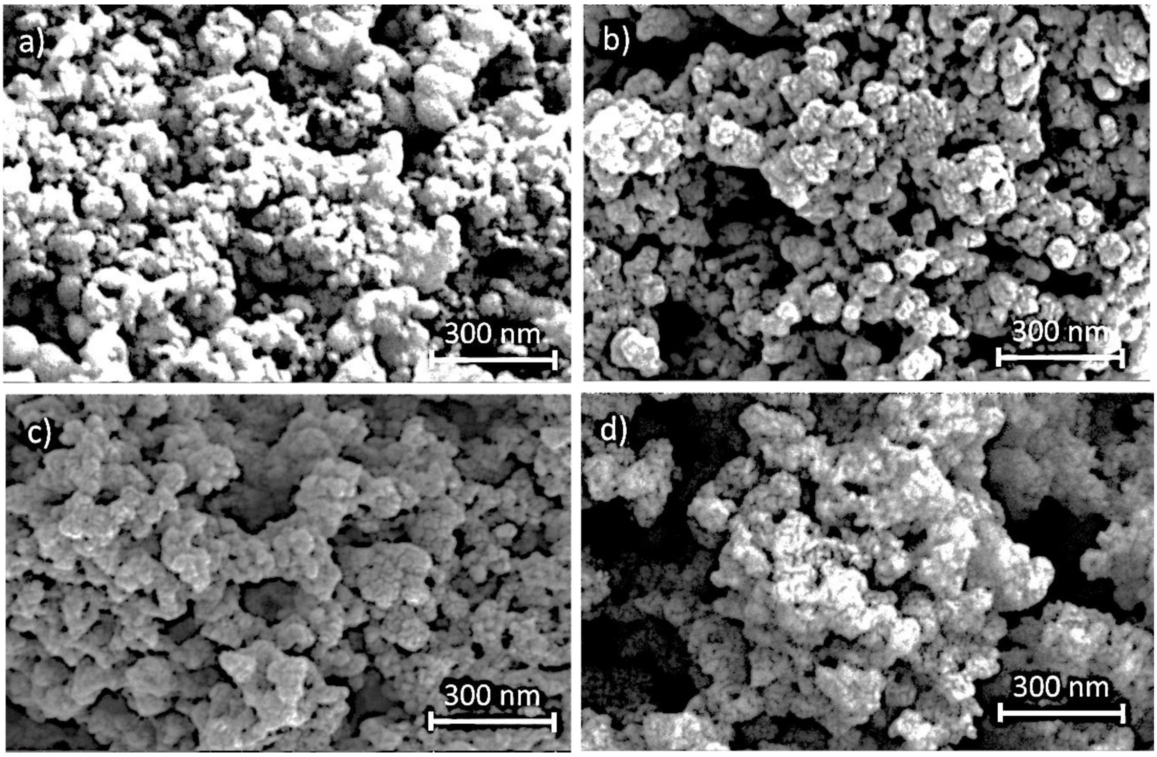
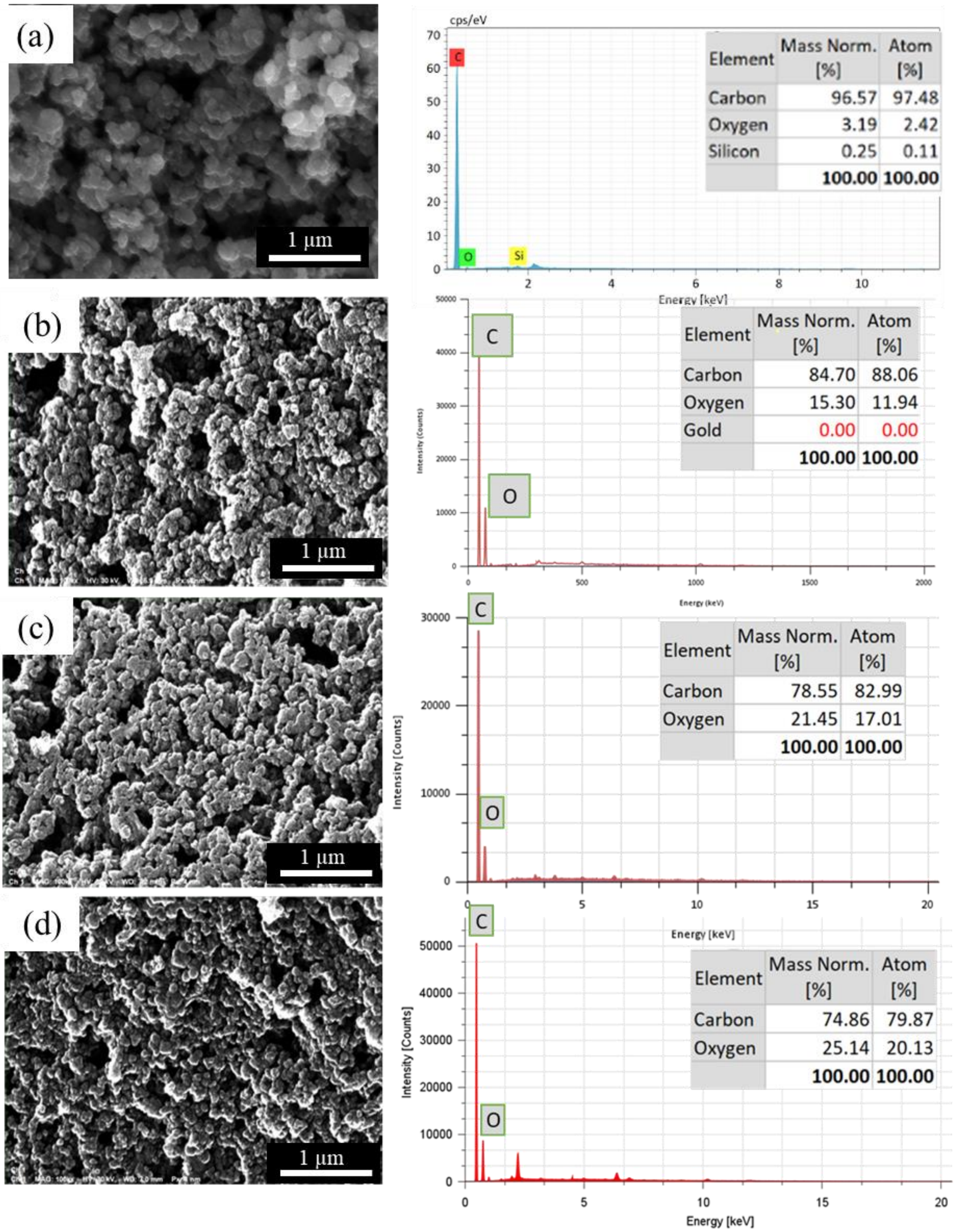


