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RESEARCH PAPER

Synthesis of Magnetic Graphene-Fe_3O_4 Nanocomposite by Electrochemical Exfoliation Method

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ABSTRACT

Superparamagnetic few-layer graphene nanocomposite (FLG-NC) can be used for many technological applications. In this work, we applied electrochemical exfoliation method as a simple, one step and economical technique to fabricate FLG-NC. The fabricated superparamagnetic FLG-NC were characterized by X-ray diffraction (XRD), Fourier transform infrared (FTIR) spectroscopy, scanning electron microscopy (SEM), field emission scanning electron microscope (FESEM), energy dispersive spectroscopy (EDS) and vibrating sample magnetometer (VSM). Analysis of XRD pattern shows the formation of graphene iron oxide nanocomposite due to the existence of magnetite (Fe_3O_4) and graphite picks. The SEM image show that the number of layers in nanocomposite is between 3 and 10. According to the results of SEM and XRD, the nanocomposite is FLG-Fe_3O_4. Hysteresis curve of the sample and size of ferromagnetic particles reduced to a critical size to represents superparamagnetic properties with saturation magnetization M_S=57.3 emu. g^{-1}. The results are useful for preparation of large magnetic moment superparamagnetic nanocomposite.

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INTRODUCTION

Graphene is a known and specific material with high electrical conductivity, high surface area and favourable biocompatibility. It has shown as a potential material to be used in technological applications such as nanoelectronics [1], sensors [2], batteries [3] and supercapacitors [4].

Large specific surface area and planar shape made graphene to be a good candidate for acting matrix role in nanocomposite. By using magnetic fillers for this matrix, the nanocomposite will be magnetized that is very useful in controlled targeted drug delivery [5], cancer drug delivery agent [6] and removal of toxic metal and dyes from aqueous solutions [7]. Therefore, production of magnetic nanocomposite without obvious toxicity is an important task for many applications. Moreover, between various types of magnetic materials, iron oxides (Fe_3O_4 and γ-Fe_2O_3) are the best choices because of their special magnetic properties, low toxicity and low cost [8].

Graphene nanosheets have often been made from expanded graphite which according to the number of layers named as few-layer graphene (FLG) and multi-layer graphene (MLG) [9].

There are several methods to produce graphene-iron oxide nanocomposite [10]. Electrochemical exfoliation method as a simple, one step and economical technique was our choice to fabricate magnetic graphene nanocomposite [11]. In this research, we used an electrochemical exfoliation method and an ammonium sulfate iron salt to produce superparamagnetic few layer graphene-iron oxide nanocomposite (FLG-IONC) representing high magnetization and superparamagnetic properties.

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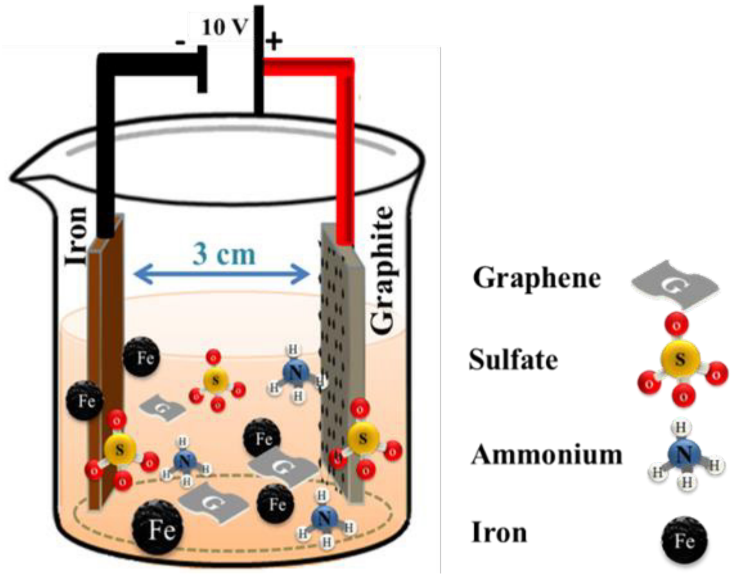

