Journal of Tropical Resources and Sustainable Science

journal homepage: jtrss.org

Facile preparation of graphene oxide silver aerogel for antibacterial

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Received 18 July 2017
Accepted 12 September 2017
Online 24 June 2018

Keywords:
graphene-metal oxide, nanocomposites, antibacterial

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Abstract

We report a simple and straightforward method for the preparation of graphene oxide-silver aerogel (GOAg) via microwave irradiation. Graphene oxide (GO) was used as a substrate for the growth of silver nanoparticles (AgNPs) and silver complex was used as a precursor. The GOAg were prepared by rapidly expose mixture of GO and silver complexes with microwave for 30 s under an ambient atmosphere. For the nanocomposites, GOAg solution were lyophilized in a freeze-dryer for 24 hours to form an aerogel. The obtained GO and GOAg nanocomposites were characterized by X-ray diffractometer (XRD) and Ultraviolet-visible spectroscopy (UV-Vis). XRD confirmed the formation GO, and GOAg while GOAg display antibacterial properties against Methicillin-resistant *Staphylococcus aureus* (MRSA) and *Aeromonas hydrophila* bacteria. We demonstrate a cost effective and easy synthesis of a GOAg with promising antibacterial properties.

1. INTRODUCTION

Graphene, a single atomic layer of sp^2 carbon atoms with a honeycomb crystal structure has attracted enormous attention from the scientific community around the globe due to its unique electrical, mechanical and thermal properties (Dikin et al., 2007). These properties also can be enhanced by composite with other materials. The studies on nanocomposites materials have attracted numerous researcher due to their intensifying properties. The consolidation of silver and graphene as nanocomposite acquire an interesting structure and has potential in various applications for instance antibacterial (de Faria et al., 2014; D. Zhang et al., 2011; Zhu et al., 2013), transparent and conductive film (Kahng et al., 2014), electrochemical immunosensor (Qu et al., 2011), catalysis (Wen et al., 2012), and sensors (Yola et al., 2014). Numerous techniques, including chemical reduction, hydrothermal, chemical vapour deposition and electromagnetic radiation (Hassan et al., 2009; Williams et al., 2008) have been used to prepare graphene. Among these methods, electromagnetic radiation has received significant attention because of its utility in the large-scale production for industry use. However, most of the irradiation sources are either limited or dangerous due to the high frequency. Hence, exploring new approaches are essential. Herein, a facile method was modified for the simple, easy and large scale of GOAg nanocomposite by microwave irradiation on aqueous solution and lyophilized processing produces a homogenous Ag nanoparticle embedded on graphene oxide sheets. This simple, fast and easy method provides an effective path for the preparation of large scale a GOAg nanocomposites.

2. MATERIALS AND METHODS

2.1. Synthesis of graphene oxide

Graphene oxide (GO) was prepared using the simplified Hummers method (Hummers Jr et al., 1958; Lim et al., 2011). 3 g of graphite was mixed into solution of H_2SO_4 and H_3PO_4 (4:1) respectively followed by $KMnO_4$. The mixture was stir for a three days period before poured onto 400 ml ice cube along with 27 ml of H_2O_2 (30%). The mixture was centrifuge at 10 000 g until the mixture reaching pH5 and use for further action.

eISSN Number: 2462-2389

2.2. Synthesis of reduced graphene oxide silver aerogel

Briefly, 100 ml ammonia (25 wt%) was slowly added into 20 mM of 10 ml of silver nitrate solution. The mixture was vigorously stirred until a clear solution was observed indicating the formation of complex before mixed with 0.5 mg/mL of aqueous GO solution (volume ratio 3:1) and sonicated for 5 min to ensure the homogeneity of the mixture. The reaction mixture was exposed to the microwave irradiation for 30 s. The final product was centrifuged at a centrifugal force of 10,000 g and washed with deionized water for three times before transfer into freeze-drying to form an aerogel.

2.3. Characterizations

The samples were characterized by Evolution™ 600, a UV-visible spectrophotometer (UV-Vis) within a range of 200-800 nm. XRD pattern of the products was recorded on a D5000 Siemen X-ray diffractometer in a 2θ range of 5-80º with monochromatic Cu Kα source (λ=1.5406 Å). The morphology and size of the samples were examined by high resolution transmission electron microscopy HRTEM (Philips CM12) operated at 100 kV.

