# Synthesis of Graphene Oxide via the Tour Method and Its Structural Characterization

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**Abstract:** Graphene oxide (GO) has emerged as one of the most versatile two-dimensional materials owing to its tunable structure and rich surface chemistry. However, the controlled synthesis of GO with high oxygen functionality and desirable morphology remains a key challenge. In this work, Graphene oxide (GO) was successfully synthesized from graphite using the modified Tour method. The synthesis process, which required approximately 7 h, was followed by freeze-drying for 24 h to obtain stable nanosheets. The structural and chemical features of the obtained GO were examined by XRD, FTIR, and TEM analyses. XRD revealed a prominent diffraction peak at $2\theta \approx 11.2°$, corresponding to an interlayer spacing of ~0.79 nm, which is more than twice that of pristine graphite, confirming efficient oxidation and intercalation of oxygen functionalities. FTIR spectra further identified hydroxyl, epoxy, carbonyl, and carboxyl groups, providing strong evidence of successful oxidation. TEM micrographs displayed thin, wrinkled sheets with transparent regions, characteristic of few-layer GO. These findings demonstrate that the adopted synthesis route enables the production of GO with expanded interlayer spacing, abundant active sites, and flexible sheet-like morphology. Such properties make the material particularly attractive for energy-related applications, including photocatalysis and hydrogen production under visible light.

**Keywords** Graphene oxide (GO); interlayer spacing; oxygen functionalities; photocatalysis; hydrogen energy

# Синтез Оксида Графена Методом Тура и Его Структурная Характеристика

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**Аннотация:** Оксид графена (GO) стал одним из самых универсальных двумерных материалов благодаря своей настраиваемой структуре и богатой поверхностной химии. Однако контролируемый синтез GO с высокой кислородной функциональностью и желаемой морфологией остается ключевой проблемой. В этой работе оксид графена (GO) был успешно синтезирован из графита с использованием модифицированного метода Тура. Процесс синтеза, который занял приблизительно 7 часов, сопровождался сушкой вымораживанием в течение 24 часов для получения стабильных нанолистов. Структурные и химические особенности полученного GO были исследованы с помощью анализов XRD, FTIR и TEM. XRD выявил выраженный дифракционный пик при $2\theta \approx 11{,}2°$, соответствующий межслоевому расстоянию ~ 0,79 нм, что более чем в два раза больше, чем у исходного графита, подтверждая эффективное окисление и интеркаляцию кислородных функциональных групп. Спектры FTIR дополнительно идентифицировали гидроксильные, эпоксидные, карбонильные и карбоксильные группы, что предоставило убедительные доказательства успешного окисления. Микрофотографии, полученные в просвечивающем электронном микроскопе, демонстрируют тонкие, морщинистые слои с прозрачными областями, характерные для малослойного оксида графена (GO). Эти результаты показывают, что выбранный метод синтеза позволяет получать GO с увеличенным межслоевым расстоянием, обилием активных центров и гибкой листовой

морфологией. Такие свойства делают материал особенно привлекательным для энергетических приложений, включая фотокатализ и производство водорода в видимом свете.

**Ключевые слова:** оксид графена (GO); межслоевое расстояние; кислородные функциональные группы; фотокатализ; водородная энергетика

# Grafen Oksidining Tur Usuli bilan Sintezi va Uning Strukturaviy Xarakteristikasi

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**Annotatsiya:** Grafen oksidi (GO) sozlanishi tuzilishi va boy sirt kimyosi tufayli eng ko'p qirrali ikki o'lchovli materiallardan biriga aylandi. Biroq, yuqori kislorod funksionalligi va kerakli morfologiyasi bilan GO ning boshqariladigan sintezi asosiy muammo bo'lib qolmoqda. Ushbu ishda grafen oksidi (GO) o'zgartirilgan Tur usuli yordamida grafitdan muvaffaqiyatli sintez qilindi. Taxminan 7 soat davom etgan sintez jarayoni barqaror nano-varaqlarni olish uchun 24 soat davomida muzlatish bilan quritilgan. Olingan GO ning strukturaviy va kimyoviy xususiyatlari XRD, FTIR va TEM tahlillari yordamida tekshirildi. XRD 2th ≈ 11,2 ° da sezilarli diffraktsiya cho'qqisini aniqladi, bu qatlamlar orasidagi masofa ~ 0,79 nmga to'g'ri keladi, bu toza grafitdan ikki baravar ko'p, kislorod funktsional guruhlarining samarali oksidlanishi va interkalatsiyasini tasdiqlaydi. FTIR spektrlari qo'shimcha ravishda gidroksil, epoksi, karbonil va karboksil guruhlarini aniqladi, bu muvaffaqiyatli oksidlanishning ishonchli dalillarini taqdim etdi. Transmissiya elektron mikrograflari bir necha qatlamli grafen oksidi (GO) ga xos shaffof hududlarga ega nozik, ajin qatlamlarini namoyish etadi. Bu natijalar shuni ko'rsatadiki, tanlangan sintez usuli qatlamlar oralig'ining ortishi, faol joylarning ko'pligi va moslashuvchan varaqga o'xshash morfologiyasi bilan GO hosil qiladi. Ushbu xususiyatlar materialni energiya uchun qo'llash, jumladan, fotokataliz va ko'rinadigan yorug'lik vodorod ishlab chiqarish uchun jozibador qiladi.

