



## Preparation of Graphene Oxide from Expanded Graphite at Different Microwave Heating Times

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### Highlights:

- Graphene oxide (GO) was successfully prepared from exfoliated graphite (EG).
- The maximum interplanar distance (d-spacing) of the GO increased from 0.344 to 0.831 nm.
- Sonication increase d-spacing between the GO layers.

**Abstract.** Exfoliated graphite (EG) was prepared by mixing graphite with  $\text{HNO}_3$  and  $\text{KMnO}_4$  at weight ratio 1:2:1 using microwave heating at times 20, 60, 80 and 120 sec. Graphene oxide (GO) was then prepared using EG as precursor by the modified Hummer's method. Atomic force microscopy (AFM), X-ray diffraction (XRD) and Fourier transform infrared (FTIR) spectroscopy analyses showed successful conversion of EG into GO. The XRD results of the GO showed that the maximum interplanar distance (d-spacing) increased from 0.344 to 0.831 nm. The AFM showed a minimum thickness of 0.519 nm for a single layer of GO prepared from EG 80 sec. The XRD examination also showed an increase in the d-spacing between the GO layers after sonication compared to before sonication.

**Keywords:** *exfoliated graphite; graphite intercalation compounds; graphene oxide; Hummer's method; graphite layers; microwave heating.*

## 1 Introduction

One of the most common atoms on earth is carbon, which forms allotropes of carbon [1]. The chemical, physical and mechanical properties of the allotropes of carbon depend on valence bond hybridization, which are hybridized to form  $sp$ ,  $sp^2$  and  $sp^3$  chemical bonds between atoms. Allotropes of carbon can be defined by valence bond hybridization  $sp^n$  [2]. There are five allotropes of carbon, namely graphite, diamond, graphene, carbon nanotubes and fullerene [3]. Graphite is an important allotrope of carbon [4]. It is a 3D stacked structure with parallel layers,

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where in each layer the carbon atoms are covalently bonded. These parallel layers are stacked and bonded by weak Van der Waals forces along the perpendicular axis to the basal plane [5]. Natural graphite has been used as host material to synthesize graphite intercalation compounds (GIC) [6].

Microwaves transfer their energy directly to target materials by radiation at the speed of light without conduction or convection [7]. Microwave heating (MW) can heat a material more uniformly compared to conventional heating. Thus, MW synthesis has been proven as a fast, energy efficient, uniform, and scalable approach [8]. MW is a good heating method, without mechanical stirring and energy loss to the environment [9]. Thus, it can be used to a small number of layers of graphene oxide [10]. The formulation of EG has been done by rapid heating of graphite or intercalation compounds [11]. When graphite layers are oxidized, they will open gradually due to the molecules or ions entering gases into the interspace of the graphite layers. During heating, the released volatile gases make GIC expand to produce EG [6,11]. The quantitative characterization of the expansion of graphite in MW-assisted exfoliation depends on the use of HNO<sub>3</sub> as intercalated ions and KMnO<sub>4</sub> as a highly oxidant agent. Agent molecules of HNO<sub>3</sub> migrate and deposit between the graphene layers of the graphite. Then, the intercalation agent molecules are vaporized under microwave heating in the intercalation and exfoliation processes and generate enough force to overcome the Van der Waals forces between the graphite layers. Moreover, the characteristics of expanding graphite in MW-assisted exfoliation depend on the weight ratio between graphite, KMnO<sub>4</sub>, HNO<sub>3</sub> and mixing time [12]. Graphite intercalation compounds synthesize expanded graphite from intercalation of graphite [13]. Thus, using MW is a rapid and efficient approach to the synthesis of the EG materials.

This process is accompanied with lightening and fuming [14]. Graphene oxide sheets are a layered material produced by the oxidation of graphite structures [15]. The surface of GO contains large numbers of hydrophilic functional groups, such as carbonyl, carboxyl, epoxy, hydroxyl and GO, that are structurally similar to graphene [15,16]. Thus, a graphene oxide layer can be as thin as a single carbon layer. GO has oxygen functional groups that are bonded to the surface [17].

GO can be used for water purification, super capacitors and as a composite in antibacterial activity, coatings and solar cells [1]. The aim of the present study was to prepare EG at different microwave heating times. After that, GO was prepared using EG as precursor by modified Hummers method. The effect of ultrasonication on GO preparation was studied.

