



## Synthesis of Functionalized Single graphene Sheets by thermal exfoliation of Graphite Oxide

ABDUL JABAR MOHMMED SALEH AL-EYANI<sup>3</sup> and NABIL ABDULLAH NOMAN ALKADASI<sup>1,2,3</sup>

<sup>1</sup>Hubei Key Lab of Materials Chemistry & Service Failure, School of Chemistry & Chemical Engineering, Huazhong University of Science and Technology, Wuhan, Hubei, 430074 China .

<sup>2</sup>Department of Chemistry, Faculty of Education, Al-baida'a , University, Yemen , P.O.Box:39189.

<sup>3</sup>Mechanical Engineering, Faculty of Engineering, Thamar University, Yemen.

\*Corresponding author E-mail: nalkadasi@yahoo.com

<http://dx.doi.org/10.13005/ojc/300405>

(Received: September 24, 2014; Accepted: November 03, 2014)

### ABSTRACT

Synthesis is described to produce single sheets functionalized graphene through thermal exfoliation of graphite oxide. Synthesis yields a single sheet structure resulting from the reaction sites involved in oxidation and reduction process. Application of graphite used for unelectrical material and used pencil meanwhile main applications of graphene sheets are electrical materials.

**Key words:** Graphite, Graphene oxide and Graphene sheets, Characterization and application.

### INTRODUCTION

Graphene is a single hexagonally flat layer of graphite, which has attracted great interest both for fundamental understanding of its unique structural and electronic properties and for important potential applications in nano-electronics and devices<sup>1-6</sup>. The unique properties of this two-dimensional (2D) material include the highest intrinsic carrier mobility at room temperature of all known materials and very high mechanical strength and thermal stability<sup>7-10</sup>. Graphene holds great promise for the development of new composite materials, emissive displays, ultrasensitive

detectors and micromechanical resonators<sup>11-13</sup>. The combination of high mobility, thermal, chemical and mechanical stability with the high surface area offers many interesting applications in a wide range of fields including heterogeneous catalysis where metallic and bimetallic nano-particle catalysts can be efficiently dispersed on the graphene sheets<sup>14-16</sup>. In many cases, the remarkable properties of single graphene layers extend to bilayers and a few layers of graphene sheets. Several methods have been reported for the production of graphene sheets including micromechanical cleavage and thermal expansion of graphite .

## EXPERIMENTAL

### Materials

Physical parameters of Graphite 99.95% , Potassium Chlorate, Sulfuric acid  $H_2SO_4$  and Nitric acid ( $HNO_3$ ), 99.9 % are reported in table 1, 2 ,3 and 4 respectively.

### Fabrication of graphene sheets

Commercial powdered natural graphite (from Aladdin Industrial Corporation Shanghai, China ) was used as our starting material. The commercial graphite has a particle size of 325 mesh with a purity of 99.99% . Graphite was oxidized following modified Method of Staudenmaier method to form graphite oxide (GO). In this method, graphite (2.5g) was first mixed with sulfuric acid (43.75 mL) and nitric acid (22.5 mL) and stirred. When graphite was uniformly dispersed, potassium chlorate (27.5 g) was added slowly and stirred for over 96 h. After the completion of oxidation reaction, the mixture was added into 4 L deionized water and then filtered. The GO was repeatedly rinsed and redispersed in a 5% solution of HCl. It was then washed continually with deionized water until the pH of the filtrate was neutral<sup>17-33</sup>. Potassium stayed in the GO even after several times of washing .Therefore multiple washing cycles used in conjunction with bath ultrasonication in fresh ethanol were used for the removal of residual potassium . After this the GO was dried in a vacuum oven at 60 °C until used. Finally, the GO was treated with the nitrogen ( $N_2$ ) and then it was heated to 1050 °C in the furnace for 30 s to form graphene sheets<sup>16-31</sup>.

### Treatment silicon wafer

silicon wafer were cut into ( 3mm x 3mm ) used Piranha solution is mixture consisting of sulfuric acid ( $H_2SO_4$ ) and hydrogen peroxide ( $H_2O_2$ ) is to cleaning silicon wafer . It is typically mixed in concentration ratios of around 3:1  $H_2SO_4:H_2O_2$  for one hour with temperature 60 °C . Then followed by triple rinsing in ethanol with ultrasonic cleaning for 30 min then with nitrogen ( $N_2$ ) . There are two main applications for piranha in wafer fabrication: it is used to remove organic contaminants from surface of the wafer during cleaning . Silicon wafer were used for SEM<sup>32</sup>.

