

Synthesis and Characterisations of Reduced Graphene Oxide Prepared by Microwave Irradiation with Sonication

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ABSTRACT: *Recently, reducing graphene oxide (GO) through microwave irradiation has been extensively explored in order to scale up the mass production of graphene. We report the simple technique to reduce GO by means of microwave irradiation combined with a sonication technique. The microwave-reduced GO (MWrGO) is formed by exposing a microwave onto GO powder in order to reduce the oxygen functional group and then followed by exfoliating via a sonication method. The time exposure of the microwave irradiation was 20 min with the powers of 450 W and 800 W. The UV-visible (UV-vis) spectra showed the evolution of GO into MWrGO indicated by the red shift of the absorption peak from 230 nm to 267 nm and disappearance of the shouldering peak at 300 nm. The reduction of the oxygen functional group has been proved by Fourier transform infrared (FTIR) spectra. Furthermore, the scanning electron microscopy-energy dispersive X-ray spectroscopy (SEM-EDS) data demonstrated further confirmation of the reduction of GO and the formation of basal planes of sp^2 carbon clusters of the sample due to the treatment. The EDS spectra revealed that the MWrGO by 800-W-irradiation had much less oxygen functional groups and much more carbon content than GO. The proposed synthesis method is simple and readily controlled for a mass production of graphene from GO.*

Keywords: graphene oxide, reduced graphene oxide, microwave reduction, sonication

1. INTRODUCTION

Graphene has been known as a future material due to its excellent properties and widely potential applications.¹ In order to exploit its various possible applications, numerous studies have been conducted to improve the yield and reproducibility of graphene.² The common methods to obtain graphene sheets are micromechanical exfoliation from graphite, direct sonication of graphite, and electrochemical exfoliation, while the examples of the bottom-up approaches are chemical vapour deposition (CVD), confined self-assembly, arc discharge, epitaxial growth on SiC, and the reduction of graphene oxide (GO).³⁻¹⁰ Among these, the reduction of GO method is considered as the most promising method to largely synthesize graphene. Generally, GO is synthesized via the Hummers' method or other methods and then could be reduced by chemical methods using different reductants, e.g., hydrazine, dimethylhydrazine, hydroquinone and NaBH₄.¹¹⁻¹⁶ This reduction technique usually takes a long time and uses hazardous reductants. In fact, there are several studies that demonstrated the reduction of GO under specific conditions such as under alkaline condition, ultraviolet-assisted method and thermal methods.¹⁷⁻¹⁹ Recently, it has been reported that the microwave irradiation could reduce the oxygen functional groups of GO through its high temperature in a short time.^{20,21} However, the optimum method of how to completely remove oxygen functional groups is still a challenge.

A general framework in transforming graphite to reduced graphene oxide (rGO) is by conducting three necessary steps, i.e., oxidation, exfoliation and reduction.²² In this article, we demonstrate the use of microwave irradiation to reduce the oxygen functional group of GO powder. In order to generate well-dispersed graphene in water without the assistant of surfactant or stabilisers, we utilised the sonication method. The GO powder, which has been exposed by microwave irradiation, is then vibrationally exfoliated by means of a sonication method. The combination of microwave irradiation and sonication method could be a simple method to produce microwave-reduced GO (MWrGO) suspension, which can be readily modified for novel materials. Moreover, this method does not involve hazardous component such as hydrazine to generate rGO so that it can be considered as an environmentally friendly method.

2. EXPERIMENTAL

2.1 Synthesis of GO

GO was prepared using the modified Hummers' method as reported elsewhere.²³ In brief, the mixture of 1.0 g graphite, 0.5 g NaNO₃ with 23 ml of H₂SO₄

(98 wt%), was prepared in a beaker glass. Then, 3.0 g of KMnO_4 was added gradually to the mixture with 1 h slow stirring in an ice bath (20°C). The stirring process was continued in a heat bath at a temperature of 40°C for 20 h. Then 200 ml of deionized (DI) water was added drop wise into the mixture and stirred for 1 h under 90°C until the colour of the mixture turned brownish. H_2O_2 (5 ml, 30%) was used to react with the excess KMnO_4 designated by the golden colour of the mixture. The final mixture was then centrifuged and washed with DI water several times until the PH value of the mixture was nearly equal to 7. The mixture was dried in an oven to get GO powder. All the above materials were purchased from Alfa Kimia CV Yogyakarta.