## **2 Materials and Experimental work**

### **2.1 Materials**

Natural graphite powder (99.8%, Sigma, Germany), nitric acid ( $\text{HNO}_3$ , 69%), concentrated sulphuric acid ( $\text{H}_2\text{SO}_4$ , 98%, Romil, UK), potassium permanganate ( $\text{KMnO}_4$ ), hydrochloric acid ( $\text{HCl}$ , 37%, CDH), hydrogen peroxide ( $\text{H}_2\text{O}_2$ , 30%, GMBH, Germany) were used for the preparation of EG, after which graphene oxide was prepared from the EG.

### **2.2 Preparation of Expanded Graphite by Microwave Irradiation**

Expanded graphite was prepared by mixing graphite powder (1 g) with concentrated  $\text{HNO}_3$  (2 g) and  $\text{KMnO}_4$  (1 g) at a weight ratio of 1:2:1 [14]. The mixture was then stirred by hand in a glass flask at room temperature for 20 minutes. The glass flasks with the mixtures were then put into a domestic microwave oven for heating for 20, 60, 80 and 120 sec to obtain four types of expanded graphite. The completeness of the graphite exfoliation process was confirmed by the disappearance of lightning and fuming, and the formation of gases. After the microwave heating process, each one of the obtained expanded graphite samples was washed using distilled water for several times until the pH reading was 7. Thereafter, the wet expanded graphite was filtered and then dried at 110 °C for six h to obtain expanded graphite (EG) powder.

### **2.3 Preparation of GO Using Expanded Graphite**

Graphene oxide (GO) was prepared by modified Hummer's method. The prepared EG (1 g) was mixed with  $\text{H}_2\text{SO}_4$  (20 ml) by magnetic stirrer for 30 min. Then, the resulting mixture was transferred to an ice bath at a mixture temperature of (10-14 °C) to avoid a rapid increase in temperature. Under magnetic stirring,  $\text{KMnO}_4$  (3 g) was slowly added to the mixture for 20 min. The resultant mixtures had a green or (dark blue) color [18]. The dark blue mixture was transferred to a water bath at (35-45 °C) and stirred with a magnetic stirrer for 30 min, when this mixture became a pasty stack. Afterwards, distilled water (50 ml) was slowly added with continuous stirring; this step produces crackles with toxic gases. The resulting mixture was transferred to a water bath at (80-98 °C) and stirred for 15 min while waiting for the dispersion color to become brown. Thereafter, 140 ml of distilled water was added, with continuous magnetic stirring of the mixture for 15 min. Subsequently, the reactions were terminated by adding 15 ml of hydrogen peroxide ( $\text{H}_2\text{O}_2$  30%) with continuous stirring for one hour, until the color of the dispersion became bright yellow. Throughout this work the notations following will be used: EG prepared by microwave heating at 700 W, 20 sec was used to prepare GO 20 sec; EG prepared by microwave heating at 700 W, 60 sec was used to prepare GO 60 sec; EG prepared by microwave heating at 700 W, 80 sec

was used to prepare GO 80 sec; EG prepared by microwave heating at 700 W, 120 sec was used to prepare GO 120 sec.

#### 2.4 Washing and Drying to Prepare GO

Several washing steps were necessary to eliminate impurities and oxidizing agents, which occurred prior to the oxidation and exfoliation. Upon the end of the oxidation and exfoliation reactions of graphite using H<sub>2</sub>O<sub>2</sub> 30%, two liters of distilled water were added to the GO mixture, which was left to settle for 24 hours. The water was poured out and the GO residual was dried at 50-60 °C. The dry precipitate of GO was washed with 100 ml of diluted HCl 10% to remove metal ions, with magnetic stirring for 10 minutes and vacuum filtering. Then, the wet precipitate of GO was washed with two liters of distilled water without vacuum filtration, because GO solution is sticky and will close the filter pores, which makes filtration difficult. Therefore, the wet GO was dried in an oven at 50 °C. The above washing, separation, purification and filtration processes are economical and easy to use [19]. Figure 1 shows the prepared GO in the present study under sunlight. The GO obtained was dispersed in distilled water (0.5 mg/ml) followed by ultra-sonication for one hour, in order to increase expansion and exfoliation by ultra-sonication and to generate GO nano layers.



Figure 1 Prepared graphene oxide (GO).

