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GRAPHENE PRODUCED WITH USING SURFACTANT FROM EXPANDED GRAPHITE

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ABSTRACT
Graphene is a single layer of graphite with hexagonal structure which have stacked layers. Graphene has recently been recognized by its researchers due to it have sp^2 bonds in hexagonal lattice, the two-dimensional structure of a single atomic thickness, and its superior electrical, electrochemical, optical, thermal, mechanical properties and lightness. Liquid phase exfoliation (LPE) is the most promising method of graphene research to produce high quality graphene in the production of single or low layer graphene layers. In addition, LPE is an easy and inexpensive method as well as enables the production of higher capacity graphene. LPE method in the production of graphene is the use of surfactant mainly to make water suitable as a means of exfoliation. Furthermore, graphene adsorbable surfactants provide an effective charge by electrostatic repulsion to prevent the re-aggregation of graphene sheets, thus providing stabilization against the recombination of suspended graphene layers. Because 1,2-Dichlorobenzene or ortho-dichlorobenzene (O-DCB) is both effective as solvent for graphene synthesis, it is one of the best production of graphene by the liquid phase exfoliation method. In this study, graphite powders were mixed in an acid solution of H_2SO_4 and HNO_3 for 12h. The resulting powder was washed with distilled water until a neutral pH was obtained and then subjected to thermal treatment to obtain expanded graphite. The resulting powder was then mixed in an ultrasonic homogenizer in a mixture of O-DCB for 2h using 50 % strength and then it was examined by Transmission electron microscope (TEM) and X-ray photoelectron spectroscopy (XPS).

Keywords: *Graphene, ortho-dichlorobenzene, Expanded Graphite, LPE.*

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1. INTRODUCTION

Graphene is known as a surprising material in recent years due to its new features related to its two-dimensional structure (Monajjemi, 2017). Its intriguing properties such as high strength, an optical transmittance of 97,7 %, carriermobility as high as 200,000 $cm^2 V^{-1} s^{-1}$ at room temperature and a Young's modulus of 0.5–1 TPa, perfect electrical conductivity and high thermal conductivity make it promising for various applications (Arao *et. al.*, 2017). Due to these features of graphene, flexible devices, high-frequency transistors, energy storage and transducers, sensors, biomedical applications and the production of new generation composites have been demonstrated that graphene can be made easily (Wei, *et. al*, 2015). Today, the most widely used methods in the production of graphene; mechanical exfoliation, chemical method (reduction of graphene oxide), chemical vapor deposition (CVD), epitaxial growth in silicon carbide, liquid phase exfoliation method (LPE), electrochemical exfoliation, the solvothermal method, arc discharge (Lee, *et. al*., 2019)

Commercialization of graphene-oriented applications is inevitable, but cost-effective and high-quality serial production is needed (Xu, *et. al*., 2014). Since the first day it was found and applied, the liquid phase exfoliation method has been proposed as the most promising method of graphene research to produce high quality single or low layer graphene sheets in solvent dispersion forms. The main advantage of this process is a simple and sizable process in which pure graphite or expanded graphite is applied directly to a solvent process to weaken the Van Der Waals tensile forces between graphene interlayers (Durge, *et. al.,* 2014). Many successes have been achieved in the production of graphene by liquid phase exfoliation method by using numerous solvent systems and suitable surfactants (Huo , *et. al*.2015). Although the Liquid Phase Exfoliation Method is a relatively easy and inexpensive method, the quality of the products produced is higher than the products produced by other methods (Zhu, *et. al*., 2013).

In this study, graphite powders were mixed in an acid solution of H_2SO_4 and HNO_3 for 12h. The resulting powder was washed with distilled water until a neutral pH was obtained and then subjected to thermal treatment to obtain expanded graphite. The resulting powder was then mixed in an ultrasonic homogenizer in a mixture of O-DCB for 2h using 50 % strength. The main purpose of this study is to investigate the effects of O-DCB which is used as solvent in graphene synthesis by liquid phase exfoliation method in graphene production.