Figure 3: FESEM images of (a) rCB, (b) GO-1, (c) GO-3, and (d) GO-5.

Energy-dispersive X-ray spectroscopy (EDX) was conducted to examine the elemental composition of graphene oxide (GO) samples synthesized using varying concentrations of $KMnO_4$, as shown in Figure 4. The analysis provides insight into the extent of oxidation by quantifying the relative proportions of carbon and oxygen in the samples [17]. Figure 4(a) shows the EDX of rCB with the carbon content (C) of 96.57 wt%.

Upon the oxidation with 1 g $KMnO_4$, the EDX spectrum of GO-1 (Figure 4(b)) shows a decrease in the carbon content to 84.70 wt%, while the oxygen content was increased to 15.30 wt%. This relatively high carbon-to-oxygen ratio suggests a low degree of oxidation [18], indicating that the oxidant concentration was insufficient to introduce a substantial number of oxygen-containing functional groups onto the rCB structure. For the GO-3 sample (Figure 4(c)), the oxygen content increased to 21.45 wt%, while the carbon content decreased to 78.55 wt%. This change reflects enhanced incorporation of oxygen-containing groups such as hydroxyl, epoxy, and carboxyl moieties, signifying more effective oxidation of the carbon matrix compared to GO-1.

The trend continues in the GO-5 sample (Figure 4(d)), where the highest oxidant concentration of 5 g $KMnO_4$ yielded oxygen content of 25.14 wt% and a reduced carbon content of 74.86 wt%. This increase in oxygen content demonstrates a greater degree of surface oxidation, confirming that higher $KMnO_4$ concentrations promote more efficient oxidation of rCB to GO. Thus, the EDX results corroborate the findings from FESEM and XRD analyses, highlighting that increasing the $KMnO_4$ concentration leads to higher oxygen incorporation and thus a greater degree of chemical modification in the synthesized GO. This trend is consistent with the progressive formation of oxygen-rich functional groups that facilitate the exfoliation and transformation of rCB into GO.



Element	Mass Norm. [%]	Atom [%]
Carbon	96.57	97.48
Oxygen	3.19	2.42
Silicon	0.25	0.11
	100.00	**100.00**

Element	Mass Norm. [%]	Atom [%]
Carbon	84.70	88.06
Oxygen	15.30	11.94
Gold	0.00	0.00
	100.00	**100.00**

Element	Mass Norm. [%]	Atom [%]
Carbon	78.55	82.99
Oxygen	21.45	17.01
	100.00	**100.00**

Element	Mass Norm. [%]	Atom [%]
Carbon	74.86	79.87
Oxygen	25.14	20.13
	100.00	**100.00**

Figure 4: EDX analysis of (a) rCB, (b) GO-1, (c) GO-3, and (d) GO-5.

4. CONCLUSIONS

This study successfully demonstrated that the concentration of potassium permanganate ($KMnO_4$) plays a critical role in governing the structural transformation and morphological characteristics of graphene oxide (GO) synthesized from recovered carbon black (rCB) derived from waste tires. Increasing $KMnO_4$ concentration facilitated greater oxidation and exfoliation, as evidenced by the shift of XRD peaks to lower angles, the formation of oxygen-containing functional groups in FTIR spectra, and the morphological evolution from spherical aggregates to thin, wrinkled GO sheets in FESEM analysis. The findings confirm that controlling the oxidant concentration enables effective tuning of GO properties, providing a sustainable and scalable route for transforming waste-derived carbon into high-value nanomaterials. GO synthesized at higher $KMnO_4$ concentrations, particularly 5 g, exhibited characteristics most consistent with well-oxidized and structurally defined GO, making it suitable for further functional and application-specific studies.

Acknowledgements

The authors gratefully acknowledge the financial support provided by Universiti Teknologi MARA (UiTM) through the Geran Insentif Penyeliaan (GIP), Grant Code: 600-RMC/GIP 5/3 (015/2024). The authors also sincerely thank the Faculty of Applied Sciences, UiTM Shah Alam, for the use of research equipment and laboratory facilities essential to this study.

Author Contributions

All authors contributed toward data analysis, drafting, and critically revising the paper and agree to be accountable for all aspects of the work.

Disclosure of Conflict of Interest

The authors have no disclosures to declare

Compliance with Ethical Standards

Not Applicable

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