### 3 Results and Discussion

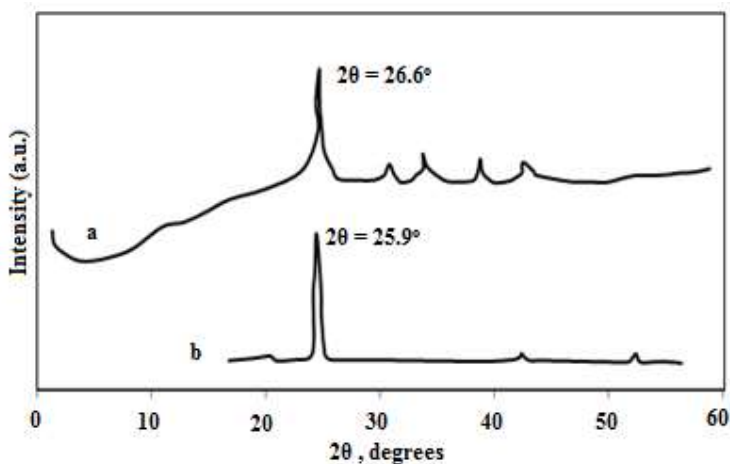
The prepared GO was characterized by X-Ray diffractometer (XRD) (MiniFlex II, Rigaku Co., Japan), Fourier transform infrared (FTIR) spectroscope (Prestige 21 Shimadzu Co., Japan) and atomic force microscope (AFM) (model: NT-MDT Ntegra, Russian Federation) to study the GO before and after ultra-sonication.

#### 3.1 X-Ray Diffraction (XRD) of Expanded Graphite

Expanded graphite (EG) was prepared by mixing graphite with HNO<sub>3</sub> and KMnO<sub>4</sub> at a weight ratio of 1:2:1. The mixtures were heated using a domestic microwave oven (700W) at different heating times. Figure 2(a) shows the XRD

## Preparation of Graphene Oxide from Expanded Graphite

results of the pristine graphite with a peak at  $2\theta = 26.6^\circ$  for plane 002, which corresponds to an interlayer distance of 0.338 nm [19]. The XRD pattern of the prepared EG 60 sec confirmed the crystalline structure of EG with a sharp peak at  $2\theta = 25.9^\circ$  corresponding to a d-spacing of 0.344 nm in plane 002, as shown in Figure 2(b).



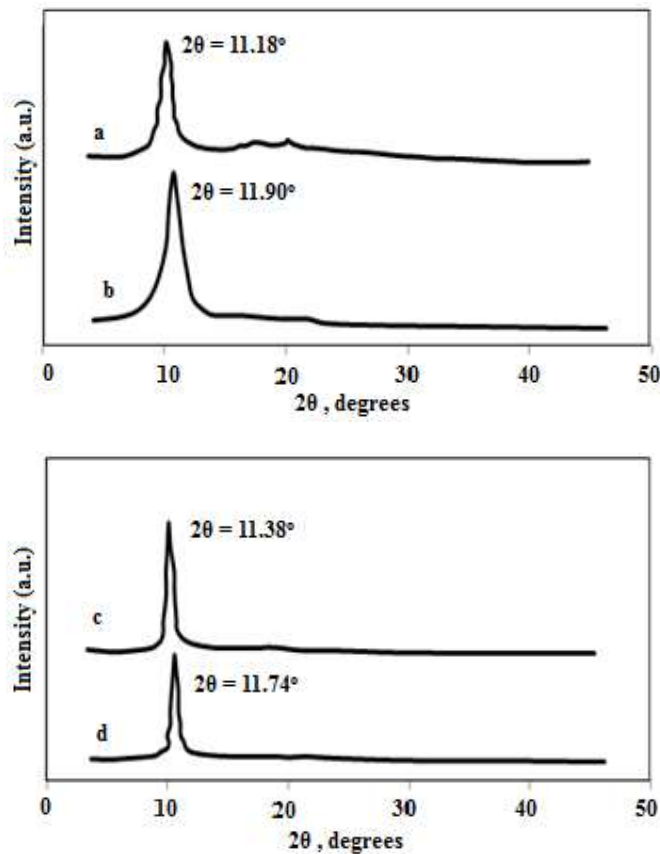
**Figure 2** XRD patterns of (a) pristine graphite [19]; (b) expanded graphite (EG).