2.4. Antibacterial test of silver-graphene oxide aerogel

All the apparatus used in this test were sterilized in an autoclave at 140 °C with pressure at 1 bar for 5 h. The Methicillin-resistant *Staphylococcus aureus* (MRSA), *Aeromonas hydrophila*, Flavobacterium spp. Salmonella spp. Streptococcus spp. and Psedomonas spp. isolated was provided by Aquaculture Research Lab, Universiti Malaysia Kelantan. The bacterial were cultured in tryptic soy broth (TSB) for 24 h at room temperature. The concentration of the colony were fixed to 10^9 CFU/ ml and cross check with an ELISA reader.

2.5. Minimum inhibitory concentration (mic) values determination

Minimum inhibitory concentration (MIC) values were determined using two-fold micro broth dilution method in 96-wells microliter plate format. The GOAg aerogel were cut into small pieces (20 mm in diameter) before were added into bacterial suspensions which inoculated into wells with the kanamycin as a control. The MIC value was determined by measure the lowest concentration of antimicrobial agent inhibits the visible growth of the inoculated bacteria after 24 h incubation.

3. RESULTS AND DISCUSSION

The UV-visible absorption spectra of GO and GOAg aerogel are shown in Fig. 1. As shown in the Fig. 1(a) the absorption peak of GO displays two bands. The highest absorption band centred at 228 nm is assigned to the π → π* transition of aromatic C-C bonds and the second shoulder band at 304 nm corresponds to n → π* transition of C=O bonds (Zhang et al., 2012). As to GOAg (Fig. 1b), after the irradiation the peak was disappeared, indicating the small reduction on the surface of GO (Baby et al., 2011). These suggest the restored electronic conjugation within graphene sheets and the removal of oxygen-containing functional groups after microwave irradiation (Pasricha et al., 2009; Xu et al., 2011; Zainy et al., 2012). The appearance of the band at 420 nm in Fig. 1(b) is attributed to the surface plasmon resonance of Ag nanoparticles which evince the presence of Ag nanoparticles on graphene oxide (Zhang et al., 2011).

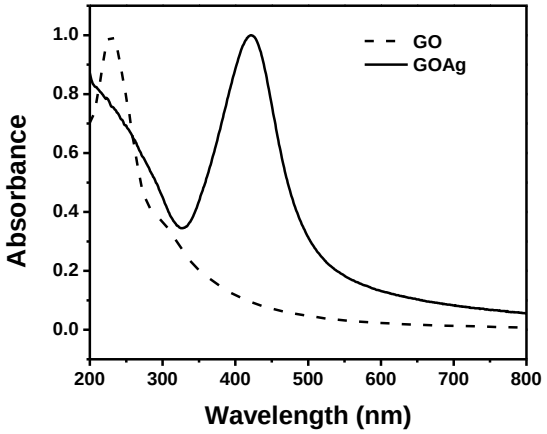

Figure 1: UV-Vis absorption of (a) GO and (b) GOAg aerogel.

The XRD spectra of the synthesized sample are shown in Fig. 2(a). From the pattern it can be seen that the characteristic GO peak centred at 10.0º corresponds to *d*-spacing of 8.29 Å, which is attributed to the (001) reflection of GO. After the irradiation of microwave on the GO containing silver complexes, the peak at 10.0º reduced while a sharp peak at 38.1º, 42º, 65º and 79º appeared, which suggests that the Ag particle had been embedded to GO layer after 30 s (Fig. 2(b)). As shown in the figure 2 (b), the XRD pattern of the GOAg can be indexed with the JCPDS file 087-0597, which correspond to the (111), (200), (220) and (311) planar crystallographic of face-centred cubic (fcc) silver nanoparticles. The formation of high purity crystalline Ag nanoparticles in the nanocomposites were confirm by a sharp and intense diffraction peak at 38.1º corresponds to crystalline Ag.