Fig. 1. Schematic representation of FLG-IONC production

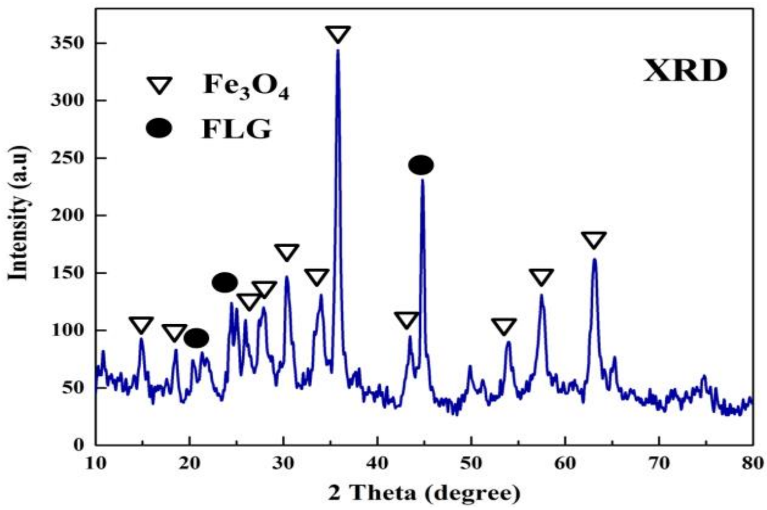

Fig. 2. XRD diffraction pattern of FLG- Fe_3O_4

MATERIALS AND METHODS

Iron (II) ammonium sulfate [$(NH_4)_2Fe(SO_4)_2.6H_2O$] and hydrochloric acid (HCl) were prepared from Merck. Graphite foil and iron plate are used as electrodes.

The fabricated FLG-IONC was characterized by X-ray diffraction (XRD) with Cu K**α radiation** between 10 and 80°. Fourier transform infrared (FTIR) spectroscopy in an optical range of 400–4000 cm^{-1}, scanning electron microscopy (SEM), field emission SEM (FESEM), energy dispersive spectroscopy (EDS) and vibrating sample magnetometer (VSM) techniques were applied to study the nanocomposite.

Synthesis of superparamagnetic FLG-IONC

For the production of FLG-IONC, firstly 0.1 M solution was prepared with stirring 7.8 g iron (II) ammonium sulfate in 200 ml distilled water. We used graphite foil as anode and iron plate as cathode with 3 cm distance between the electrodes. Applied voltage was 10 V DC for 3 hours, as shown schematically in Fig. 1. After the fabrication, magnetic nanocomposite products were separated by magnet and rinsed several times with distilled water and dried at 100 ° C.