2.2 Microwave Irradiation and Sonication Process

The microwave oven used in this article was a commercially available microwave oven under air atmosphere with a 2.45 GHz frequency while the sonication apparatus was a homemade sonication generator with a power of 130 W. The microwave irradiation was conducted on 50 mg GO powder placed into a small quartz tube (1 cm in diameter and 10 cm in length) under air atmosphere for 20 min with two different powers, namely 450 W and 800 W. Figure 1 illustrates the reduction process of GO into MWrGO. The GO powder is irradiated by microwave in order to reduce the oxygen functional groups of GO and turn GO into MWrGO. Afterwards, the GO and the irradiated GO samples are dispersed in deionized water and then sonicated for 1 h in order to get well-dispersed samples. The resulted samples are the GO suspension, the suspension of MWrGO by 450 W (MWrGO 450 W) and the suspension of MWrGO by 800 W (MWrGO 800 W). The characterisation of the samples is then conducted right away. The microwave oven was purchased from UFO Electronics Yogyakarta.

2.3 UV-visible (UV-vis) Characterisation

The absorbance of samples was measured using the UV-vis spectrophotometer Shimadzu UV-2450. The resulted spectra were analysed to obtain information of molecular transitions of graphite, GO, MWrGO 450 W and MWrGO 800 W. The UV-vis spectrophotometer is available in the Integrated Laboratory, Faculty of Mathematics and Natural Sciences, Universitas Negeri Yogyakarta, Indonesia.

2.4 Fourier Transform Infrared (FTIR) Characterisation

The FTIR spectra of the samples were measured using the Thermo Nicolet Avatar 360. The resulted spectra are analysed to qualitatively compare the oxygen functional groups on the samples. The FTIR unit is available in the Integrated Laboratory, Universitas Islam Indonesia, Yogyakarta.

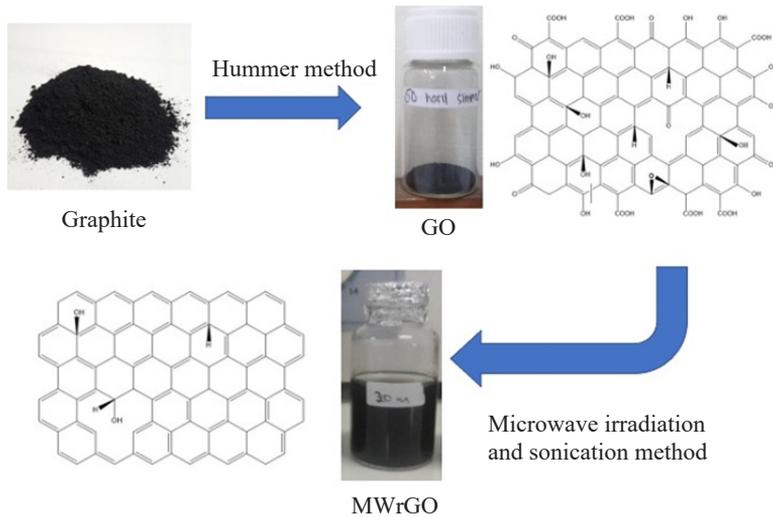


Figure 1: The synthesis route of MWrGO starting from the graphite powder being processed by the Hummers' method into GO then followed by a microwave irradiation and sonication method.

2.5 Scanning Electron Microscopy-Energy Dispersive X-ray Spectroscopy (SEM-EDS) Characterisation

SEM-EDS characterisation was performed to investigate the surface morphology and the atomic contents of the samples. SEM images and EDS spectra of GO, MWrGO 400 W and MWrGO 800 W were taken using JEOL 6510 LA with an operational voltage of 20 kV. The SEM-EDS unit is available in the Integrated Laboratory, Universitas Islam Indonesia, Yogyakarta.