## RESULTS AND DISCUSSION

The GO was reduced to graphene sheet by heating to 1150 °C. Plate 1 ,2( SEM ) and plate 7 (TEM) shows the top-view TEM images of the graphite oxide plate ( SEM ) 3 , 4 and plate 8 (TEM). TEM images of the original graphite plate

**Table 1: General Characteristics of Graphite 99.95%**

Trade Name	Graphite 99.95 %
Appearance	Black
Size	325 mesh
Company	Aladdin Industrial Corporation Shanghai, China

**Table 2: General characteristics of Potassium Chlorate**

Trade Name	KClO <sub>3</sub>
Appearance	White
Molecular weight	122.55
Company	China

**Table 3: General Characteristics of Sulfuric Acid  $H_2SO_4$**

Molecular formula	Sulfuric Acid( $H_2SO_4$ ), 99.9 %
Appearance	liquid
Molecular weight	98.08
Concentration	95 - 98 %
Company	Sinopharm chemical reagent Co ,Ltd ,China

**Table 4. General Characteristics of Nitric Acid ( $HNO_3$ ), 99.9 %**

Molecular formula	Nitric Acid( $HNO_3$ ), 99.9 %
Appearance	liquid
Molecular weight	63.01
Concentration	65 – 68 %
Company	Sinopharm chemical reagent Co ,Ltd ,China



Photo 1: Graphite in water



Photo 2: GO green before dry  
GO.dry in 60°C water



Photo 3: Graphene Sheet in water

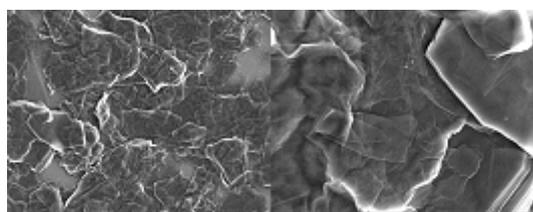


Plate 1: SEM of Graphene sheets  
dispersed in water

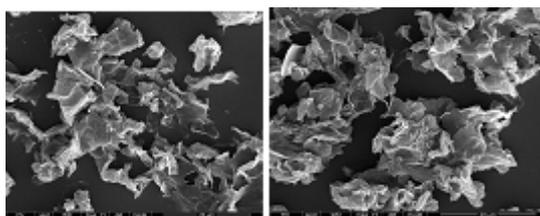


Plate 2: SEM of Graphene  
sheets dispersed in ethanol

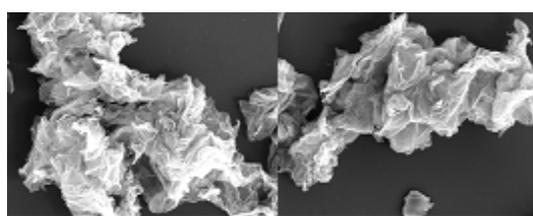


Plate 3: SEM of Graphene Oxide  
(Dry) dispersed in water

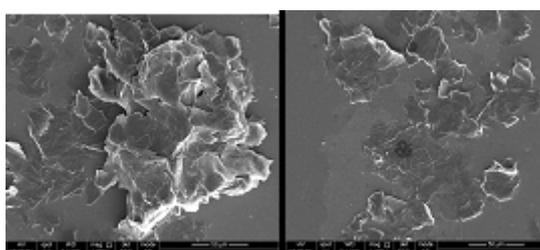


Plate 4: SEM of Graphene  
Oxide ( Dry ) dispersed in ethanol

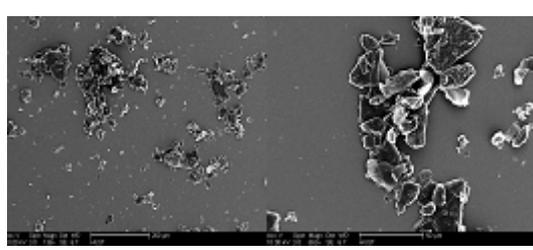
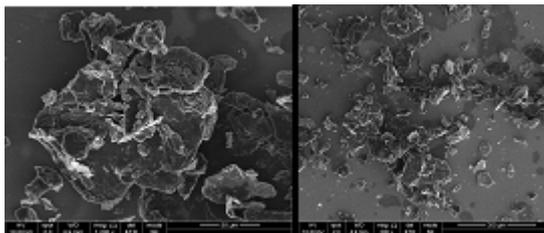
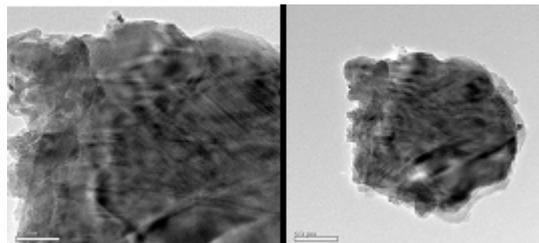


