Supplementary Information

Rolling and unrolling of graphene sheets via in-situ generation of superparamagnetic iron oxide nanoparticles

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Experimental

Materials: Graphite powder (<20μm) was obtained from Sigma Aldrich, H_2SO_4 (98%) , 50% Hydrogen peroxide (H_2O_2), Potassium permanganate ($KMnO_4$), were purchased from Merck India. Ferrous Sulphate heptahydrate ($FeSO_4.7H_2O$, 98.5%, Merck), Ferric Chloride hexahydrate ($FeCl_3.6H_2O$,97% Sigma Aldrich) . Ammonium hydroxide (25%, Merck) Cetyl trimethyl ammonium bromide, CTAB (98%Merck) were used for the synthesis of Fe_3O_4 nanoparticles. Aqueous solutions were prepared with ultrapure water (18 MΩ). All chemicals were analytical grade and used without further purification.

Synthesis of GO

Graphene oxide (GO) was prepared by modified Hummer's method which employed acid oxidation of graphite powder at low temperature. Graphite powder was added to 98% sulphuric acid and stirred for 12 h at room temperature. Potassium permanganate (4 g) was gradually added after cooling the solution below 10 ^oC. Finally the reaction was arrested by adding 150 ml water and 30% 10 ml H_2O_2. The suspension

was washed several times with distilled water and eventually filtered using filtration apparatus. The endmost product was dried at 60 ^{0}C under vacuum for 12h.

Reduction to rGO

The GO powder (1g/ml) was suspended in 20 ml distilled water and bathsonicated for 20 minutes. The solution was then transferred to Teflon-lined stainless steel hydrothermal autoclave and was kept in air oven at 180 ^{0}Cfor 6 hours.

Preparation of Fe_3O_4 decorated graphene nanoscroll

Graphene nanoscroll was synthesised by chemical coprecipitation of Fe_3O_4 from suspended GO solution. In a typical experiment, $FeCl_3.6H_2O$ and $FeSO_4.7H_2O$ [Fe^{2+}: Fe^{3+}=1:2] were dissolved in 40 ml deionised water with vigorous stirring and was added to suspended hydrothermally reduced GO solution. To this solution source under vigorous stirring, NH_4OH was added drop-wise. Nitrogen gas was continuously purged throughout the reaction. The solution mixture was then transferred to hydrothermal autoclave and heated to 100 ^{0}C for 24 hours. Dark brown mixture of Fe_3O_4 –GNS was obtained, washed by deionized water and finally, the product was separated by filtration. Lastly the Fe_3O_4 –GNS was dried at 60 ^{0}C under vacuum.

Material characterisation

Transmission Electron Microscopy (TEM) images were obtained using HRTEM JEOL JEM -2100. XPS measurements were performed on PHI 5000 Versa Probe using Al Kα x-ray source.The XRD patterns were taken by High Resolution X-Ray diffractometer (PANalytical x'pert PRO) equipped with Cu-Kα as radiation source (ʌ = 1.5405 A^0) with a step width of 0.05^0. Magnetic measurements of Fe_3O_4 nanoparticles were done using Quantum Design make, Versa lab, and physical property measurement system. The hysteresis curve was measured under a magnetic field strength sweeping from -15 to +15 kOe at room temperature.

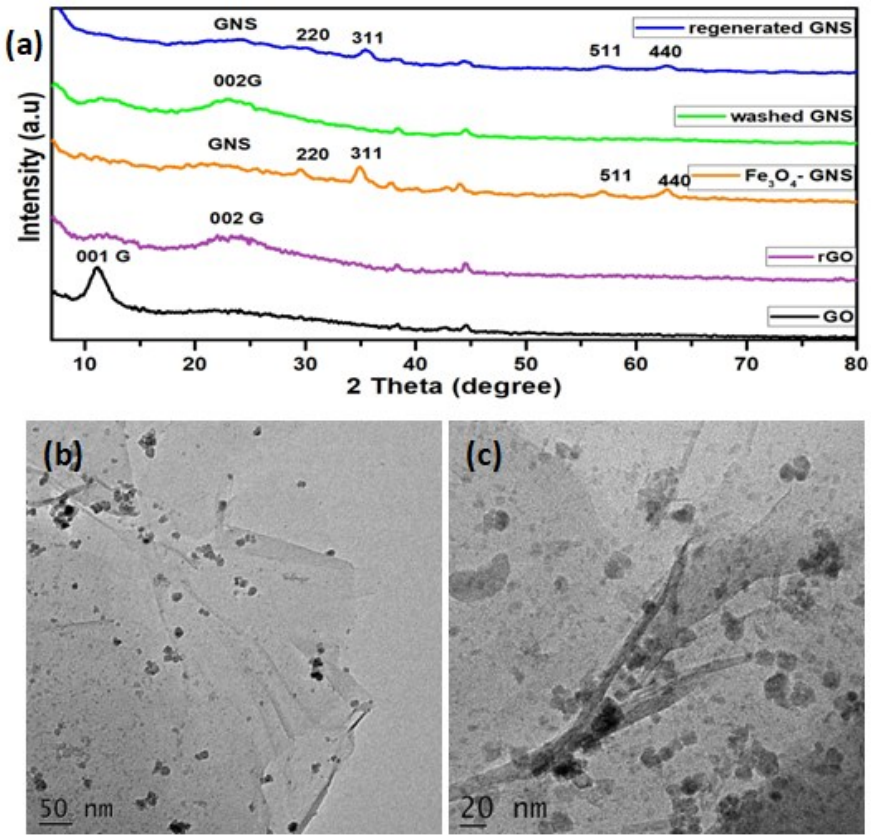

Figure S1 (a) XRD images of GO, rGO, Fe_3O_4 – GNS, GNS washed using HCl to remove iron oxide nanoparticles, regenerated GNS by decoration with Fe_3O_4 (b) TEM image of GO (without hydrothermal reduction)- Fe_3O_4 hybrid.