**Kalit so'zlar:** grafen oksidi (GO); qatlamlararo masofa; kislorod funktsional guruhlari; fotokataliz; vodorod energiyasi.

**Introduction.** Carbon nanomaterials possess an exceptionally wide range of practical applications, spanning from medicine[1] to electrochemistry[2]. Their multifunctionality primarily arises from the peculiarities of their electronic structure, which enable the development of 2D and 3D functional materials (such as graphene and multilayered structures) with unique physicochemical properties[3]. Graphene is a two-dimensional, one-atom-thick allotrope of carbon, consisting of $sp^2$-hybridized carbon atoms arranged in a hexagonal lattice. Despite its outstanding properties, the processability and large-scale production of highly exfoliated, pure graphene remain a significant challenge, as this issue has persisted for a long time. Consequently, research efforts have increasingly shifted toward alternative materials[4].

However, large-scale production of these materials remains challenging and costly. In this context, reduced graphene oxide can serve as a cost-effective alternative to graphene, particularly for the preparation of nanocomposite materials[5]. One of the closest alternatives to graphene is reduced graphene oxide (RGO). Notably, RGO is a derivative of graphene, and its scalable production is typically achieved through the exfoliation and subsequent reduction of graphene oxide (GO).

The most widely used methods for the synthesis of graphite oxide are the Brodie method[6], Staudenmaier method[7], Hofmann method[8], and Hummers method[9], along with their various modified and improved versions. In these approaches, graphite powder is first chemically treated with acids, followed by the intercalation of alkali metals or their compounds into the graphitic layers, which

facilitates the subsequent breaking of these layers into smaller fragments. During the oxidation process, hydroxyl, epoxy, and carboxyl functional groups are incorporated onto the surface of graphene layers, thereby improving the dispersibility of the material and creating favorable conditions for subsequent chemical functionalization.

In the synthesis of graphene oxide, the Tour method was employed. While the Hummers method is based on conventional oxidizing systems, the Tour method utilizes a combination of strong oxidants, enabling the production of highly oxidized and well-dispersed graphene oxide. This approach promotes the formation of a greater number of oxygen-containing functional groups in graphene oxide, thereby enhancing its potential for subsequent chemical processing and broadening its application prospects.

The Tour method employed for the synthesis of graphene oxide (GO) offers several advantages over conventional approaches. The process was completed within only 7 hours, which is significantly shorter compared to the traditional Hummers method (12–24 hours), thereby ensuring savings in both time and energy. As a result of the synthesis, graphite residues were minimized, and highly oxidized GO was obtained. During the drying stage, the use of a freeze-dryer prevented the restacking of GO layers, thereby preserving the morphology and yielding a highly dispersible powder. Moreover, this method helped maintain the stability of oxygen-containing functional groups.

Overall, the applied synthesis and drying conditions demonstrate superior efficiency, time savings, and product quality compared to methods reported in the literature.

**Experimental Section**

**Materials and chemicals**

All chemicals used in this study were purchased from Sigma-Aldrich and used without further purification. Graphite flakes [99.8%, natural graphite], sulfuric acid (98%), potassium permanganate, ortho-phosphoric acid, hydrogen peroxide [30% (w/v)], hydrochloric acid, were used as starting materials for the synthesis of GO by the Tour method.