2. EXPERIMENTAL PROCEDURE

Hexagonal graphite powders was waited at 90 ºC for 2 h to remove the humidity. The dried HG was mixed with saturated acid consisting of concentrated H_2SO_4 and HNO_3 for 12 h to form the graphite intercalated compound (GIC). It was carefully washed with distilled water. Then, GIC was heated at 1000 ºC to form expanded graphite (EG).

The graphene-sheets were still bonded with weak van der Waals forces at some points in EG. Therefore, EG was incurred to a final exfoliation to obtain graphene-sheet by ultra-sonication in O-DCB. The suspension of 0.07 mg/ml concentration of EG with O-DCB was prepared. The solution of EG was kept at multi-frequency ultra-sonication homogenizer for 1h. Then, the mixture was centrifuged at 5000 rpm for 8 hours to remove O-DCB. The low density material suspended at the top layer of centrifuged solution was collected for further characterization.

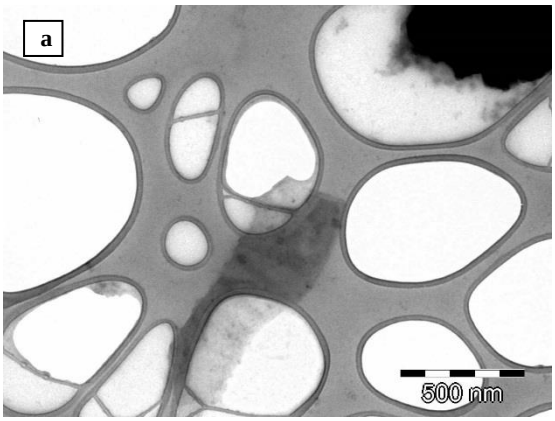
Transmission electron microscope (HRTEM) (JEOL Jem 1100) was used to investigate the microstructure of graphene samples. The samples were chractersized via X-ray photoelectron spectroscopy (XPS) (Specs-Flex) and Raman Spectroscopy (WITech alpha 300-R 512 nm wavelenght).

3. RESULTS AND DISCUSSION

Graphite consists of a stack of flat graphene-layer. Bonding between layers is via weak van der Waals bonds, which allows layers of graphite to be easily separated, or to slide past each other. The aim of this study is exfoliation of graphite layers in order to product the graphene. A solvent is necessary to separate the layers. The distance of layers should be sufficient in order to enter the solvent atoms between layers. So, hexagonal graphite powders were expanded by acid and thermal treatments. After those processes, the gap between hexagonal sheets extends and the weak bonds between the sheets easily break off via entering-solvent-atoms. Following this process increases the distance between the hexagonal layers and solvent atoms easily fill these gaps, provide the rupture of the weak bonds between the layers.

In previous our studies, different solvents (Such as DMF, NMP eg.) were used for graphene synthesis via Liquid Phase Exfoliation (LPE). It was shown in literature that O-DCB also is suitable a solvent because of its surface properties.

TEM images of samples were given after LPE process by using O-DCB as solvent. As the figure shows that, exfoliation was succeed and layers were separated from each other's. The width of the layers was 200 nm above. There is a dark-colored contamination in figure 1.a. It is believed that the contamination was sediments of solvent while using the production of amorphous carbon or graphene.



