

One Step Process of Producing Reduced Graphene Oxide from Silantek Sub-Bituminous Coal Using Microwave Irradiation Heating

M.N. Iqbal^{1,2}, M.I. Azlan^{1,3}, I. Khudzir^{2,3}

¹Faculty of Applied Sciences, University Teknologi MARA, Arau, Perlis, 02600, Malaysia

²Faculty of Applied Sciences, University Teknologi MARA, Shah Alam, Selangor, 40450, Malaysia

³Coal and Biomass Energy Research Group, Universiti Teknologi MARA, Shah Alam, Selangor, 40450, Malaysia

*Corresponding author E-mail: mohdiqbal@ yahoo.com

Abstract

Reduced graphene oxide (rGO) was prepared from purified Silantek sub-bituminous coal by using microwave irradiation heating with the presence of FeCl_3 and ZnCl_2 as graphitizing and activating agents, respectively. The preparation was carried out at microwave power level ranging from 600 to 1000W. The results showed that rGO produced from sub-bituminous coal has same characteristics as produced from graphite. X-ray Diffraction (XRD) spectra showed that the sharp peak presence at $2\theta = 26^\circ$ had decreased and broaden after microwave irradiation heating due to the removal of various oxide groups with interlayer distance reduced to 0.337nm similar to that of graphite. Further, Raman spectra showed a high degree graphitization of rGO produced at 900 and 1000W of microwave irradiation power level. Moreover, the degree of crystallinity of rGO produced from coal is 1.31 (G900) and 1.15 (G1000), which is similar to rGO produced from graphite. Thus, this suggest that purified Silantek sub-bituminous coal upon heating using microwave irradiation method is potentially to be used as a carbon precursor to produce rGO.

Keywords: crystal structure; graphene; graphitization; microwave irradiation system; reduced graphene oxide; sub-bituminous coal

1. Introduction

Graphene is one of carbon allotrope and the most popular carbon material because it's potential in various applications for human life such as medical, chemical and industrial processes. Graphene is a layer of graphite which can be conceived as a two-dimensional of single layer carbon bonded atom in a basic structural element of hexagonal lattice structure [1-4]. The structure of graphene consists of sp^2 carbon bonded atom structure that attached each other by strong covalent bonding in form of one-atom-thick planar sheet with a high crystallinity and electric conductivity properties [5-8]. Ever since it first isolated by Geim and Novoselov in 2004 [9-11], graphene has been making a remarkable impact in many areas of science and technology due to its remarkable physicochemical properties such as high specific surface area of $2630\text{m}^2\text{g}^{-1}$ (theoretically), electron transport capabilities, superlative mechanical strength (Young's modulus, 1100 GPa), unparalleled thermal conductivity (5000 W/m/K) and excellent electrical conductivities [3]. Thus, these unique physicochemical properties of graphene promising great potential that can provide new approaches and critical improvements in many biochemical and nano-technological fields such as electronic devices and biosensors [12-16].

Practically, rGO is one of graphene related material which usually refers to graphene that is produced through oxidation and reduction process of graphite [17-19]. At present, there are several techniques have been developed in order to produce graphene such as micromechanical exfoliation, chemical vapor deposition, transfer printing, epitaxial growth and the reduction of graphene oxide

(GO) [7,8]. However, the reduction of GO is one of a promising alternative path toward high-yield production of graphene especially for commercialization due to low production cost and easily up-scalable process. GO is a carbon material that consists of a single layer of carbon bonded atom of graphite structure that has been attached by oxygen functional groups on both of its basal planes and edges, which is good to be as a candidate for carbon precursor to produce rGO [20-22]. Technically, GO consists of layers of graphite oxide because it was usually produced from graphite that has been oxidized by using oxidation method such as Hummers method, with the presence of suitable organic solvents or strong acids [5,6].