**Synthesis of Copper(I) Oxide Nanoparticles**

In this study, graphene oxide was synthesized using the Tour method. The synthesis procedure involved the following steps: Graphite was dispersed in a mixture of sulfuric acid ($H_2SO_4$) and phosphoric acid ($H_3PO_4$) at a ratio of 9:1. Potassium permanganate ($KMnO_4$) was then gradually added to the mixture at 0 °C (ice bath) under continuous stirring. After 3 hours of stirring, the reaction mixture was heated in an oil bath to 50 °C and maintained at this temperature for 2 hours. Upon completion of the oxidation process, hydrogen peroxide ($H_2O_2$) was added to remove the excess permanganate. The resulting product was repeatedly washed with hydrochloric acid solution (HCl, 5%) and deionized water until a neutral pH was obtained. The purified precipitate was filtered and dried using a freeze dryer. X-ray diffraction (XRD) analysis confirmed the successful formation of graphene oxide (GO). In addition, transmission electron microscopy (TEM) revealed the layered sheet-like morphology of GO, while the FT-IR spectra clearly indicated the presence of oxygen-containing functional groups. Together, these characterization techniques verify the successful synthesis and structural integrity of graphene oxide. The synthesis process is illustrated in Figure 1

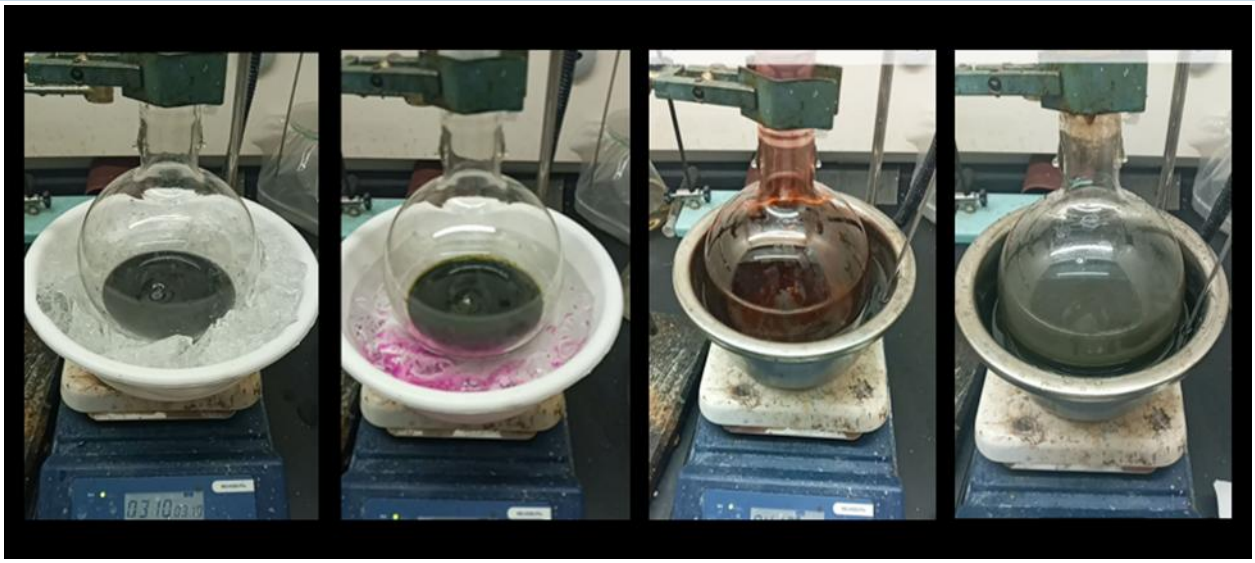

**Figure 1.** Photographs describing the preparation process of GO by Tour's method: (A) before addition of potassium permanganate; (B) after oxidation; (C) after pouring on ice; (D) after addition of $H_2O_2$.

**Results and Discussion**

**X-ray diffraction (XRD) analysis**

The crystalline structures of pristine graphite and graphene oxide (GO) were investigated by X-ray diffraction (Fig. X). The diffraction pattern of graphite exhibits a sharp and intense peak at 2θ ≈ 26.4°, corresponding to the (002) reflection, with an interlayer spacing of d(002) ≈ 0.34 nm, as calculated by Bragg's equation. This value is consistent with the well-ordered, closely packed graphitic layers reported in the literature.

In contrast, the diffraction pattern of GO shows the disappearance of the graphite (002) peak and the appearance of a new reflection at 2θ ≈ 11.2°, which can be indexed to the (001) plane. The corresponding interlayer spacing is calculated using **Bragg's law**:

$$n\lambda = 2d\sin\theta$$

where $n$ is the diffraction order (taken as 1), $\lambda$ is the wavelength of the Cu Kα radiation (0.15406 nm), and $\theta$ is the Bragg angle. For graphite, the calculated interlayer spacing is:

$$d_{001} = \frac{\lambda}{2\sin\theta} = \frac{0.15406}{2\sin(5.6°)} \approx 0.80\ nm$$

This expanded interlayer distance, more than twice that of pristine graphite, is attributed to the introduction of oxygen-containing functional groups (–OH, –COOH, –O–) and the intercalation of water molecules between the graphene sheets during the oxidation process.