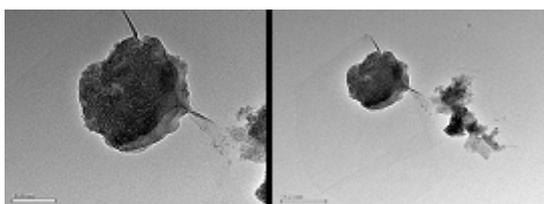
Plate 5: SEM of Graphite dispersed in water



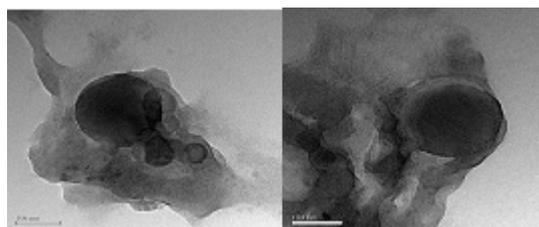
**Plate 6: SEM of Graphite dispersed in ethanol**



**Plate 7: TEM of Graphene sheets**



**Plate 8: TEM of Graphene Oxide**



**Plate 9: TEM of Graphite**

(SEM) 5,6 and plate 9 (TEM). A small flake of graphite particle is seen in . The size of the graphene sheet is about 9.35 $\mu\text{m}$  from SEM photo. The surface of graphene shows several large meandering wrinkles. The thickness of graphene can be clear in high-magnification TEM image.

application of graphene sheets in electrical material. The surface of graphene sheets shows several large meandering wrinkles. The thickness of graphene can be determined from the high-magnification TEM image .

### CONCLUSIONS

Single sheets functionalized graphene through thermal exfoliation of graphite oxide . Main

### ACKNOWLEDGEMENTS

I acknowledge the financial support from UNESCO/Government of China (Great wall) and Al-baida'a ,University , Yemen .

### REFERENCES

1. K.S .Novoselov, A.K .Geim, S.V. Morozov, D. Jiang, Y. Zhang, S.V .Dubonos, et al. Electric field effect in atomically thin carbon films. *Science* **2004**; 306: 666–9.
2. Novoselov KS, Jiang D, Schedin F, Booth TJ, Khotkevich VV, Morozov SV, et al. Two-dimensional atomic crystals. *Proc Natl Acad Sci USA* **2005**;102: 10451–3.
3. Geim AK, Novoselov KS. The rise of graphene. *Nat Mater* **2007**; 6:183–91.
4. Novoselov KS, Geim AK, Morozov SV, Jiang D, Zhang Y, Dubonos SV, et al. Twodimensional gas of massless Dirac fermions in grapheme. *Nature* **2005**; 438: 197-202.
5. Partoens B, Peeters FM. From graphene to graphite: electronic structure around the K point. *Phys Rev B* **2006**; 74: 075404.
6. Ghorbani, H.R., *Orient J Chem.*, **2014**, 30(2), 803-806.
7. Graf D, Molitor F, Ensslin K, Stampfer C, Jungen A, Hierold C, et al. Spatiallyresolved Raman spectroscopy of single- and few-layer graphene. *Nano Lett* **2007**;7:238–42.
8. Stankovich S, Dikin DA, Dommett GHB, Kohlhaas KM, Zimney EJ, Stach EA, et al. Graphene-based composite materials. *Nature* **2006**; 442: 282–6.
9. McAllister MJ, Li JL, Adamson DH, Schniepp HC, Abdala AA, Liu J, et al. Single sheet