Practically, it has been reported and identified by previous study that the oxygen functional groups in GO structure consist of hydroxyl and epoxy groups on the basal plane. Further GO also consist of smaller amounts of carboxyl, carbonyl, phenol, lactone and quinone at the edges of carbon sheet [5,6]. All of the properties of GO such as mechanical, electrical and electrochemical properties are strongly affected by the presence of oxygen functional groups within its structure which possibly limits the direct application of GO in electrically active materials and devices [23]. Thus, the properties of GO can be improved through reduction process to produce rGO, either through chemical, thermal or H_2 plasma reduction process [5]. Further, the properties of rGO such as conductivities still lack behind the pristine graphene due to the residual functional group that remaining after the reduction process that cannot be totally removed [21]. However, nowadays this related graphene material (known as rGO) has emerged as a novel material in nano-carbon research that can be readily sold in tons due to scalable for large production at low cost [22].

Usually, graphite is the most chosen material as carbon precursors to produce GO because it is much easier to convert into GO by oxidation process, and finally turn into rGO product by reduction process. [24] produced GO from purified natural graphite by using oxidation process of Hummers and Offeman methods as a precursor to produce rGO. They found that the GO produced has 2 θ peak at 10°. Further, they also reported that the Raman spectra of GO produced has the G and D band peaks at around 1355cm⁻¹ and 1598cm⁻¹ which indicate that the material has a graphite structure. Then, the GO product was reduced into rGO and it was found that the peak of 2 θ at 10° has been reduced and disappeared. Further, [25] reported in their study on the production of GO as a rGO precursor from pristine graphite that the degree of crystallinity of the material will low if the material has high oxygen functioning groups in their structure [25]. It seems that graphite is very popularly to produce rGO which is as a carbon precursor to produce graphene. This made the production of graphene to be very expensive due to the limited resource of graphite. Moreover, the process to produce rGO from graphite provided two-step processes of oxidation and reduction processes.

Recently, some researchers found that GO can also be produced from any carbonaceous material (raw material that contain carbon element) such as coal [26,27] and biomass waste [28,29]. However, the carbonaceous material has to be graphitized first before it can be converted into GO in order to produce a crystalline structure like graphite [26]. Coal is abundantly available and inexpensive resource rich in carbon content that has a potential to be used as carbon precursor to produce GO and graphene. Quan et al. [27] has reported to produce rGO from Taixi coal. They found that the coal sample has to be graphitized first at a very high temperature of 2400°C before it can be converted into GO by using Hummers method. They reported that the GO produced has the same characteristics with the GO produced from graphite. Then, the GO produced was reduced into rGO via thermal reduction process. However, the production process from coal to rGO involved at least three step processes and required a very high temperature and long production time. In another study, Yating et al. [30] also produced GO as a precursor to produce rGO from Taixi anthracite coal. The rGO produced from GO derived from coal has similar properties as GO derived from graphite. After that, the GO material was reduced into rGO through ultrasonication process. However, they also reported the same result and faced same problem like Quan et al. [27] which the coal sample must go through three steps processes of graphitization, oxidation and reduction in order to produce rGO.

In this study Silantek sub-bituminous coal was used as a carbon precursor to produce rGO using microwave irradiation heating system in the presence of FeCl₃ as graphitizing and activating agents. All of three processes of graphitization, oxidation and reduction were conducted in only one single step process by using microwave irradiation system as a heating media. Thus, the production time to produce rGO was significantly reduced. The coal sample was treated in order to remove impurities by leaching off the inorganic matter prior to the reaction to produce rGO. As far as the literature is concern, there is no other research been reported that used sub-bituminous coal alongside with microwave irradiation heating method to produce the rGO in a single step process.

2. Experimental

2.1. Coal pre-treatment

The Silantek sub-bituminous coal sample was collected from Sarawak, Malaysia. The sample was ground into a fine powder and sieved into a uniform size of less than 2.0 mm in order to make sure the sample was completely mixed with graphitizing and activating agent. Then, the sample was heated overnight in an oven at 100°C to remove moisture. The pre-treatment process on the dried coal sample was carried out to remove impurities and to generate

high carbon content by using different chemical treatments such as of hydrogen peroxide (H₂O₂) and sulphuric acid (H₂SO₄), hydrochloric acid (HCl) and hydrofluoric acid (HF) [31,32]. At first, 10g of coal sample was poured into a solution of HCl and HF. Then, the mixture was stirred for 5 hours at a temperature of 55°C. Then, the sample was washed by distilled water in order to remove excess HCl and HF. After that, the sample was poured into a solution of H₂O₂ with the presence of two or three drops of H₂SO₄ and stirred for 2 hours at temperature of 55°C. After the treatment process the sample was washed by using distilled water and dried overnight in an oven at temperature of 100°C [31,32]. The treated sample was analyzed by using XRF. The proximate and ultimate analyses of the untreated and pre-treated coal samples were carried out using ASTM D2974 method and are shown in Table 1.

