

## Tuning of magnetic behaviour in nitrogenated graphene oxide functionalized with iron oxide

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### ABSTRACT

Two-dimensional graphene oxide (GO) nanomaterials offer interesting physical/chemical properties and have been explored for potential use in electronics, magnetic, catalysis, and energy storage applications, because of its high charge-carrier mobility and high specific surface area. This study investigates the electronic and magnetic properties of nitrogenated graphene oxide ( $GO-N_x$ ), and nitrogenated graphene oxide functionalized with iron oxide ( $GO-N_x:Fe$ ). Four different nitrogen (N) precursor viz. ammonium hydroxide ( $NH_4OH$ ), hexamethylenetetramine ( $C_6H_{12}N_4$ ), acetonitrile ( $C_2H_3N$ ), and carbamide ( $CH_4N_2O$ ) are used to synthesis " $GO-N_x$ " using chemical route. As  $C_6H_{12}N_4$  based- $GO-N_x$  shows higher ferromagnetism, thus it is further functionalized with iron oxide using three different iron (Fe) precursors viz. ferric oxide ( $Fe_2O_3$ ), ferroso-ferric oxide ( $Fe_3O_4$ ), and iron oxide-hydroxide ( $FeOOH$ ) to tune this room temperature ferromagnetism (RTFM). The electronic structure of " $GO-N_x$ " and " $GO-N_x:Fe$ " are characterized using C 1s, O 1s, N 1s and Fe 2p core-level X-ray photoelectron spectroscopy (XPS) and their corresponding magnetic behaviours are correlated with the SQUID-measured M-H loops. The magnetic moment changes due to the conversion and formation of different nitrogen bonded carbon along with the different phase of iron oxide. The tuning of magnetization in " $GO-N_x$ " and " $GO-N_x:Fe$ " using different N-precursors and/or Fe-precursors is an efficient route to tailor the electronic/magnetic properties of graphene-oxide for different electronic and magnetic device applications.

### 1. Introduction

The emergence of magnetic response in non-magnetic graphene oxide (GO) due to different defects has stimulated extensive research interest that may lead to the novel magnetic device applications [1]. Magnetism in GO and its derivatives is of particular interest since the lightweight magnet could open up the new ways to design adaptable and flexible information storage system [2]. However, the subject of magnetism in graphene-related materials remains controversial. The room temperature ferromagnetism have been observed in reduced-GO [3], nitrogen functionalized-GO [3] and hydrogenated graphene [4]. This creates an imbalance in the crystal lattice, inducing a magnetic moment [4]. Subsequently, the theoretical and experimental studies show the coexistence of ferromagnetic response along with the anti-ferromagnetic behaviour in graphene-oxide and graphene-related materials that might be due to different defects, disorders and edge states [5–10]. However, most of the investigated dominant ferromagnetic properties originated from graphene oxide (GO). During the