### 3.1.1 XRD of GO Before Ultrasonication

The X-ray diffraction patterns (XRD) of the prepared GO from exfoliated graphite (EG) were measured before sonication, which showed that the EG peak had disappeared and a new peak for GO had appeared. Figure 3(a) shows the XRD pattern of the prepared GO 20 sec with a new sharp peak at  $2\theta = 11.18^\circ$  and d-spacing at 0.79 nm. Figure 3(b) shows the XRD pattern of the prepared GO 60 sec with a new sharp peak at  $2\theta = 11.90^\circ$  and d-spacing at 0.743 nm. Figure 3(c) shows the XRD pattern of the prepared GO 80 sec with a new sharp peak at  $2\theta = 11.38^\circ$  and d-spacing at 0.78 nm. Figure 3(d) shows the XRD pattern of the prepared GO 120 sec with a new sharp peak at  $2\theta = 11.74^\circ$  and d-spacing at 0.75 nm. These results confirm the complete conversion of the expanded graphite into graphene oxide. Additionally, the absence of impurity peaks explains the high purity of the prepared GO, as shown in Figure 3.

Oxidation of EG to GO occurred due to oxygen-group intercalation on the inner and outer surfaces of the EG, which leads to a loose stack of GO sheets [20]. During the steps of oxidation of graphite, the graphite sheets react with oxygen functional groups, which increases the distance between the layers in accordance

with the degree of oxidation of the EG and the quantity of molecules inserted into the interlayer spacing [21,22].



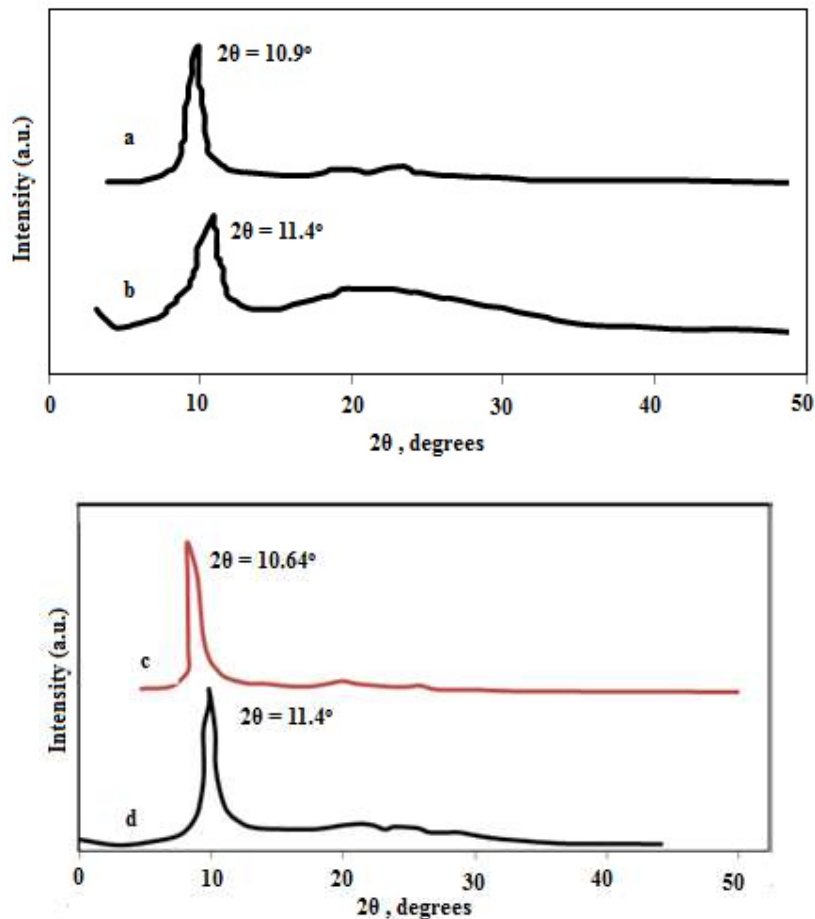
**Figure 3** XRD patterns of the prepared GO: (a) GO 20 sec; (b) GO 60 sec; (c) GO 80 sec; (d) GO 120 sec.

### 3.1.2 XRD of GO After Ultrasonication

About 0.5 mg/mL of prepared GO was sonicated in an ultra-sonication bath for one hour in distilled water. Figure 4(a) shows the X-ray diffraction patterns (XRD) of the sonicated GO 20 sec with a sharp peak at  $2\theta = 10.9^\circ$  and d-spacing at 0.81 nm. For the sonicated GO 60 sec, the sharp peak was at  $2\theta = 11.4^\circ$  and d-spacing at 0.76 nm, as shown in Figure 4(b). For the sonicated GO 80 sec, the sharp peak was at  $2\theta = 10.64^\circ$  with d-spacing at 0.83 nm, as shown in Figure 4(c). The sonicated GO 120 sec showed a sharp peak at  $2\theta = 11.4^\circ$  with d-spacing at

## Preparation of Graphene Oxide from Expanded Graphite

0.78 nm, as shown in Figure 4(d). These results indicate an increase of the interlayer distances of GO after sonication and a shift down in two theta ( $2\theta$ ).