2.2. Sample preparation

Silantek sub-bituminous coal, ferric chloride (FeCl₃) and zinc chloride (ZnCl₂) were used as carbon precursor, graphitizing agent and activating agent, respectively for the preparation of rGO from coal. Typically, 3g of pre-treated coal sample was mixed with 9g of ZnCl₂ in a 50ml of 3M FeCl₃ solution [29]. Then, the mixture was stirred at temperature of 80°C for 2 hours to ensure uniform mixing before it was heated overnight at 100°C in conventional ventilated oven.

2.3. Preparation of reduced graphene oxide

In a quartz reactor, the mixture of Silantek coal, FeCl₃ and ZnCl₂ was placed inside the microwave heating system. Microwave heating process was carried out in a 2.45 GHz laboratory microwave at power levels of 600, 700, 800, 900 and 1000W at 10min of irradiation time and denoted as G600, G700, G800, G900 and G1000, respectively. After the microwave heating process completed, the sample was leached using 50% of HCl for 2 hours in order to remove excess FeCl₃ and ZnCl₂ within the sample followed by washing with distilled water to pH 6.5-7 and dried overnight in an oven at 100°C [29].

2.4. Characterizations

The elemental composition of materials present in untreated and pre-treated coal samples were analyzed using X-Ray Fluorescence (XRF). Surface functional groups of G600, G700, G800, G900 and G1000 samples produced were measured using a Bruker Equinox 55 of Fourier transform infrared (FTIR) spectrometer. The crystal structure for the sample of rGO produced was carried out by XRD techniques using Rigaku D/Max-2500 diffractometer with Cu-K α radiation. The powdered of G600, G700, G800, G900 and G1000 samples were mounted directly onto a zero background holder and were scanned at a rate of 0.02°/2 s in the 2 θ range of 5–80° [29]. Raman spectroscopy was used in order to determine the chemical bonds and symmetry of molecules of the product through vibration information, as well as to investigate the defect and carbon disordered within the rGO structure. Raman spectra for the sample were measured by using Raman Spectrometer of Renishaw. The process was done by applying laser excitation at 633nm with a high-resolution gradient from 150 to 2500cm⁻¹ [29]. After that, the resulting data was analyzed by using the Thermo Scientific Software.

3. Result and discussions

3.1. Treated and untreated coal analysis

From Table 1 it shows that Silantek coal has a high composition of carbon and found suitable as a source of carbon to produce rGO. The percentages of carbon for both proximate and ultimate analyses significantly increased from 67.30 to 73.60% and 73.35 to

78.84%, respectively after the ($\text{H}_2\text{O}_2 + \text{H}_2\text{SO}_4$) pre-treatment process. The increased in the percentages of carbon seems to correlate with the ash constituent analysis on the untreated and treated samples using semi-quantitative XRF as shown in Table 2.

Table 1: Proximate and Ultimate Analyses of Treated and Untreated Coal

Analysis	Coal Samples		
	Untreated	Treated (HCl + HF)	Treated ($\text{H}_2\text{O}_2 + \text{H}_2\text{SO}_4$)
Proximate			
Fixed Carbon	67.30	72.18	73.60
Volatile Matter	22.21	25.33	26.27
Ash	10.49	2.49	0.13
Ultimate			
Carbon	73.35	74.74	78.84
Hydrogen	4.45	4.33	4.25
Nitrogen	1.46	1.21	1.16
Sulfur	0.16	0.00	0.01
Oxygen (calculated by difference)	20.58	19.72	15.74