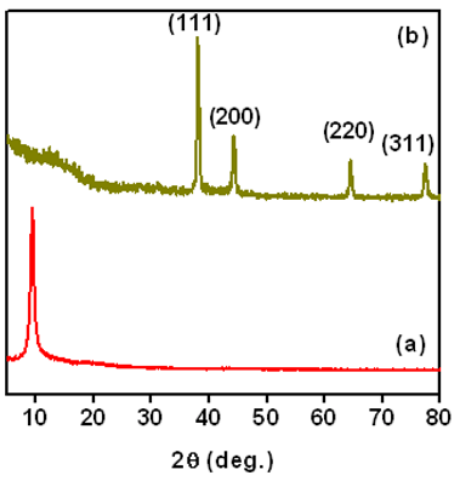

Figure 2: XRD patterns of the (a) GO and (b) GOAg aerogel.

eISSN Number: 2462-2389

The morphological of the samples were characterized using the HRTEM as shown in Fig. 3. The images demonstrate that the thin layer of GO produced almost transparent and the area of the surface was up to micrometre. After the introduction of $Ag(CH_3O_2)$ the surface of GO was deposited with Ag nanoparticles of less than 20 nm in size. Based on the finding above, we postulated that the Ag precursor was intercalated within the GO layers during the grinding process. The microwave irradiation provides sufficient energy to the electrons in GO to be excited to the Ag ions, simultaneously reducing Ag ions to metallic Ag.

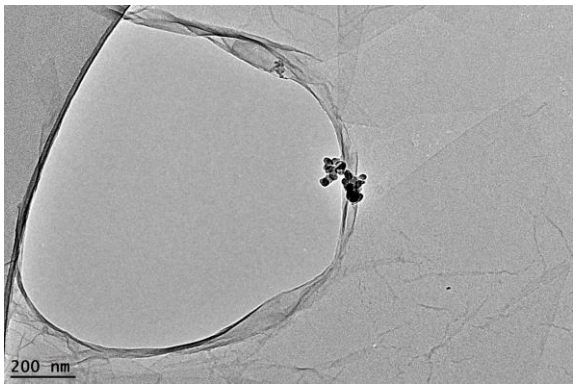

Figure 3: Transmission electron microscopy image of GOAg aerogel.

Table 1: Minimum Inhibitory Concentration (MIC) values of GOAg aerogel against bacterial

Bacteria species	Minimum Inhibitory Concentration of Kanamycin (mg/L)	Minimum Inhibitory Concentration of GOAg aerogel (mg/L)
Staphylococcus aureus	30.5	60.5
Aeromonas hydrophila	61.5	100.0
Flavobacterium spp.	33.5	50.0
Salmonella spp.	31.5	30.0
Streptococcus spp	16.3	40.5
Pseudomonas spp.	35.5	30.5

Table 1 shows the screening of bacterial growth in soy broth. Compared to the Kanamycin the bacterial could not survive in the presence of GOAg aerogel. The finding of the present study showed that GOAg inhibit the growth of various bacterial species, hence may justify the importance of GOAg in aquaculture uses. The increasing of antibiotic resistance case among pathogenic bacteria due to extensive used of commercial antibiotic in managing aquatic human health led to most of the antibiotic was no longer effective in controlling bacterial diseases. Therefore, it is crucial to explore an alternative way to apply as an antimicrobial agent. Furthermore, this study was successfully documented inhibitory activity of GOAg aerogel against various types of bacteria.

4. CONCLUSION

Graphene oxide-Ag aerogel was successfully synthesized by microwave irradiation for 30 s in ambient atmosphere. The data revealed that the Ag ions was reduced to Ag nanoparticles simultaneously deposited on the graphene oxide sheet with the size of 20 nm in size. This method clearly offers that a simple and cost-effective approach for the preparation of graphene oxide silver aerogel and can be apply as antimicrobial.

ACKNOWLEDGEMENT

This work was supported financially by the SGPL-MyRA grant no: R/MyRA/A08.00/00648A/002/2016/000378 from Universiti Malaysia Kelantan, Transdisciplinary Research Grant Scheme (TRGS) grant no: R/TRGS/A0.700/00387A/007/2016/000391 and the Fundamental Research Grant Scheme (FRGS) grant no: R/FRGS/A08.00/00648A/001/2016/000368 from the Ministry of Higher Education Malaysia.

REFERENCES

Baby, T. T., & Ramaprabhu, S. (2011). Synthesis and nanofluid application of silver nanoparticles decorated graphene. *Journal of Materials Chemistry, 21*(26), 9702-9709.

Dai, C., Yang, X., & Xie, H. (2011). One-step synthesis of reduced graphite oxide-silver nanocomposite. *Materials Research Bulletin, 46*(11), 2004-2008.

de Faria, A. F., Martinez, D. S. T., Meira, S. M. M., de Moraes, A. C. M., Brandelli, A., Filho, A. G. S., & Alves, O. L. (2014). Anti-adhesion and antibacterial activity of silver nanoparticles supported on graphene oxide sheets. *Colloids and Surfaces B: Biointerfaces, 113*(0), 115-124.