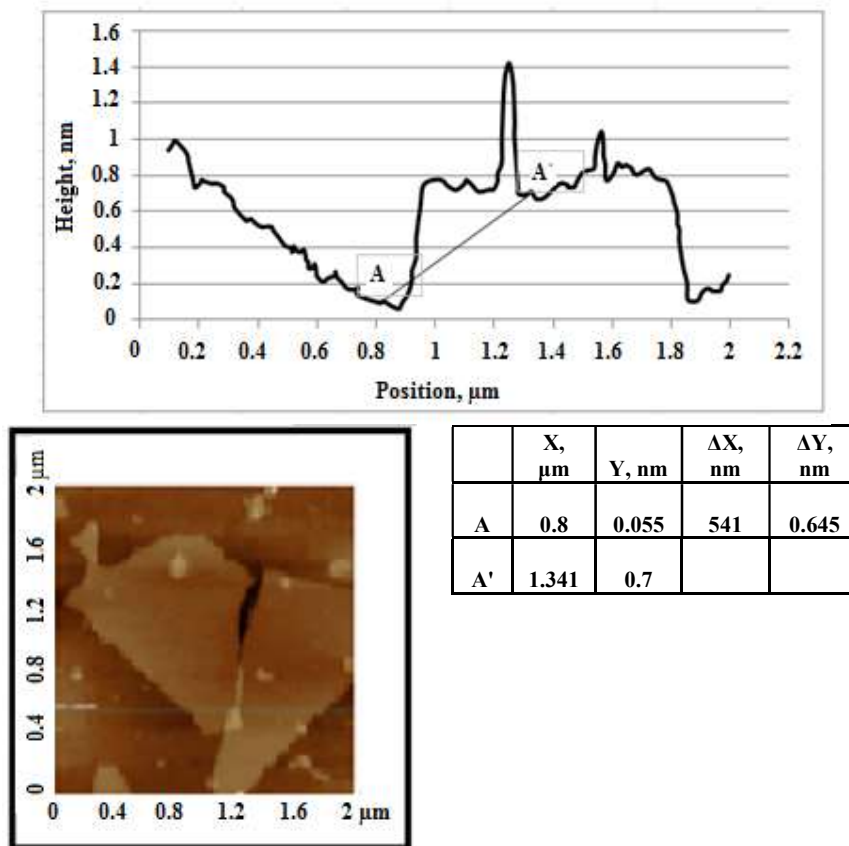


**Figure 4** XRD patterns of the prepared GO after sonication: (a) GO 20 sec; (b) GO 60 sec; (c) GO 80 sec; (d) GO 120 sec.

### 3.2 Atomic Force Microscopy (AFM)

Atomic force microscopy (AFM) was used to measure and characterize the graphene oxide. Figure 5 shows that the thickness and lateral size of GO 20 sec were 0.645 nm and 541 nm respectively. Song, *et al.* [23] reported that the GO layers had a thickness of  $2 \pm 3$  nm, which is thicker than a single layer of graphene. The characteristic thickness of single GO layers is  $\sim 0.8$  nm, which is  $\sim 0.44$  nm

thicker than that of graphene. Therefore, these results indicate the formation of a single layer of GO thicker than a single layer of graphene [23]. Therefore, these results indicate the formation of a single layer of GO.