Table 2: XRF Analysis for the Sample of Treated and Untreated Coal

Compound	Coal Samples		
	Untreated	Treated (HCl + HF)	Treated ($\text{H}_2\text{O}_2 + \text{H}_2\text{SO}_4$)
Fe_2O_3	50.2340	22.5380	0.1810
SO_3	12.2290	15.0630	0.3420
SiO_2	12.1570	7.3720	0.0127
CaO	8.9080	3.8650	0.0578
Al_2O_3	4.4310	1.4670	0.0171
TiO_2	4.0180	7.2720	0.2210
K_2O	2.9490	2.3730	Traces
Ag_2O	2.8610	3.3470	0.0412
SrO	0.4770	0.2070	0.0053
CuO	0.4050	0.1620	0.0025
NO	0.2590	0.2560	0.0052
Cl	0.1970	35.250	0.494
ZnO	0.1560	0.0833	0.0005
ZrO_2	0.1540	0.1880	0.0026
Others	0.5650	0.2543	0.0057
Ag_2O	2.8610	3.3470	0.0412

In Table 2, XRF result shows that Fe_2O_3 , SO_3 and SiO_2 were the major mineral constituents in the coal and were greatly reduced after the pre-treatment process especially by using ($\text{H}_2\text{O}_2 + \text{H}_2\text{SO}_4$). Further, it shows that the presence of sulphuric acid in hydrogen peroxide solution enhances the removal of mineral constituents from the coal samples. Apparently, the percentages of Fe_2O_3 and SO_3 were significantly reduced from 50% to 0.18% and 12% to 0.34%, respectively. Moreover, the pre-treatment process by using peroxide did not lead to addition of significant amount of oxygen to the residual coal samples that would cause concern for dilution of the sulfur levels. These results seem to agree with the previous study reported by Samit et al. [31].

3.2. Reduced Graphene Oxide analysis

All FTIR spectra of untreated and treated Silantek coal and after microwave irradiation heating process of G1000 are shown in Figure 1. FTIR analyses were conducted by depositing rGO samples onto germanium and measuring the transmission spectrum. From Figure 1(a) for the untreated sample it shows the presence of high amount of oxygen containing group which are mainly due to C=O, C-O and O-H at peak of 1580 , 1050 and 2950cm^{-1} , respectively [25]. However, when the sample was treated with $\text{H}_2\text{O}_2 + \text{H}_2\text{SO}_4$ (see Figure 1(b)), the peak at 1050cm^{-1} reduced its intensity with the appearance of broad absorption band at 1250cm^{-1} which is due to the bending of =C-H that might be caused by the removal of some oxygen after pre-treatment process. Further, there appears to be absorption peak at 1580cm^{-1} that was due to the skeletal vibrations of un-oxidized graphitic domains after microwave irradiation heating process. Apparently, the absorption

peak at 2950cm^{-1} had disappeared with the emergence of new broad absorption band at 3200cm^{-1} , which indicates the presence of O-H groups that is specific to the GO and rGO that is still remains in the material [27]. GO is strongly hydrophilic and easily exfoliated in water to form stable colloidal dispersions because of the large amounts of hydroxyl (-OH), carboxyl (-C(=O)OH), carbonyl (-C=O) and epoxide functional groups attached to the basal or edge plane [33]. It was found that the oxygen functional group in that material cannot be removed totally when the sample was converted into rGO.