3. RESULTS AND DISCUSSION

The GO powder in this work was prepared from graphite by using the modified Hummers' method. It is known that even though the interlayer distance of GO is extended, GO has a lot of oxygen functional groups which should be reduced in order to obtain rGO with comparable quality to pristine graphene.²⁴ Thus, we employed the microwave irradiation to reduce the oxygen functional groups to a certain extent. We irradiated the GO powder with two different wattages of the microwave to investigate how the microwave power influences the GO reduction. After the microwave treatment, we exfoliated the irradiated GO powder by means of the sonication method (see Figure 1). As a result, we obtained the suspension of MWrGO samples with lower oxygen content than that of GO. The

data of UV-vis spectroscopy (Figure 2), FTIR spectra (Figure 3), SEM images (Figure 4a–c), and EDS spectra (Figure 4d) demonstrate that our method successfully reduces the oxygen functional groups of GO. Furthermore, the basal planes of sp^2 carbon clusters of graphene were improved due to the microwave and sonication treatment.

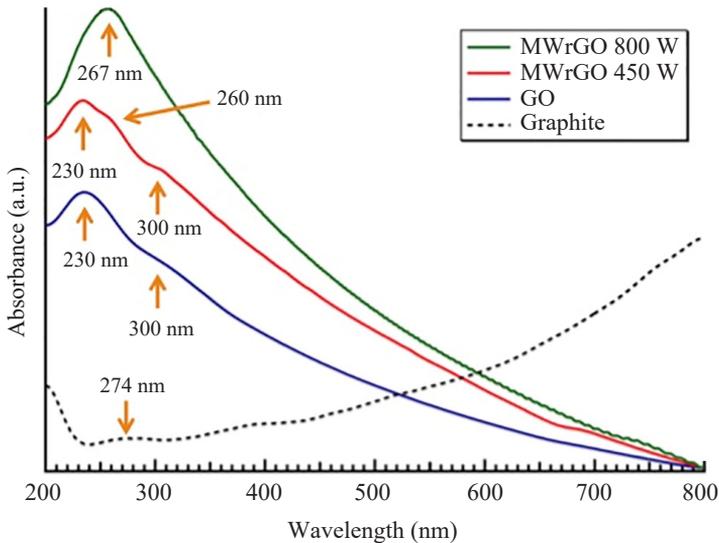


Figure 2: The UV-vis absorption spectra of graphite, GO, MWrGO 450 W and MWrGO 800 W. The spectra show the change in the absorption profile of graphite into GO and MWrGO.

The UV-vis spectroscopy is one of powerful methods to examine the molecular transition of carbon-based materials. The UV-vis results of graphite, GO, MWrGO 450 W and MWrGO 800 W can be observed in Figure 2. Original graphite shows a less pronounced peak at around 274 nm and strong absorption in the UV range. This is in accordance with Fan et al. who found the UV-vis peak of graphite to be 274 nm.²⁴ The UV-vis absorption of GO has two signature bands which are the peak at 230 nm and the shoulder at 300 nm. The peak located at 230 nm corresponds to the existence of $\pi \rightarrow \pi^*$ molecular transitions whereas the second shouldering peak at 270 nm indicates $n \rightarrow \pi^*$ molecular transitions.²⁵ Furthermore, these $n \rightarrow \pi^*$ molecular transitions indicate the presence of sp^3 clusters in GO, which should be reduced to obtain the desired graphene structure.

The UV-vis absorption peak of the GO samples at 230 nm shows a gradual red-shift to 270 nm with the increase of the microwave power. The MWrGO 450 W sample exhibits a peak at 230 nm with a small bump at 260 nm. This could be

attributed to the fact that the amount of the oxygen functional groups starts decreasing and the sp^2 carbon clusters gradually form. However, the shouldering peak at 300 nm observed in this sample indicates that the MWrGO 450 W sample still has a considerable number of sp^3 carbon clusters. Meanwhile, the absorption peak of the MWrGO 800 W shifts to 267 nm corresponding to the more reduction of oxygen functional groups and the more formation of sp^2 clusters in GO.²⁵ In addition, the shouldering peak at 300 nm in this sample is suppressed indicating that the sp^3 clusters on the sample are dramatically reduced. This behaviour might demonstrate that GO is gradually reduced and the sp^2 clusters of graphene are gradually formed due to microwave irradiation. This finding will be further confirmed by FTIR data, SEM images and EDS spectra of the samples.