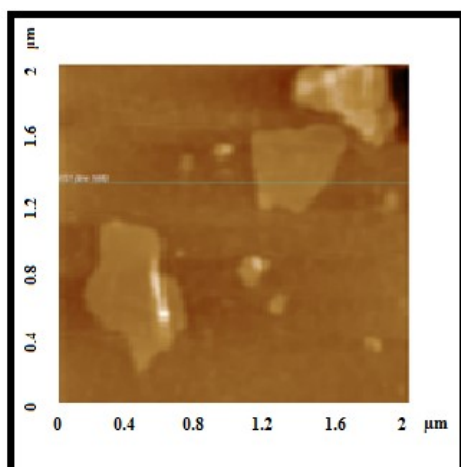
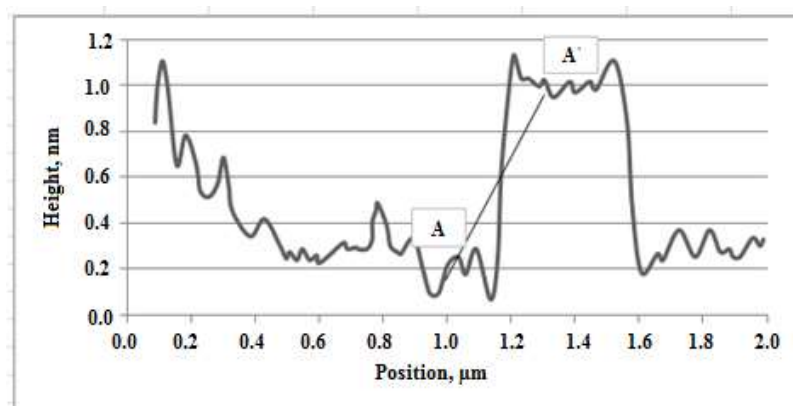


**Figure 5** AFM of prepared GO 20 sec.

The thickness, lateral size and morphology of the prepared GO 60 sec were 0.835 nm and 220 nm respectively as shown in Figure 6. The result indicates the formation of a single layer of GO.



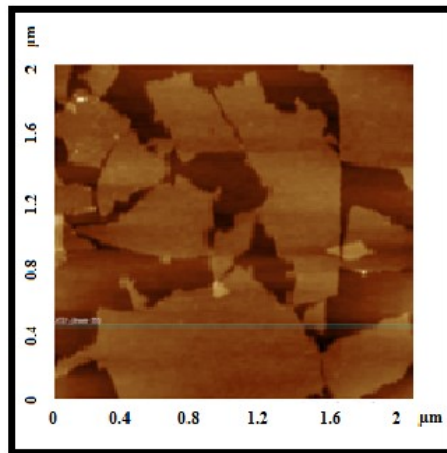
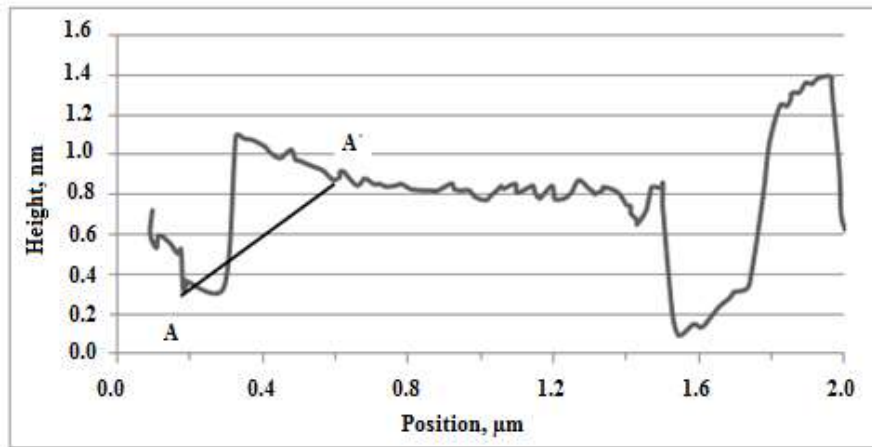
## Preparation of Graphene Oxide from Expanded Graphite



	X, $\mu\text{m}$	Y, nm	$\Delta\text{X}$ , nm	$\Delta\text{Y}$ , nm
A	1.027	0.145	220	0.835
A'	1.247	0.98		

**Figure 6** AFM of prepared GO 60 sec.

The thickness, lateral size and morphology of GO 80 sec were 0.519 nm and 439 nm, respectively, as shown in Figure 7. This indicates the formation of a single layer of graphene oxide. This thickness is the lowest compared with other works, where the thickness of single-layer GO was  $\sim 0.9\text{--}2.3$  nm [24]. The thickness of a single layer of GO in the one-step and two-step methods of the modified Hummer method was  $\sim 0.80$  nm [24,25].