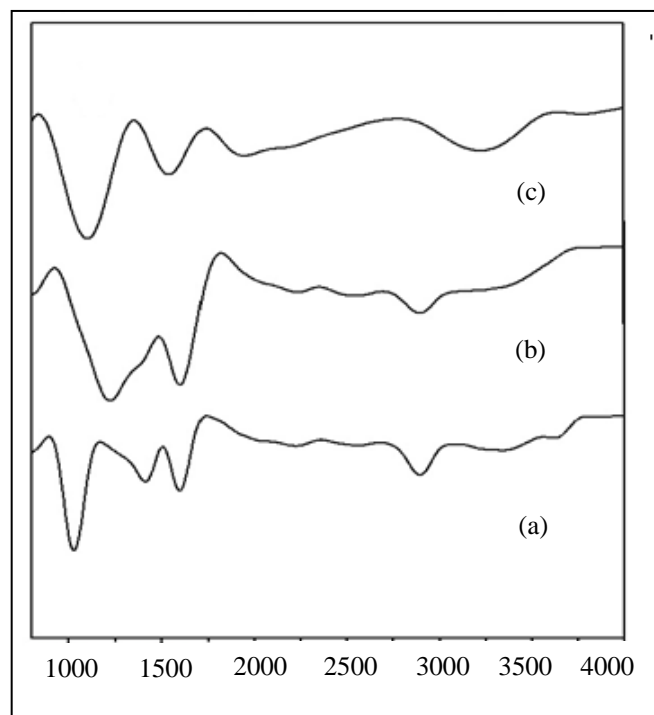


Fig. 1: FTIR spectra for (a) untreated coal, (b) ($\text{H}_2\text{O}_2 + \text{H}_2\text{SO}_4$) treated coal and (c) microwave irradiation coal (G1000).

XRD spectra were used to compare and obtain information about the crystallinity of untreated Silantek coal, G600, G700, G800, G900 and G1000. Figure 2 shows that untreated Silantek coal exhibited a broad peak at $2\theta = 24^\circ$ and a sharp peak at $2\theta = 26^\circ$. The amorphous broad peak at $2\theta = 24^\circ$ is due to the amorphous plane of Hematite (Fe_2O_3) which is the major ash constituents within the coal sample. The strong sharp peak that presence at $2\theta = 26^\circ$ is assigned to (002) plane similar to that of graphite plane, implying the high crystallinity of the coal sample [27].

The strong sharp peak presence at $2\theta = 26^\circ$ becomes weaker and broaden after the coal was treated with graphitizing agent and heating process due to the intercalation of functional groups into carbon of graphite-like structure, indicating the formation of rGO [25]. In XRD spectra of G600, G700, G800, G900 and G1000, it was found that the plane of (002) at $2\theta = 24^\circ$ becomes undetectable due to the removing of Hematite and other oxygen functional group, seems to be agreed with the result of FTIR. Further, a sharp peak at $2\theta = 26^\circ$ become broaden due to elongation of carbon chain within the material along with the increasing of microwave power level. This observation seems to agree with results of rGO produced from graphite and other coals [24, 25, 27, 30]. Table 3 shows the basal plane distance for untreated coal sample and after microwave heating process. Basal plane distance calculated from (002) plane of the sample can indicate the ordering in graphite layers within its structure. The basal plane distance calculated from XRD diffractogram of Silantek coal is 0.886nm . The basal plane distance of the sample decrease from 0.485nm of G600 to 0.337nm of G1000 after the microwave irradiation process due to the removal of oxide functional group within the material. Further,

the basal plane distance for G900 and G1000 seems to be identical as rGO produced from graphite material [24].