- functionalized graphene by oxidation and thermal expansion of graphite. *Chem Mater* **2007**; *19*: 4396–404.
10. Stankovich S, Piner RD, Nguyen ST, Ruoff RS. Synthesis and exfoliation of isocyanate-treated graphene oxide nanoplatelets. *Carbon* **2006**; *44*: 3342–7.
  11. Stankovich S, Piner RD, Chen X, Wu N, Nguyen ST, Ruoff RS. Stable aqueous dispersions of graphitic nanoplatelets via the reduction of exfoliated graphite oxide in the presence of poly(sodium 4-styrenesulfonate). *J Mater Chem* **2006**; *16*: 155–8.
  12. Li X, Zhang G, Bai X, Sun X, Wang X, Wang E, et al. Highly conducting graphene sheets and Langmuir–Blodgett films. *Nat Nanotechnol* **2008**; *3*: 538–42.
  13. Hernandez Y, Nicolosi V, Lotya M, Blighe FM, Sun ZY, De S, et al. High-yield production of graphene by liquid-phase exfoliation of graphite. *Nat Nanotechnol* **2008**; *3*: 563–9.
  14. Lotya M, Hernandez Y, King PJ, Smith RJ, Nicolosi V, Karlsson LS, et al. Liquid phase production of graphene by exfoliation of graphite in surfactant/water solutions. *J Am Chem Soc* **2009**; *131*: 3611–20.
  15. Pu NW, Wang CA, Sung Y, Liu YM, Ger MD. Production of few-layer graphene by supercritical CO<sub>2</sub> exfoliation of graphite. *Mater Lett* **2009**; *63*: 1987–9.
  16. Liu N, Luo F, Wu H, Liu Y, Zhang C, Chen J. One - step ionic - liquid - assisted electrochemical synthesis of ionic-liquid-functionalized graphene sheets directly from graphite. *Adv Funct Mater* **2008**; *18*: 1518–25.
  17. Li D, Müller MB, Gilje S, Kaner RB, Wallace GG. Processable aqueous dispersion of graphene nanosheets. *Nat Nanotechnol* **2008**; *3*: 101–5.
  18. Liu Z, Robinson JT, Sun X, Dai H. PEGylated nano-graphene oxide for delivery of water insoluble cancer drugs. *J Am Chem Soc* **2008**; *130*: 108767.
  19. Wu H, Wang J, Kang X, Wang C, Wang D, Liu J, et al. Glucose biosensor based on immobilization of glucose oxidase in platinum nanoparticles/graphene/chitosan nanocomposite film. *Talanta* **2009**; *80*(1): 403–6.
  20. Stoller MD, Park S, Zhu Y, An J, Ruoff RS. Graphene-based ultracapacitors. *Nano Lett* **2008**; *8*: 3498–502.
  21. Hsiao MH, Liao SH, Yen MY, Teng CC, Lee SH, Pu NW, et al. Preparation and properties of a graphene reinforced nanocomposite conducting plate. *J Mater Chem* **2010**; *20*: 8496–505.
  22. Liu Y, Ren L, He Y, Cheng HP. Titanium-decorated graphene for high-capacity hydrogen storage studied by density functional simulations. *J Phys Condens Matter* **2010**; *22*: 445301.
  23. Schniepp HC, Li JL, McAllister MJ, Sai H, Herrera-Alonso M, Adamson DH, et al. Functionalized single graphene sheets derived from splitting graphite oxide. *J Phys Chem B* **2006**; *110*: 8535–9.
  24. Viculis LM, Mack JJ, Mayer OM, Hahn HT, Kaner RB. Intercalation and exfoliation routes to graphite nanoplatelets. *J Mater Chem* **2005**; *15*: 974–8.
  25. Elias DC, Nair RR, Mohiuddin TMG, Morozov SV, Blake P, Halsall MP, et al. Control of graphene's properties by reversible hydrogenation: evidence for graphane. *Science* **2009**; *323*: 610–3.
  26. Rao CNR, Biswas K, Subrahmanyam KS, Govindaraj A. Graphene, the new nanocarbon. *J Mater Chem* **2009**; *19*: 2457–69.
  27. Saito R, Fujita M, Dresselhaus G, Dresselhaus MS. Electronic structure of chiral graphene tubules. *Appl Phys Lett* **1992**; *60*: 2204–6.
  28. Liang Y, Wu D, Feng X, Müller MB, Kaner RB. Dispersion of graphene sheets in organic solvent supported by ionic interactions. *Adv Mater* **2009**; *21*: 1679–83.
  29. Li D, Müller MB, Gilje S, Kaner RB, Wallace GG. Processable aqueous dispersions of graphene nanosheets. *Nat Nanotechnol* **2008**; *3*: 101–5.
  30. Paredes JI, Villar-Rodil S, Martínez-Alonso A, Tasco'n JMD. Graphene oxide dispersions in organic solvents. *Langmuir* **2008**; *24*: 10560–4.
  31. Li JT, Li M, Li JH, Sun HW. Removal of disperse blue 2BLN from aqueous solution

- by combination of ultrasound and exfoliated graphite. *Ultrason Sonochem* **2007**; *14*:62–6.
32. Shen J, Hu Y, Li C, Qin C, Ye M. Synthesis of amphiphilic graphene nanoplatelets. *Small* **2009**; *5*: 82–5.
33. Yizhi W, Cheng Z, Ye Y, Ziwen W, Weijia S, Huijie W and Xiaoliang X. Fabrication of wafer – Zize Monolayer close –packed colloidal crystals via slope self –assembly and thermal treatment . *Langmuir* **2013**; *29*: 14017-14023.