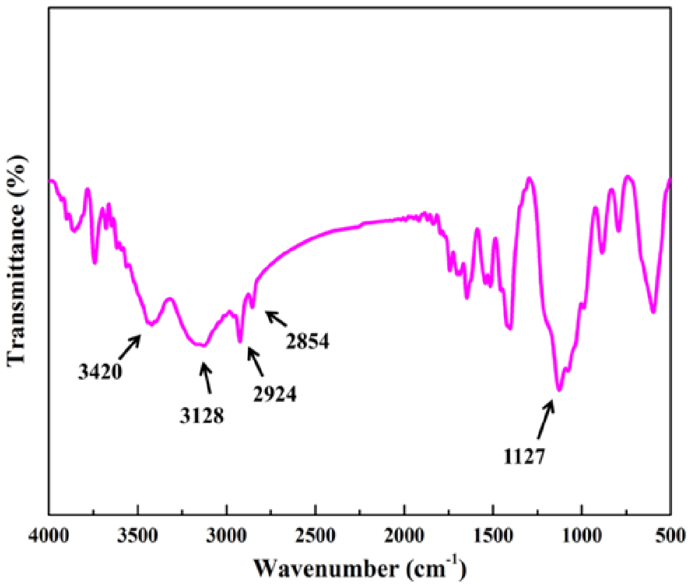

Fig. 3. FTIR spectrum of FLG-Fe_3O_4

RESULTS AND DISCUSSION

Fig. 2 shows XRD pattern that most of the iron oxide particles are magnetite and maghemite (Fe_3O_4 and γ-Fe_2O_3) according to the peaks in 14, 18, 26, 27, 30, 35, 43, 49, 53, 57, 63°. Characteristics and properties of iron oxide phases were reported by Ramimoghadam et al [12], and according to their review, our sample with black/ brownish black colour is Fe_3O_4 (magnetite). Also, peaks in 2θ=20, 25 and 44° are related to the presence of FLG. According to these acheivements, the name of the sample is FLG-Fe_3O_4.

The FTIR spectrum of our final production sample is given in Fig. 3. Absorption peak at 3420 cm^{-1} is related to stretching vibration of N-H [13]. The peak at 3128 cm^{-1} is related to stretching vibration of the coordinated –OH group [14]. The CH_2 bands at 2924 cm^{-1} and 2854 cm^{-1} can be observed. A strong absorption peak at 1127 cm^{-1} is related to double band S=O or single band C-O.

Fig. 4 shows the hysteresis loop of FLG-Fe_3O_4 representing the fact that the sample is superparamagnetic and the specific saturation magnetization (M_S) is 57.3 emu.g^{-1}. As shown in the insets of Fig. 4, the magnetic FLG-Fe_3O_4 particles are suspended in the solution (left side) and after being exposed to an external magnetic field, the magnetic FLG-Fe_3O_4 nanocomposite is separated from the solution (right side).

Fig. 5a and Fig. 5b show SEM and FESEM images of FLG-Fe_3O_4. The FLG configuration can be seen clearly in Fig. 5a. Also in Fig. 5b we can see the distribution of iron oxide particles on the surface. Fig. 5c is a SEM image of sample in a 500 nm window. In this picture, iron oxide nanoparticles in the range of 44 nm, 58 nm and 98 nm can be observed. The EDS test of FLG-Fe_3O_4 is given and shown in Fig. 5d and Fig. 5e. Inset tables in this picture show the mass percent of Fe, O and C at the specified points EDS A and EDS B, determined from panel b. We compared the mass percent of each element between tables. It is found that EDS A based on high mass percentage of iron and oxygen with low carbon is related to iron oxide. The EDS B addresses the FLG that contains iron oxide particles between graphene sheets,Therefore, results of XRD, SEM, FESEM and EDS indicate the formation of FLG-Fe_3O_4. Furthermore, from the results of the VSM, it is found that the FLG-Fe_3O_4 nanocomposite show superparamagnetic like properties and because of its high saturation magnetization it is favourable for the use in different applications.

CONCLUSIONS

Superparamagnetic FLG-Fe_3O_4 nanocomposite was successfully synthesized by simple, one step and economical electrochemical exfoliation method. Our product is formed as FLG with layer number between 3 and 10 with embedded magnetic nanoparticles of Fe_3O_4. Accessible, industrial and cost-effective iron cathode makes