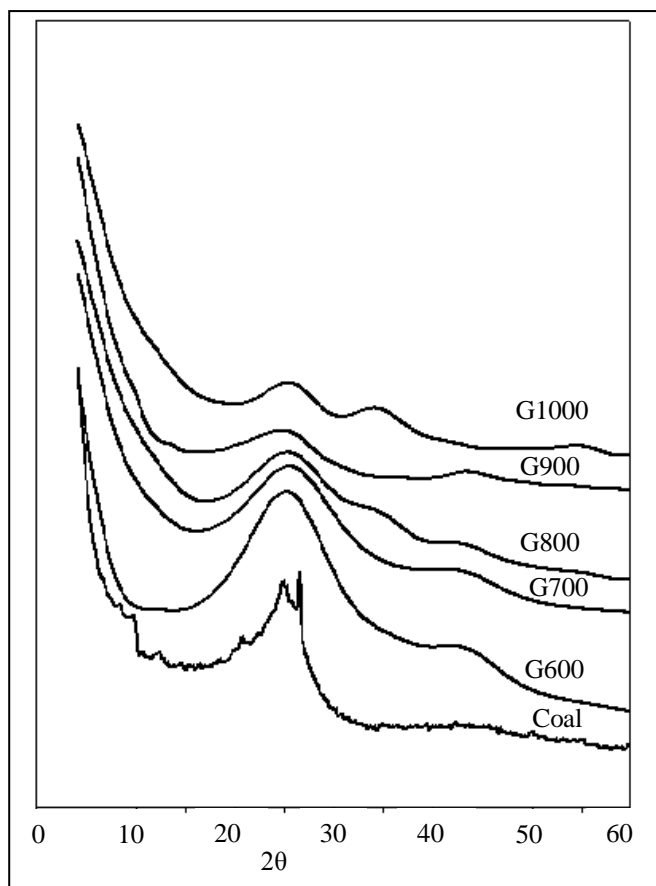


Fig. 1: XRD spectra for raw Silantek coal and rGO produced from Silantek coal (G600, G700, G800, G900 and G1000).

Table 3: Basal Plane Distance for the Sample of Coal and rGO (G600, G700, G800, G900 and G1000)

Samples	Basal Plane Distance (nm)
Coal	0.886
G600	0.485
G700	0.367
G800	0.341
G900	0.337
G1000	0.337

Raman spectroscopy is used in order to distinguish the ordered and disordered of carbon structures of the sample. In general, for carbonaceous material the bands D and G are the most studied, while the 2D bands is important to differentiate between the GO material and graphene material. The D band mode, in the range of 1200-1400 cm^{-1} is associated with imperfections or defects in the sp^2 carbon structure of the material such as disordering structure, reduction in crystal size, distortion in the network, and presence of heteroatoms). In contrast, the G band mode in the range of 1500-1600 cm^{-1} , indicates the presence of sp^2 carbon networks [10]. Figure 3 shows the Raman spectra for G600, G700, G800, G900 and G1000 that were treated at different microwave irradiation power levels at 30 min of irradiation time. Apparently, the Raman spectra shows that all the material produced were rGO due to the appearance of the D and G bands at the wavelength around 1300 and 1600 cm^{-1} , respectively. All of rGO produced was not formed as highly graphitizing material due to the low I_D/I_G ratio as shown in Table 4 because they have lower G band than D band with exception to the rGO produced at 900 and 1000W power level.

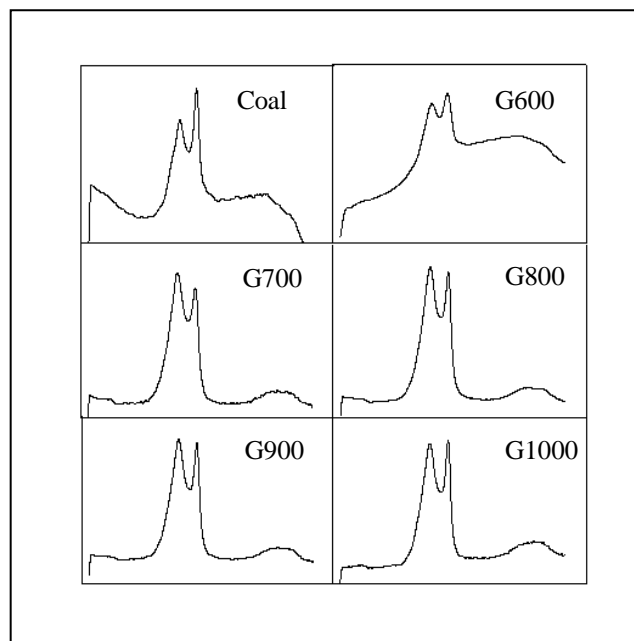


Fig. 1: XRD spectra for raw Silantek coal and rGO produced from Silantek coal (G600, G700, G800, G900 and G1000).

Table 4: I_D/I_G Ratio for the Samples Coal and rGO (G600, G700, G800, G900 and G1000)

Samples	I_D/I_G Ratio
Coal	0.88
G600	1.89
G700	2.58
G800	2.73
G900	1.31
G1000	1.15

The increasing in I_D/I_G value was due to the increasing of D band due to increasing of defects such as vacancies and disorder degree after heating process. The lower value of G band than D band was due to the residual remaining of oxygen functioning group into the carbon structure that was not completely removed, and these results are consistent with the results observed from FTIR analysis [25]. The higher value G band than the D band was observed for G900 and G1000, and this condition might have resulted from the removal of more stable oxygen functionalities. Hence, the 2D band of G1000 produced shows a higher broad peak than the others with a nearly symmetrical shape centred at 2800 cm^{-1} indicating that the rGO produced has higher degree of graphitization. Thus, all of the materials produced can be classified as rGO by the fact that the G band signatures showed a graphite structure and the D band indicates defects on the edges or surfaces of the rGO produced [25].