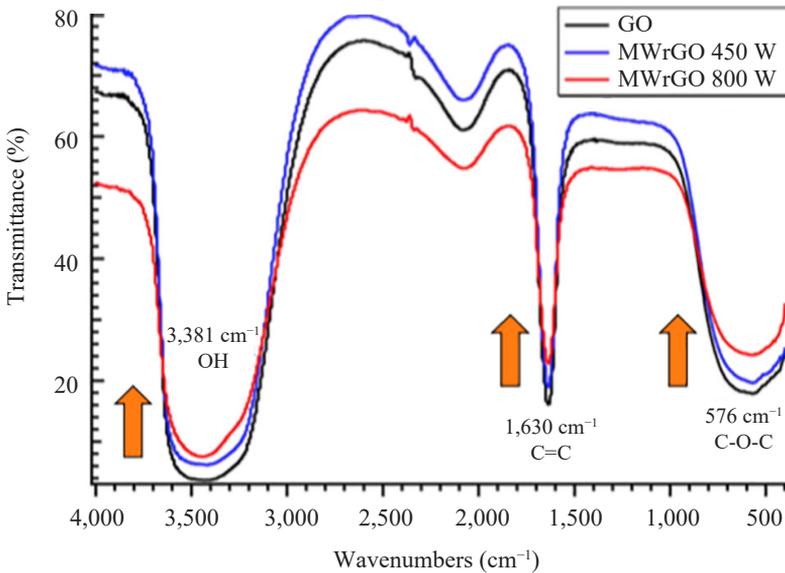


Figure 3: FTIR spectra of graphite, GO, MWrGO 450 W and MWrGO 800 W. The MWrGO spectra show the reduction of absorption peak designating the oxygen functional groups of GO. The orange arrows show the decrease of the functional groups.

The FTIR spectra of GO, MWrGO 400 W and MWrGO 800 W show functional groups contained in the samples (see Figure 3). Three strong absorption bands at 576 cm^{-1} , $1,630\text{ cm}^{-1}$ and $3,381\text{ cm}^{-1}$ are present in all samples. The strong absorption band at 576 cm^{-1} corresponds to C-O-C stretching vibration. The presence of an absorption band around $1,630\text{ cm}^{-1}$ is due to C=C stretching vibration. Another band at $3,381\text{ cm}^{-1}$ corresponds to stretching vibration of oxygen containing functional groups of hydroxyls (OH). These results are in accordance with the FTIR results obtained by Zhang et al. and indicate the

presence of oxygen functional groups.²⁶ It may be observed that the bands of all the samples have similar values. However, the intensities of these three bands are different for each sample. The FTIR spectrum of the GO has stronger absorption at three bands compared to the two spectra of the MWrGO. It means that GO has higher numbers of oxygen functional groups compared to two other samples. Furthermore, the amount of the oxygen functional groups of MWrGO 450 W is in between GO and MWrGO 800 W. In other words, the MWrGO 800 W samples have the fewest amounts of the oxygen functional groups among others. From this FTIR data, it can be observed that the reduction of oxygen functional groups occurs after the irradiation of the microwave. These reductions of oxygen functional groups demonstrate the successful preparation of rGO via microwave irradiation followed by the sonication method.

Interestingly, the absorption band at $1,630\text{ cm}^{-1}$ does not exhibit any differences among the three samples. This band indicates the C=C structure that becomes the signature of the honeycomb ring of graphene or sp^2 clusters in GO.²⁶ In this study, the resulted MWrGO still preserves the sp^2 clusters and yet it loses several amount of oxygen functional groups. This phenomenon is favourable since the reduction process of GO into rGO should consider the formation and recovery of sp^2 clusters so that it resembles the structure of graphene, which only consists of sp^2 clusters.²⁷ Furthermore, this indicates that the microwave irradiation only reduces the oxygen functional groups and still preserves the sp^2 clusters of GO or even recovers sp^3 clusters to become sp^2 clusters. During the irradiation treatment, the thermal energy from the microwave favours the further clustering of the sp^2 phase provoking connection among honeycomb rings and transforming it to the graphene structure.