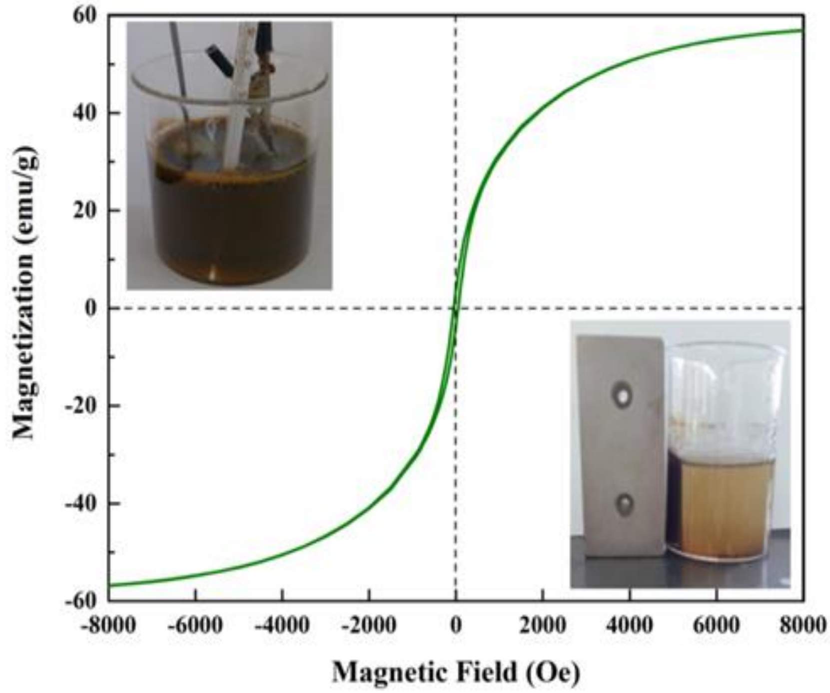

Fig. 4. Magnetic hysteresis loop of FLG- Fe_3O_4. The insets are the images of FLG- Fe_3O_4 dispersions before (top left) and after (bottom right) exposing to an external magnetic field

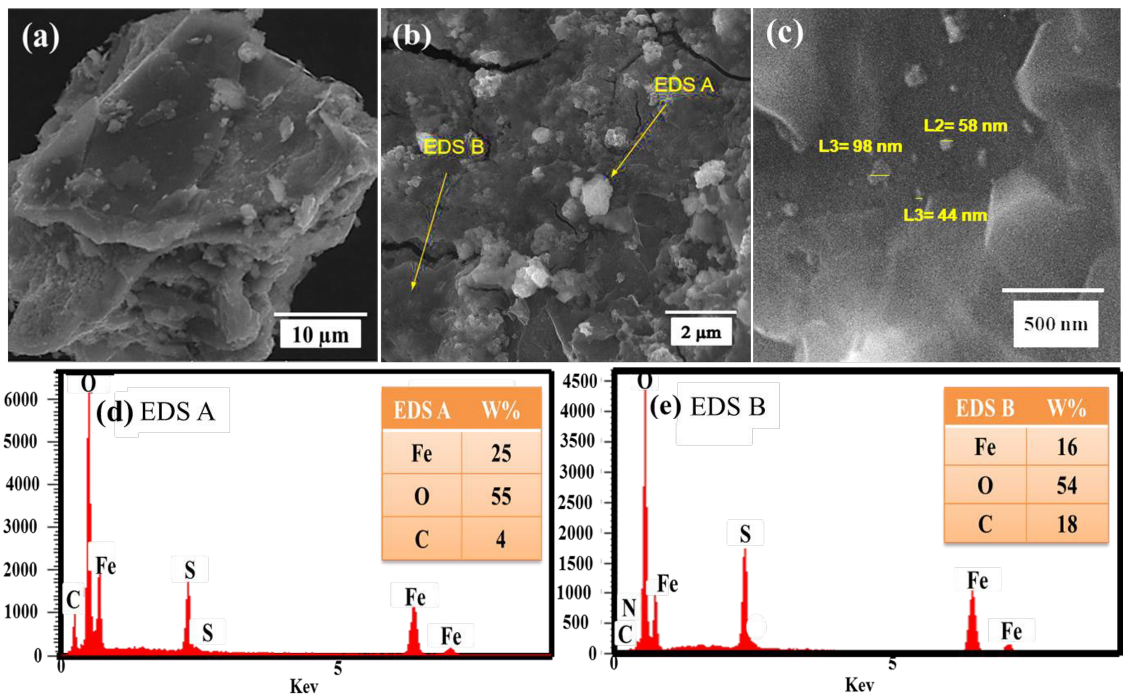

Fig. 5. (a) SEM (b) FESEM (c) SEM image of FLG- Fe_3O_4 in 500 nm (d) EDS of point EDS A (e) EDS of point EDS B

it possible to be applied in mass production of FLG-Fe_3O_4 samples. The specific saturation magnetization of FLG-Fe_3O_4 is 57.3 $emu.g^{-1}$ which is suitable for many applications such as hyperthermia, drug delivery, supercapacitors, etc.