4. Conclusion

Reduced graphene oxide (rGO) was successfully produced from Silantek sub-bituminous coal by using microwave irradiation heating system in the presence of graphitizing agent and activating agent. The rGO that was obtained at 900 and 1000W of microwave power level has a high degree of graphitization in comparison to the others due the higher removal of oxygen functioning group and the decreasing of disordered carbon within the material. Further, rGO produced at 900 and 1000W microwave irradiation also has same crystallite size with rGO produced from other coal and graphite. However, it was found that rGO produced at 900W was the best product in term of production cost due to it lower power used compared to 1000W. This finding shows that rGO can be produced from coal with a single step process by using microwave irradiation heating system and the resulting material has the same characteristic as rGO produced from graphite.

Acknowledgement

We thank all parties that helped us for finishing this study on production of rGO from coal, especially all technicians for analyses process such as XRD and Raman spectrometer. We also thank to Faculty of Applied Science, UiTM Shah Alam which supported us to finish this study.

References

- [1] Velram BM, Kin-tak L, David H & Debes B (2018). Graphene-based materials and their composites: A review on production, applications and product limitations. *Composites Part B* 142, 200–220.
- [2] Tiannan Z, Feng CKai L, Hua D, Qin Z, Jiwen F & Qiang F (2011). A simple and efficient method to prepare graphene by reduction of graphite oxide with sodium hydrosulfite. *Nanotechnology* 22 (045704), 6.
- [3] Josphat P, Patrick G & Thad CM (2017). General overview of graphene: Production, properties and application in polymer composites. *Materials Science and Engineering B* 215, 9–28.
- [4] Khenfouch M, Buttner U, Mimouna B & Malik M (2014). Synthesis and Characterization of Mass Produced High Quality Few Layered Graphene Sheets via a Chemical Method. *Science Resources* 3, 7-14.
- [5] Ren S, Rong P & Yu Q (2018). Preparations, properties and applications of graphene in functional devices: A concise review. *Ceramics International* 44, 11940–11955.
- [6] Virendra S, Daeha J, Lei Z, Soumen D, Saiful IK & Sudipta S (2011). Graphene based materials: Past, present and future. *Progress in Materials Science* 56, 1178–1271.
- [7] Novoselov KS, Jiang D, Schedin F, Booth TJ, Khotkevich VV, Morozov SV & Geim AK (2005). A roadmap for graphene. *PNAS* 102 (30), 10451–10453.
- [8] Bhagya LD, Jamshid MN, Dermot B & Sumsun N (2017). Graphene and derivatives – Synthesis techniques, properties and their energy applications. *Energy* 140, 766–778.
- [9] Randviir EP, Brownson DAC & Banks CE (2014). A decade of graphene research: production, applications and outlook. *Materials Today* 17 (9), 426–432.
- [10] C. Shamik & B. Rajasekhar (2014). Recent advances in the use of graphene-family nano-adsorbents for removal of toxic pollutants from wastewater. *Advance in Colloid and Interface Science* 204, 35–56.
- [11] Zhong Y, Zhen Z & Zhu H (2017). Graphene: Fundamental research and potential applications. *FlatChem* 4, 20–32.
- [12] Bin W, Yan-hong C & Lin-jie Z (2011). High yield production of graphene and its improved property in detecting heavy metal ions. *New Carbon Materials* 26, 31-35.
- [13] Zhang C, Lv W, Zhang W, Zheng W, Wu M, Wei W, Tao Y, Li Z & Yang Q (2014). Reduction of graphene oxide by hydrogen sulfide: a promising strategy for pollutant control and as an electrode for Li-S Batteries. *Journal of Advance Energy Materials* 4, 175–182.