The surface morphology of the sample was investigated by using SEM as shown in Figure 4a–c. It can be observed that the surface of the GO sample (Figure 4a) is smoother than those of MWrGO surfaces (Figure 4b–c) due to the good interaction between GO interlayers through functional oxygen groups.²⁸ This morphology qualitatively demonstrates that the oxygen functional groups of the GO sample are the largest among the other samples. However, the MWrGO samples turn out to be more transparent and more crumpled than the GO sample. The transparency of the MWrGO sheet indicates that more GO layers were exfoliated after microwave irradiation and sonication process so that they resulted in a thinner sheet. Meanwhile, the crumpled surfaces of both MWrGO 450 W dan MWrGO 800 W indicate a high surface-to-volume aspect ratio of those two samples.²⁹ This high aspect ratio can be attributed to the fact that oxygen functional groups are reduced to a certain extent and the basal planes of the sp^2 carbon clusters are improved after the microwave and sonication treatment.³⁰

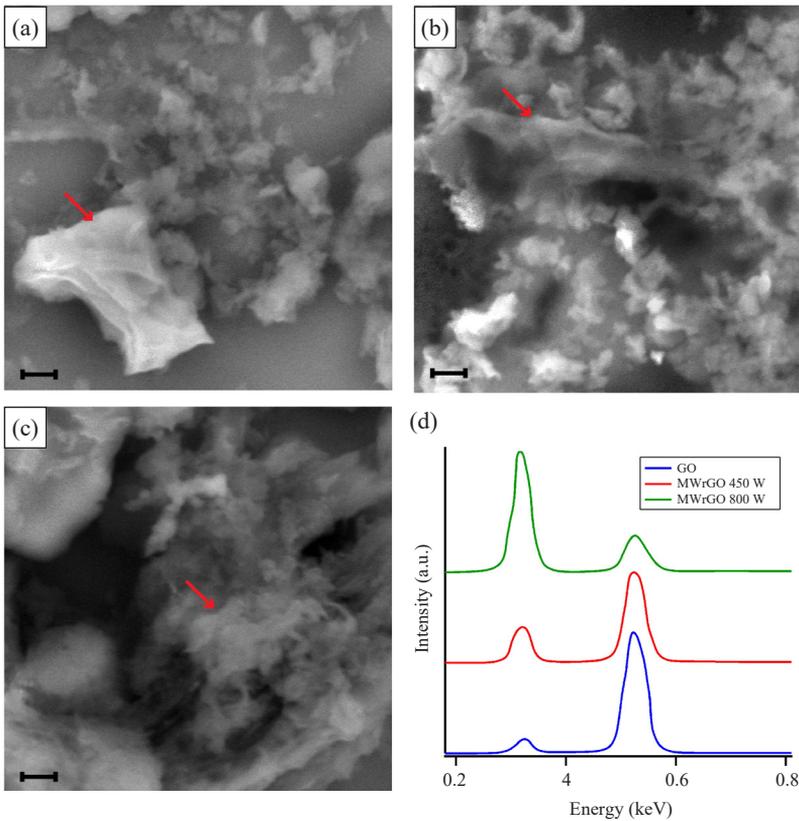


Figure 4: SEM images of the (a) GO samples, (b) MWrGO 450 W, (c) MWrGO 800 W deposited on glass substrates. The scale bar in the images is 1 μm, and (d) The EDS spectra are taken from specific spots indicated by red arrows.

To verify the reduction of oxygen functional groups of GO after the microwave and sonication treatment, we performed the EDS analysis on the samples. The EDS analysis could provide information of the relative change of the chemical state of the sample, such as the oxygen content (O Kα) on the graphene sheet.³¹ We investigated the chemical elements which were present on the specified spot of each sample which is indicated by a red arrow in Figure 4a–c. The EDS spectra of the samples show two characteristic X-ray energies at ~0.28 keV and ~0.53 keV which correspond to carbon content (C Kα) and O Kα, respectively. It can be observed from Figure 4d that the relative intensity of O Kα on the GO sample is the highest among the others while its C Kα shows the lowest intensity. Furthermore, from Table 1, it can be seen that the atomic percentage (at. %) of the O Kα decreasing from 60% for GO sample to 43% and 26% for MWrGO 450 W and MWrGO 800 W, respectively. Interestingly, the atomic percentage