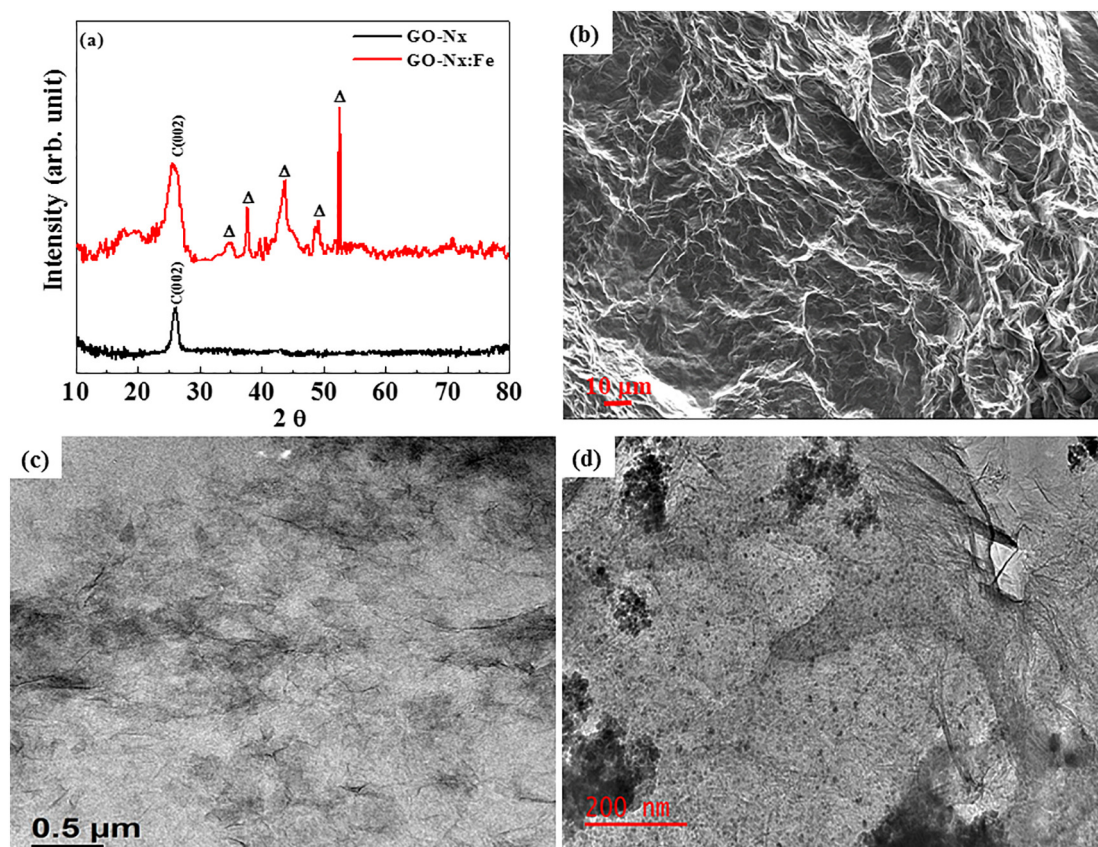
preparation of GO by the chemical process, it seems unlikely that micron-sized magnetic impurities could remain within the GO membranes. It is considered that all of these contentious make the GO as a magnetic material and that is supported by experimental as well as theoretical observations. Despite of different reports about the views and the relationship among the paramagnetism, diamagnetism and ferromagnetism in nitrogenated graphene-oxide and their composites related materials, we have further explored the magnetism of functionalized graphene-oxide materials using different N-precursor and Fe-precursors.

In our earlier report [3], we have observed that GO functionalized with nitrogen using urea, that exhibits a drastic enhancement in their magnetic response. In the present study, we have used nitrogenated-GO ( $GO-N_x$ ) functionalized with iron oxide to improve the magnetic properties of GO for different magnetic applications. Initially, we have synthesized GO by the Hummers method [11] and subsequently functionalized with nitrogen using four different N-precursor ( $NH_4OH$ ,  $C_6H_{12}N_4$ ,  $C_2H_3N$  and  $CH_4N_2O$ ) for the preparation of four different type

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**Fig. 1.** (a) X-ray diffraction of the as synthesized  $C_6H_{12}N_4$  based “GO- $N_x$ ” and  $Fe_2O_3$ -based “GO- $N_x:Fe$ ” respectively ( $\Delta$  are “Fe” and/or oxides of ‘Fe’); (b) field emission scanning electron microscopy (FESEM) image of as synthesized  $C_6H_{12}N_4$  based “GO- $N_x$ ”; (c) transmission electron microscopy (TEM) image of as synthesized  $C_6H_{12}N_4$  based “GO- $N_x$ ” and (d) TEM image of as synthesized  $Fe_2O_3$ -based “GO- $N_x:Fe$ ”.

**Table 1a**

Compositional and quantificational (at. %) analysis results obtained from X-ray photoelectron spectroscopy.