Thus, the disappearance of the characteristic (002) reflection of graphite and the emergence of the (001) peak of GO provide clear evidence for the successful oxidation of graphite into graphene oxide.

In the literature, the interlayer spacing of GO samples synthesized by various methods is typically reported to be in the range of 0.70–0.85 nm (based on the Hummers method and its modifications)[10, 11]. Our result fully corresponds to this range, indicating the successful synthesis of GO.

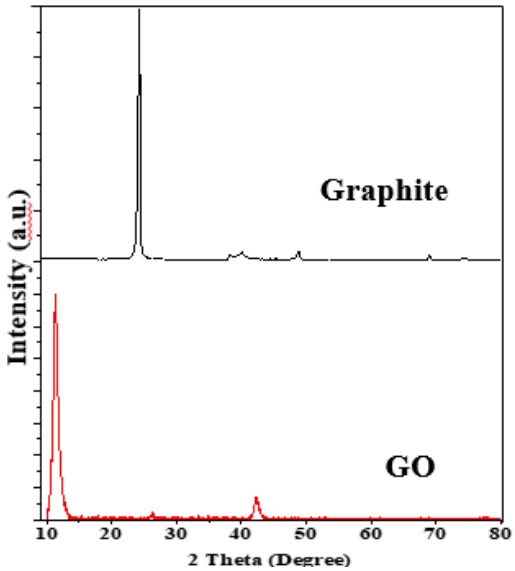


**Figure 2**. X-ray structures of graphite and GO

**FTIR Analysis:**

The chemical structure of graphene oxide (GO) was investigated using FTIR spectroscopy (Figure 3). The spectrum exhibited a broad peak at ~3200 $cm^{-1}$, corresponding to hydroxyl (–OH) groups and adsorbed water molecules. A strong band observed at ~1720 $cm^{-1}$ was assigned to the C=O stretching vibration, indicating the presence of carboxyl and carbonyl groups. The peak at ~1620 $cm^{-1}$ was attributed to the skeletal vibrations of aromatic C=C bonds. At lower frequencies (~1200–1050 $cm^{-1}$), absorption bands associated with C–O–C (epoxy) and C–O (alkoxy) stretching vibrations were detected, while peaks around ~870–900 $cm^{-1}$ correspond to out-of-plane vibrations of aromatic C–H bonds.

These results confirm the presence of hydroxyl, carbonyl, carboxyl, and epoxy groups in the GO structure. This provides evidence of the successful oxidation of graphite and is consistent with the XRD analysis, which revealed an expansion of the interlayer spacing due to the incorporation of oxygen-containing functional groups and intercalated water molecules.

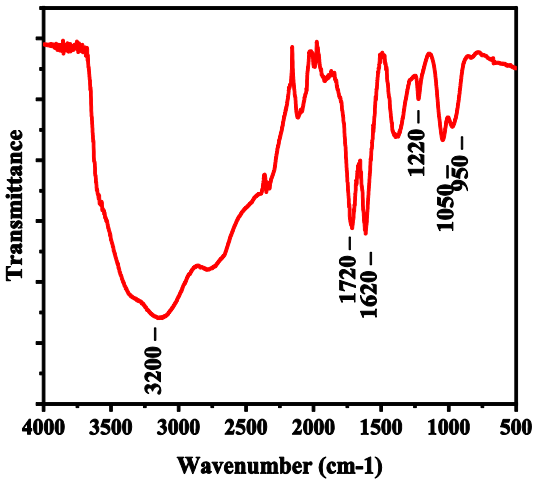


**Figure 3.** FT-IR spectrum of graphene oxide (GO)

**Morphological and Elemental Analysis**

The morphology and chemical composition of the synthesized graphene oxide (GO) were investigated using transmission electron microscopy (TEM) and energy-dispersive X-ray spectroscopy (EDS). Fig. 4 (a) presents the TEM image of GO, which clearly shows thin, wrinkled, and semi-transparent sheet-like structures, characteristic of exfoliated nanosheets. The darker contrasts correspond to overlapping multilayers, whereas the lighter regions indicate few-layer or monolayer GO sheets. The nanosheets exhibit lateral dimensions ranging from several hundred nanometers up to a few micrometers. The observed wrinkles and folds are attributed to the disruption of the $sp^2$ carbon lattice by oxygen-containing functional groups.