Dikin, D. A., Stankovich, S., Zimney, E. J., Piner, R. D., Dommett, G. H., Evmenenko, G., . . . Ruoff, R. S. (2007). Preparation and characterization of graphene oxide paper. *Nature, 448*(7152), 457-460.

Hassan, H. M. A., Abdelsayed, V., Khder, A. E. R. S., Abouzeid, K. M., Terner, J., El-Shall, M. S., . . . El-Azhary, A. A. (2009). Microwave synthesis of graphene sheets supporting metal nanocrystals in aqueous and organic media. *Journal of Materials Chemistry, 19*(23), 3832-3837.

Hummers Jr, W. S., & Offeman, R. E. (1958). Preparation of graphitic oxide. *Journal of the American Chemical Society, 80*(6), 1339.

Kahng, Y. H., Kim, M.-K., Lee, J.-H., Kim, Y. J., Kim, N., Park, D.-W., & Lee, K. (2014). Highly conductive flexible transparent electrodes fabricated by combining graphene films and inkjet-printed silver grids. *Solar Energy Materials and Solar Cells, 124*(0), 86-91.

Lim, H. N., Huang, N. M., Lim, S. S., Harrison, I., & Chia, C. H. (2011). Fabrication and characterization of graphene hydrogel via hydrothermal approach as a scaffold for preliminary study of cell growth. *International journal of nanomedicine, 6*, 1817-1823.

Pasricha, R., Gupta, S., & Srivastava, A. K. (2009). A facile and novel synthesis of Ag-graphene-based nanocomposites. *Small (Weinheim an der Bergstrasse, Germany), 5*(20), 2253-2259.

eISSN Number: 2462-2389

Qu, F., Lu, H., Yang, M., & Deng, C. (2011). Electrochemical immunosensor based on electron transfer mediated by graphene oxide initiated silver enhancement. *Biosensors and Bioelectronics*, *26*(12), 4810-4814.

Wen, C., Shao, M., Zhuo, S., Lin, Z., & Kang, Z. (2012). Silver/graphene nanocomposite: Thermal decomposition preparation and its catalytic performance. *Materials Chemistry and Physics*, *135*(2–3), 780-785.

Williams, G., Seger, B., & Kamt, P. V. (2008). TiO2-graphene nanocomposites. UV-assisted photocatalytic reduction of graphene oxide. *ACS Nano*, *2*(7), 1487-1491.

Xu, Z., Gao, H., & Guoxin, H. (2011). Solution-based synthesis and characterization of a silver nanoparticle-graphene hybrid film. *Carbon*, *49*(14), 4731-4738.

Yola, M. L., Gupta, V. K., Eren, T., Şen, A. E., & Atar, N. (2014). A novel electro analytical nanosensor based on graphene oxide/silver nanoparticles for simultaneous determination of quercetin and morin. *Electrochimica Acta*, *120*(0), 204-211.

Zainy, M., Huang, N. M., Vijay Kumar, S., Lim, H. N., Chia, C. H., & Harrison, I. (2012). Simple and scalable preparation of reduced graphene oxide–silver nanocomposites via rapid thermal treatment. *Materials Letters*, *89*(0), 180-183.

Zhang, D., Liu, X., & Wang, X. (2011). Green synthesis of graphene oxide sheets decorated by silver nanoprisms and their anti-bacterial properties. *Journal of Inorganic Biochemistry*, *105*(9), 1181-1186.

Zhang, Y., Ma, H. L., Zhang, Q., Peng, J., Li, J., Zhai, M., & Yu, Z. Z. (2012). Facile synthesis of well-dispersed graphene by γ-ray induced reduction of graphene oxide. *Journal of Materials Chemistry*, *22*(26), 13064-13069.

Zhang, Z., Xu, F., Yang, W., Guo, M., Wang, X., Zhang, B., & Tang, J. (2011). A facile one-pot method to high-quality Ag-graphene composite nanosheets for efficient surface-enhanced Raman scattering. *Chemical Communications*, *47*(22), 6440-6442.

Zhu, Z., Su, M., Ma, L., Ma, L., Liu, D., & Wang, Z. (2013). Preparation of graphene oxide–silver nanoparticle nanohybrids with highly antibacterial capability. *Talanta*, *117*(0), 449-455.

eISSN Number: 2462-2389