GO- $N_x$ /GO- $N_x:Fe$ composites	XPS elemental and quantification data				Magnetization	
	C (at. %)	O (at. %)	N (at. %)	Fe (at. %)	$M_s$ (emu/g)	$H_c$ (Oe)
$NH_4OH$ -based “GO- $N_x$ ”	68.6	29.6	1.8	–	$4.4 \times 10^{-3}$	43
$C_6H_{12}N_4$ -based “GO- $N_x$ ”	90.6	8.0	1.4	–	$5.7 \times 10^{-3}$	53
$C_2H_3N$ -based “GO- $N_x$ ”	81.3	17.2	1.5	–	$3.6 \times 10^{-3}$	35
$CH_4N_2O$ -based “GO- $N_x$ ”	79.8	18.7	1.5	–	$3.1 \times 10^{-3}$	31
$Fe_2O_3$ -based “GO- $N_x:Fe$ ” composites	59.7	29.8	1.7	8.8	$4.3 \times 10^{-2}$	48
$FeOOH$ -based “GO- $N_x:Fe$ ” composites	57.5	31.6	1.1	9.8	$2.9 \times 10^{-2}$	32
$Fe_3O_4$ -based “GO- $N_x:Fe$ ” composites	53.9	34.3	1.9	9.2	$1.5 \times 10^{-2}$	18

of “GO- $N_x$ ”. We have observed that  $C_6H_{12}N_4$ -based “GO- $N_x$ ” has the highest magnetization among these four types of “GO- $N_x$ ”. To tune this magnetization further, we have functionalized this  $C_6H_{12}N_4$ -based GO- $N_x$  with three different Fe-precursors ( $Fe_2O_3$ ,  $Fe_3O_4$  and  $FeOOH$ ) for the preparation of “GO- $N_x:Fe$ ” composites. The electronic structure and bonding properties of “GO- $N_x$ ” and “GO- $N_x:Fe$ ” were studied using C 1s, N 1s, O 1s and Fe 2p core-level X-ray photoelectron spectroscopy (XPS) studies have been carried out and establish a correlation with their magnetic behaviours for future electronic and magnetic device

applications.

## 2. Experimental details

Initially, GO is synthesized by the Hummers method [11], and then mixed with four different N-precursor viz.  $NH_4OH$ ,  $C_6H_{12}N_4$ ,  $C_2H_3N$  and  $CH_4N_2O$  to synthesize GO- $N_x$  in a household microwave at 700 W for 150 s. 50 ml of aqueous solution of GO (0.08 g) was prepared by ultra-sonicating for 50–60 min. Then the equal molar concentration of each N-precursor viz.  $NH_4OH$ ,  $C_6H_{12}N_4$ ,  $C_2H_3N$  and  $CH_4N_2O$  was added with GO separately and sonicated for another 5 min. The mixture was then put in a standard household microwave (2450 MHz, 700 W) and heated for 300 s at a maximum output power. The solution was then cooled to room temperature and centrifuge at 3000 rpm for 30 min. The clear supernatant obtained after centrifugation was separated. The obtained brown/black precipitate was further cleaned in deionised water and centrifuged at 3000 rpm for 30 min. The final slurry was left to dry in the vacuum oven at 50 °C overnight to obtain nitrogenated GO (GO- $N_x$ ). This treatment with nitrogen induces oxygen reduction and carbon purification, facilitating nitrogen substitution in GO- $N_x$ . After the preparation of four type of GO- $N_x$ , we have further functionalized the  $C_6H_{12}N_4$  based GO- $N_x$  with three different type of Fe-precursors viz.  $Fe_2O_3$ ,  $Fe_3O_4$  and  $FeOOH$  separately to synthesize three different “GO- $N_x:Fe$ ” composites. The “GO- $N_x:Fe$ ” composites were synthesized by co-precipitating pre-hydrolysed  $Fe_2O_3$ ,  $Fe_3O_4$  and  $FeOOH$  separately in the presence of GO. In this process, requisite amount of GO- $N_x$  (15 mg) was dispersed into de-ionised water (30 ml) by ultra-sonication for half an hour. To this suspension, 50 ml solution of  $Fe_3O_4/Fe_2O_3/FeOOH$  (50 mg each) in de-ionised water was added at the room temperature. Then the temperature of the solution was raised to 85 °C. After that the solution being stirred for 1 h and then cooled to room temperature. The

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