CONFLICT OF INTEREST

The authors declare that there is no conflict of interests regarding the publication of this manuscript.

REFERENCES

1. Blake P, Brimicombe PD, Nair RR, Booth TJ, Jiang D, Schedin F, et al. Graphene-Based Liquid Crystal Device. Nano Letters. 2008;8(6):1704-8.
2. Zhang Y-H, Chen Y-B, Zhou K-G, Liu C-H, Zeng J, Zhang H-L, et al. Improving gas sensing properties of graphene by introducing dopants and defects: a first-principles study. Nanotechnology. 2009;20(18):185504.
3. Yoo E, Kim J, Hosono E, Zhou H-s, Kudo T, Honma I. Large Reversible Li Storage of Graphene Nanosheet Families for Use in Rechargeable Lithium Ion Batteries. Nano Letters. 2008;8(8):2277-82.
4. Stoller MD, Park S, Zhu Y, An J, Ruoff RS. Graphene-Based Ultracapacitors. Nano Letters. 2008;8(10):3498-502.
5. Yang X, Zhang X, Ma Y, Huang Y, Wang Y, Chen Y. Superparamagnetic graphene oxide–Fe3O4 nanoparticles hybrid for controlled targeted drug carriers. Journal of Materials Chemistry. 2009;19(18):2710.
6. Pooresmaeil M, Namazi H. β-Cyclodextrin grafted magnetic graphene oxide applicable as cancer drug delivery agent: Synthesis and characterization. Materials Chemistry and Physics. 2018;218:62-9.
7. Pooresmaeil M, Mansoori Y, Mirzaeinejad M, Khodayari A. Efficient Removal of Methylene Blue by Novel Magnetic Hydrogel Nanocomposites of Poly(acrylic acid). Advances in Polymer Technology. 2016;37(1):262-74.
8. Arévalo P, Isasi J, Caballero AC, Marco JF, Martín-Hernández F. Magnetic and structural studies of Fe3O4 nanoparticles synthesized via coprecipitation and dispersed in different surfactants. Ceramics International. 2017;43(13):10333-40.
9. Choi W, Lahiri I, Seelaboyina R, Kang YS. Synthesis of Graphene and Its Applications: A Review. Critical Reviews in Solid State and Materials Sciences. 2010;35(1):52-71.
10. Young RJ, Kinloch IA, Gong L, Novoselov KS. The mechanics of graphene nanocomposites: A review. Composites Science and Technology. 2012;72(12):1459-76.
11. Du Y, Luo X-L, Xu J-J, Chen H-Y. A simple method to fabricate a chitosan-gold nanoparticles film and its application in glucose biosensor. Bioelectrochemistry. 2007;70(2):342-7.
12. Ramimoghadam D, Bagheri S, Hamid SBA. Progress in electrochemical synthesis of magnetic iron oxide nanoparticles. Journal of Magnetism and Magnetic Materials. 2014;368:207-29.
13. Wen Q, Wang Y, Xu K, Li N, Zhang H, Yang Q, et al. Magnetic solid-phase extraction of protein by ionic liquid-coated Fe@graphene oxide. Talanta. 2016;160:481-8.
14. Chrissafis K, Paraskevopoulos KM, Papageorgiou GZ, Bikiaris DN. Thermal and dynamic mechanical behavior of bionanocomposites: Fumed silica nanoparticles dispersed in poly(vinyl pyrrolidone), chitosan, and poly(vinyl alcohol). Journal of Applied Polymer Science. 2008;110(3):1739-49.