- [14] Lorenzo G, Vishnu TC, Di W, Chris B, Piers A & Tapani R (2012). Graphene for energy harvesting/storage devices and printed electronics. *Particuology* 10, 1–8.
- [15] Soldano C, Mahmood A & Dujardin E (2010). Production, properties and potential of graphene. *Carbon* 48, 2127-2150.
- [16] Li W, Bi W, Shimin W (2015). Well-dispersed PEDOT: PSS/graphene nanocomposites synthesized by in situ polymerization as counter electrodes for dye-sensitized solar cells. *Journal of Materials Science* 50, 2148–2157.
- [17] Zhao B, Liu P, Jiang Y, Pan D, Tao H, Song J, Fang T & Xu W (2012). Supercapacitor performances of thermally reduced graphene oxide. *Journal of Power Sources* 198, 423-427.
- [18] Zhu X, Zhu Y, Murali S, Stoller MD & Ruoff RS (2011). Nanostructured reduced graphene oxide/Fe₂O₃ composite as a high-performance anode material for lithium ion batteries. *American Chemical Society* 5, 3333-3338.
- [19] Cheng IF, Xie Y, Gonzales RA, Brejna PR, Sundrarajan JP, Kengne BAF, Aston DE, Macllroy DN, Foutch JD & Griffiths PR (2011). Synthesis of graphene paper from pyrolyzed asphalt. *Carbon* 49, 2852-2861.
- [20] Ling S & Bunshi F (2013). Mass production of graphene oxide from expanded graphite. *Material Letters* 109, 207–210.
- [21] Samuel J, Rowley N, Edward PR & Ahmed SAD, Craig EB (2018). An overview of recent applications of reduced graphene oxide as a basis of electroanalytical sensing platforms. *Applied Materials Today* 10, 218–226.
- [22] Yun-zhen C, Gao-yi H, Yao-ming X, Hai-han Z & Jian-hua D (2017). A comparative study of graphene oxide reduction in vapor and liquid phases. *New Carbon Materials* 32 (1), 21-26.
- [23] Nanting L, Shaochun T, Yumin D & Xiangkang M (2014). The synthesis of graphene oxide nanostructures for supercapacitors: a simple route. *Journal of Material Science* 49, 2802–2809.
- [24] Chao X, Ru-sheng Y & Xin W (2014). Selective reduction of graphene oxide. *New Carbon Material* 29 (1), 61–66.
- [25] Jianfeng S, Tie L & Yu L (2012). One-step solid state preparation of reduced graphene oxide. *Carbon* 50, 2134-2140.
- [26] Saikia BK, Boruah RK & Gogoi PK (2009). A X-ray diffraction analysis on graphene layers of Assam coal. *Journal of Chemical Science* 121, 103-106.
- [27] Zhou Z, Zhao Z, Zhang Y, Meng B, Zhou A & Qiu J (2012). Graphene sheets from graphitized anthracite coal: preparation, decoration and application. *Energy and Fuels* 26, 5186-5192.
- [28] Ruan G, Sun Z & Peng Z (2011). Growth of graphene from food, insects and waste. *American Chemical Society Nano* 5, 7601-7607.
- [29] Li S, Chungui T, Meitong L, Xiangying M, Lei W, Ruihong W, Jie Y & Honggang F (2013). From coconut shell to porous graphene-like nano-sheets for high-power supercapacitors. *Journal of Materials Chemistry A* 1, 6462–6470.
- [30] Yating Z, Guoyang L, Jiangtao C, Anning Z, Xiaoqian Z & Jieshan Q (2013). Preparation of graphene from Taixi anthracite and its photocatalyst performance for CO₂ conversion. *Journal of Nanoengineering and Nanosystems* 228, 161–164.
- [31] Samit M, Mahiuddin S & Borthakur PC (2001). Demineralization and Desulfurization of Subbituminous Coal with Hydrogen Peroxide. *Energy & Fuels* 15, 1418-1424.
- [32] Linares SA, Martin GI, Salinas MC & Serrano TB (2000). Activated carbons from bituminous coal: effect of mineral matter content. *Fuel* 79, 635–643.
- [33] Cheng KW, Guo JW & Jin FD (2013). Controlled functionalization of graphene oxide through surface modification with acetone. *Journal of Material Science* 48, 436–3442.