of the C K α increased with the reduction of O K α from 19% for GO samples to 42% and 71% for MWrGO 450 W and MWrGO 800 W, respectively. This evolution of O K α and C K α on the samples confirms the reduction process of functional oxygen groups of GO after being irradiated by the microwave and sonicated. In addition, this EDS result proves that the basal planes of the sp² carbon of GO are improved after the treatment. The more power of the irradiation is used, the more functional oxygen groups of GO are reduced and the more sp² clusters are improved. This result also further indicates that the main mechanism in which the O K α is reduced via microwave irradiation is due to the direct heat agitation of the microwave. The heat produced by the microwave irradiation breaks the oxygen bonds, hence reducing the oxygen functional group upon the basal planes of the sp² carbon and provoking the recovery of sp² carbon clusters. However, further increasing the power of the microwave may also affect the sp² carbon bonds, which may produce an unwanted defect of the MWrGO produced.²²

Table 1. EDS elemental analysis of GO, MWrGO 450 W and MWrGO 800 W deposited on glass substrates. The Silicon (Si) contents appearing in all samples are due to the substrates.

Sample	Elements	Atom. Conc. (at. %)	Weight (wt%)
GO	C	18.97	13.02
	O	59.96	54.81
	Si	11.33	18.18
	Others	9.74	13.99
MWrGO 450 W	C	41.98	31.48
	O	42.94	42.90
	Si	9.18	16.10
	Others	5.90	9.52
MWrGO 800 W	C	71.08	63.18
	O	25.87	30.63
	Si	1.23	2.56
	Others	1.82	3.64

Based on the UV-vis, FTIR, SEM and EDS data, we observed that GO can be reduced into graphene (rGO) by using microwave irradiation followed by a sonication technique. In our method, we reduce the oxygen functional groups of GO by microwave irradiation, and then exfoliate the irradiated GO using sonication technique. This technique enables the reduction of GO without using hazardous materials and it is easily conducted. Thus, the combination of microwave irradiation and sonicated exfoliation may open opportunity to produce graphene in a large scale. This method is more eco-friendly than other oxygen reduction methods, e.g., oxidation and thermal expansion of graphite, UV-assisted

photocatalytic reduction, electrochemical polymerisation and oxygen reduction method under alkaline conditions utilised the Staudenmaier methods.^{17–19,32} However, a drawback of this method is in the use of the Hummers' method in synthesising the GO. Although we do not utilise hazardous chemicals in the reduction of GO, the Hummers' method still yields hazardous chemicals.³³ Hence, to fully provide an eco-friendly method in obtaining graphene through the reduction of GO, the latter should also be produced via alternative eco-friendly methods, such as using ultrasonication.³⁴

4. CONCLUSION

We report the possibility of reducing oxygen functional groups of GO via microwave irradiation followed by sonication. The UV-vis spectra show the red shift of 230 nm into 270 nm and the disappearance of the shouldering peak at 300 nm, which indicates the reduction of GO. This reduction process is in accordance with FTIR spectra indicating fewer oxygen functional groups of the MWrGO. The SEM-EDS data show further confirmation of the reduction of GO and the formation of basal planes of sp^2 carbon clusters of the sample. The resulted MWrGO 800 W exhibits less oxygen functional groups and more sp^2 carbon clusters than the MWrGO 450 W. This demonstrates that the more power of the irradiation is used, the more functional oxygen groups of GO are reduced and the more sp^2 clusters are improved. Furthermore, we have demonstrated a facile approach to synthesis rGO via microwave irradiation followed by sonicated exfoliation, which results in a substantial removal of oxygen functionalities of the GO. This method avoids the usage of hazardous chemical reductants that may introduce additional functional groups that are undesirable for large-scale production of graphene sheets. Future studies may include an investigation upon the surface of the MWrGO material obtained based on the adsorption or desorption isotherms and pore size distribution curves. It is also sensible to provide additional characterisations towards the MWrGO material produced, e.g., using X-ray diffraction (XRD) and high-resolution transmission electron microscopy (HR-TEM).

5. ACKNOWLEDGEMENTS

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