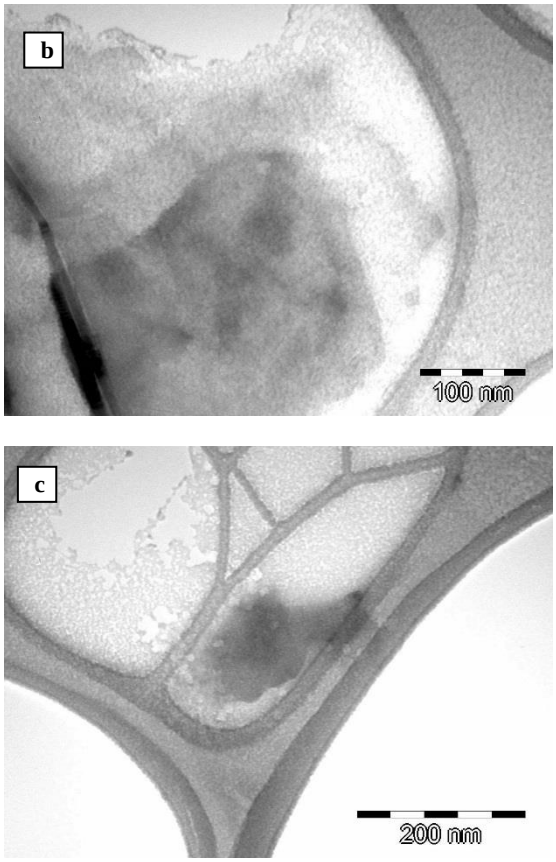
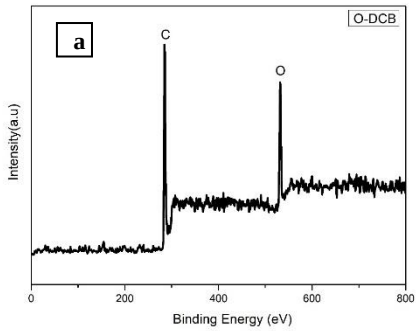


Fig. 1. (a),(b),(c) TEM micrographs of high magnification of synthesized graphene layers

In figure 2, XPS spectra analysis of samples were given. Figure 2a. shows general XPS spectra, Figure 2b. shows fitting of XPS peaks. As the figure shows that, synthesized graphenes contain C-C bonds over 80 %. In addition, C-O bonds exist in samples. We think that C-O bonds formed when EG was produced. Because, graphite powders were treated with acids and powders were heated to high temperature.



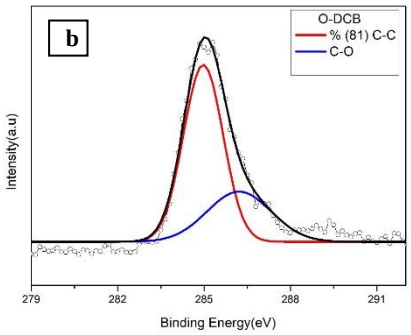


Fig. 2. (a) general XPS spectra, (b) XPS spectra fitting of synthesized Graphene layers

Figure 3 shows the Raman spectra of synthesized graphenes. In Raman spectra of graphenes, 3 characteristic peaks were shown. D peak (at 1350 cm^{-1}) relate to defects in graphenes (in sheets basal plane or sheet edges). G peak (at 1582 cm^{-1}) relates to C atoms vibrations having sp^2 hibritazition in hexagonal lattice. 2D peak (at 2700 cm^{-1}) indicates graphenes existence in powders. In Raman spectra of our samples, these peaks exist. The intensity of 2D peak is weak. This show that not only graphene exist in samples but also graphene nanosheets. The intensity of D peak is comparatively high. This show that defects exist in synthesized powders. We think that these defect formed during ultrasonic process.

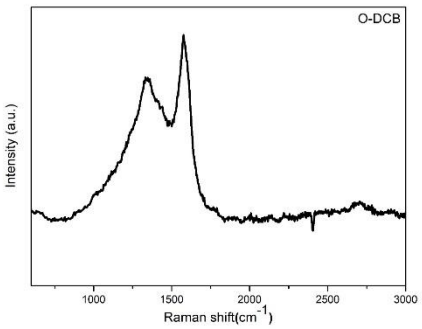


Fig. 3. Raman Spectra of synthesized graphene layers

In this study, the analyzes show that production of graphene nanosheets were achieved with Liquid-Phase Exfoliation method by using O-DCB as solvent.

4. CONCLUSION

In this study, O-DCB was used as solvent in graphene synthesis via LPE method. In first step, EG was produced. Subsequently, EG was sonicated in O-DCB in order to synthesis graphene. It shown that graphene was synthesized by using O-DCB. But, graphene nanosheets also form together with graphene in samples.

Acknowledgments

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