	,μm	Y,nm	ΔX, nm	ΔY, nm
A	0.181	0.301	439	0.519
A'	0.62	0.82		

**Figure 7** AFM of prepared GO 80 sec.

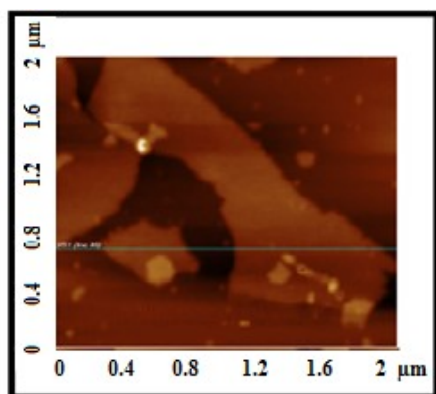
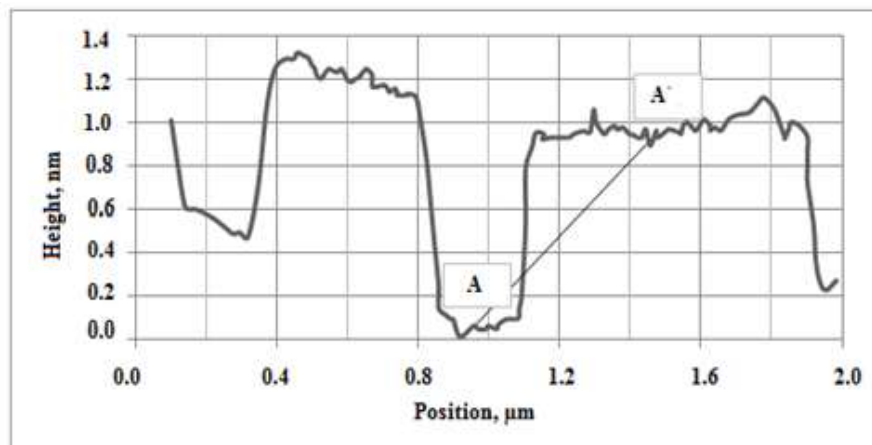
For GO 120 sec, the thickness, lateral size and morphology were 0.867 nm and 533 nm respectively, as shown in Figure 8. These results also indicate the formation of a single layer of graphene oxide.

### 3.3 Fourier Transmission Infrared (FTIR) of EG and GO

FTIR analysis was used to find the functional groups on the surfaces of the exfoliated graphite and the graphene oxide. Figure 9(a) shows the FTIR of the prepared EG using microwave heating with the specific band maximum in the

## Preparation of Graphene Oxide from Expanded Graphite

3300~3121  $\text{cm}^{-1}$  region due to the stretching and vibration of O-H hydroxyl [20,26]. The peak at 1330  $\text{cm}^{-1}$  is assigned to C-O (epoxy) and the peak at 1296  $\text{cm}^{-1}$  is assigned to the C-O group. There is an O=C=O peak at 2383  $\text{cm}^{-1}$ . The peaks at 1443  $\text{cm}^{-1}$  of C=O, 1076  $\text{cm}^{-1}$  and 1330  $\text{cm}^{-1}$  are related to C-O (epoxy) and C-OH (hydroxyl), respectively [21,27].



	X, $\mu\text{m}$	Y, nm	$\Delta\text{X}$ , nm	$\Delta\text{Y}$ , nm
A	0.917	0.013	533	0.867
A'	1.45	0.88		

**Figure 8** 8 AFM of prepared GO 120 sec.

The GO surface has oxygenated groups, such as epoxy (C-O), hydroxyl (-OH), carboxyl (C=O) groups, which were shown by the FTIR. Figure 9(b) shows the FTIR peaks for the GO prepared from oxidation of EG. The peak 3437  $\text{cm}^{-1}$  exhibits the characteristics of hydroxyl (O-H). The peak at 1728  $\text{cm}^{-1}$  indicates

carboxyl bands (C=O), while the in-plane C=C ( $sp^2$  carbon) skeletal stretching vibrations at  $1616\text{ cm}^{-1}$ ; and the peak at  $1400\text{ cm}^{-1}$  are for aromatic C=C. The peak at  $1365\text{ cm}^{-1}$  indicates the presence of epoxy (C-O) and the peak at  $1076\text{ cm}^{-1}$  is for C-OH [26-28]. It is clear from Figure 9(b) that the OH peak for GO at  $3487\text{ cm}^{-1}$  was enhanced, indicating that more OH functional groups were attached to the GO. Furthermore, there were more enhancements in the carboxyl bands (C=O) at  $1728\text{ cm}^{-1}$  for GO.