Fig. 4 (b) displays the EDS spectrum, which demonstrates dominant peaks corresponding to carbon (C, ~0.3 keV) and oxygen (O, ~0.5 keV). The quantitative results, summarized in Table 1, show high atomic fractions of carbon (73.69%) and oxygen (18.37%), confirming the effective oxidation of graphite and the successful formation of graphene oxide. Minor peaks of Si, S, and Cu were also detected, originating from residual reagents or the supporting grid, but these do not belong to the intrinsic composition of the material.

Overall, the combined TEM and EDS results verify that the obtained material consists of thin, layered nanosheets with abundant oxygen functionalities, providing high surface area and active sites, which make GO a promising candidate for photocatalytic and energy-related applications.

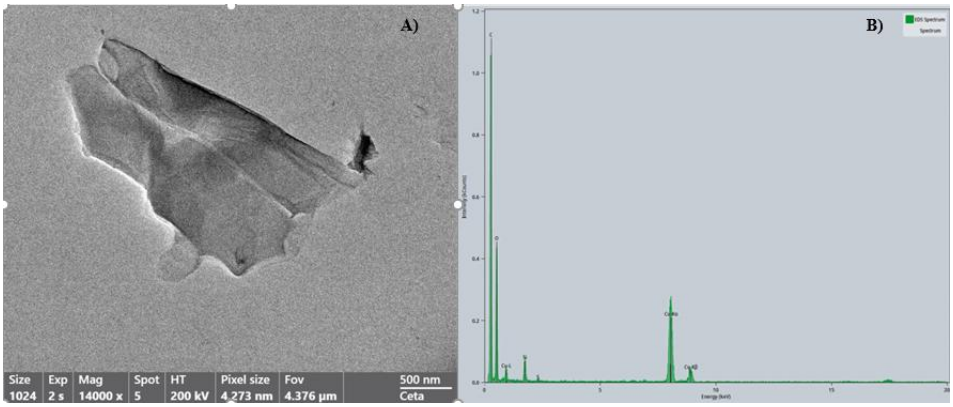


**Figure 4.** a) TEM image of graphene oxide (GO) showing thin and layered sheet-like morphology, b) EDS spectrum of GO showing dominant peaks of carbon (C, ~0.3 keV) and oxygen (O, ~0.5 keV), confirming the successful formation of graphene oxide.

**Table 1.** Elemental composition obtained from EDS, showing high contents of carbon (C) and oxygen (O), which are consistent with the structural characteristics of graphene oxide (GO).

Analysis of EDS Spectrum C)

| Z | Element | Family | Atomic (%) | Fraction | Atomic (%) | Error | Mass (%) | Fraction | Mass Error (%) | Fit Error (%) |
|---|---|---|---|---|---|---|---|---|---|---|
| 6 | C | K | 73.69 | | 2.85 | | 54.13 | | 2.86 | 2.64 |
| 8 | O | K | 18.37 | | 3.03 | | 17.98 | | 3.03 | 0.47 |
| 14 | Si | K | 1.24 | | 0.24 | | 2.12 | | 0.42 | 1.19 |
| 16 | S | K | 0.13 | | 0.03 | | 0.26 | | 0.05 | 5.16 |
| 29 | Cu | K | 6.57 | | 0.92 | | 25.51 | | 2.90 | 0.19 |

## Conclusion

Graphene oxide (GO) was successfully synthesized from graphite using the modified Tour method. Structural analyses, including XRD, FT-IR, and TEM, confirmed the effective oxidation of graphite, the incorporation of oxygen-containing functional groups, and the formation of layered nanosheets. Compared to conventional methods, this approach offers shorter synthesis time, higher oxidation efficiency, and better preservation of morphology through freeze-drying. In addition, energy-dispersive X-ray spectroscopy (EDS) analysis further verified the chemical composition of the synthesized GO. The spectrum displayed dominant peaks corresponding to carbon (C) and oxygen (O), with atomic fractions of 73.69% and 18.37%, respectively. These results strongly confirm the presence of oxygen functionalities within the carbon framework, supporting the structural features of graphene oxide. Overall, the obtained GO, characterized by a large surface area, stable oxygen functionalities, and high purity as confirmed by EDS, exhibits strong potential for energy-related applications such as photocatalysis and hydrogen production.

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