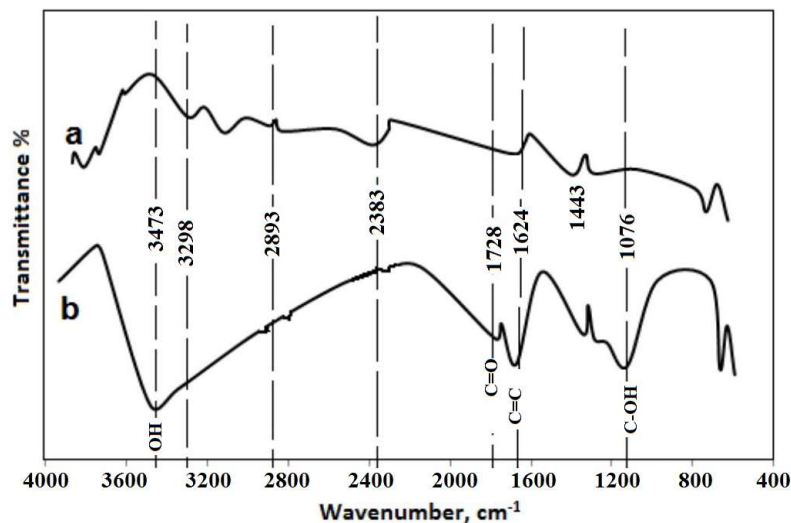


Figure 9 FTIR: (a) EG; (b) GO prepared from EG at 80 sec.

### 3.4 Effect of Microwave Time

The AFM, XRD and FTIR images indicated the successful preparation of GO from EG. The quality, thickness and distance of layers and morphology of the GO thin layer depend on the graphite oxidation process, the degree of oxidation and the conditions used to prepare the GO, as well as on the irradiation time in the microwave. The variation of d-spacing and diffraction angle peaks versus microwave heating times for the GO prepared from EG before sonication and after sonication are shown in Table 1. In this work, the optimum d-spacing was 0.78 nm (before sonication) for GO 80 sec, corresponding to  $2\theta = 11.38^\circ$ . After sonication, the optimum d-spacing for the prepared GO 80 sec was 0.83 nm, corresponding to  $2\theta = 10.64^\circ$ , as shown in Table 1. Furthermore, the AFM results showed that the smallest layer thickness was 0.519 nm for GO 80 sec. Thus, the AFM results or GO layer thickness (Figure 7) were in good agreement with an XRD d-spacing of 0.831

## Preparation of Graphene Oxide from Expanded Graphite

nm. Both results indicate that the optimum microwave heating time was 80 sec, which gave a maximum d-spacing of 0.831 nm and a minimum thickness of a single layer of graphene oxide of 0.519 nm. The percentage increment in d-spacing of the GO was calculated based on the d-spacing of the exfoliated graphite, which was 0.344 nm. The maximum percentage increase in d-spacing was 141% for the GO prepared from EG at 80 sec after sonication, as shown in Table 1.

**Table 1** Values of  $2\theta$  and d-spacing of the GO before and after sonication.

Time (sec)	Before GO sonication			After GO sonication				
	$2\theta^\circ$	d-spacing (nm)	d-spacing Increment %	$2\theta^\circ$	d-spacing (nm)	Lateral's size (nm)	GO thickness (nm)	d-spacing Increment %
20	11.18	0.79	130	10.9	0.81	541	0.645	135
60	11.9	0.74	115	11.4	0.76	220	0.835	121
80	11.38	0.78	127	10.64	0.83	439	0.519	141
120	11.74	0.75	118	11.4	0.78	533	0.867	127

## 4 Conclusions

In this research expanded graphite was prepared by mixing graphite with  $\text{HNO}_3$  and  $\text{KMnO}_4$  at a weight ratio of 1:2:1 using microwave irradiation for heating at four time periods (20, 60, 80 and 120 sec). After that, GO was successfully prepared from the EG. The AFM, XRD and FTIR analyses showed successful and complete conversion of EG into one single layer of GO. The thickness of the GO layer, the inter-planar distance (d-spacing) and morphology of the GO layers depends on the degree of graphite exfoliation (microwave irradiation time) as a precursor to prepare GO. Thus, in this study the optimum microwave irradiation time of EG was 80 sec, which provided a maximum d-spacing of 0.831 nm and a minimum thickness of 0.519 nm for the GO. Also, the XRD results indicated that the increase in d-spacing of the GO after sonication was 0.831 nm compared to 0.79 nm before sonication. The maximum percentage increase in d-spacing was 41% for the GO prepared from EG at 80 sec after sonication.

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## Preparation of Graphene Oxide from Expanded Graphite

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