The XRD pattern of GO shows peaks at 11^0 and 44.4^0 corresponding to 001 and 101 diffraction planes respectively. After hydrothermal reduction, a broad peak at 24.3^0 developed indicating the deep reduction of GO. The peaks at 30.1^0, 35.5^0, 57.1^0, 62.6^0 corresponds to (220), (311), (511) and (440) planes of Fe_3O_4 (JCPDS 75-0033) while the broad peak at 24^0 corresponds to GNS. After washing, all peaks corresponding to magnetite disappeared indicating complete removal of iron oxide from the hybrid.

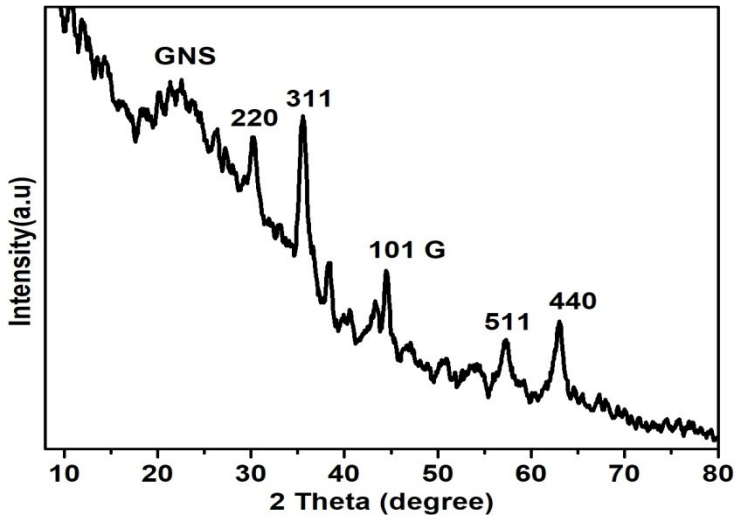

Figure S2 XRD pattern of Fe_3O_4-GNS

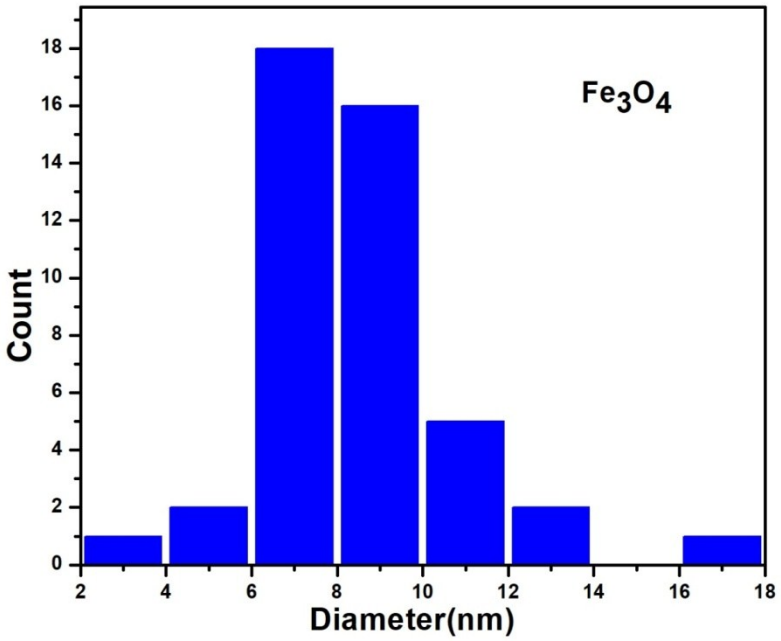

Figure S3 Histogram of statistical analysis for Fe_3O_4 nanoparticles in Fe_3O_4-GNS

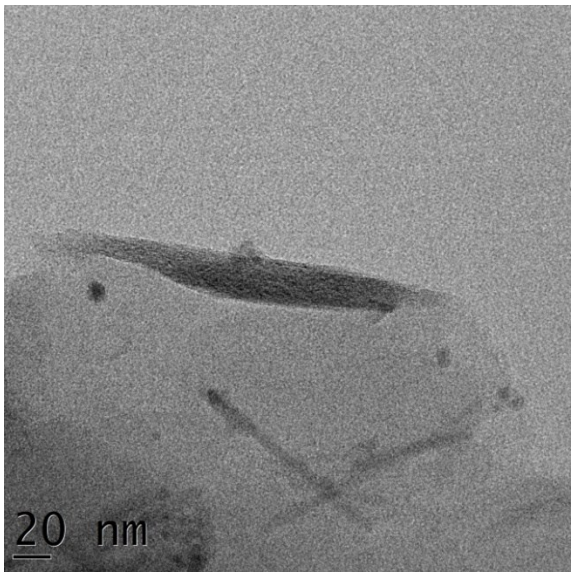

Figure S4 A portion of graphene sheet tending to scroll

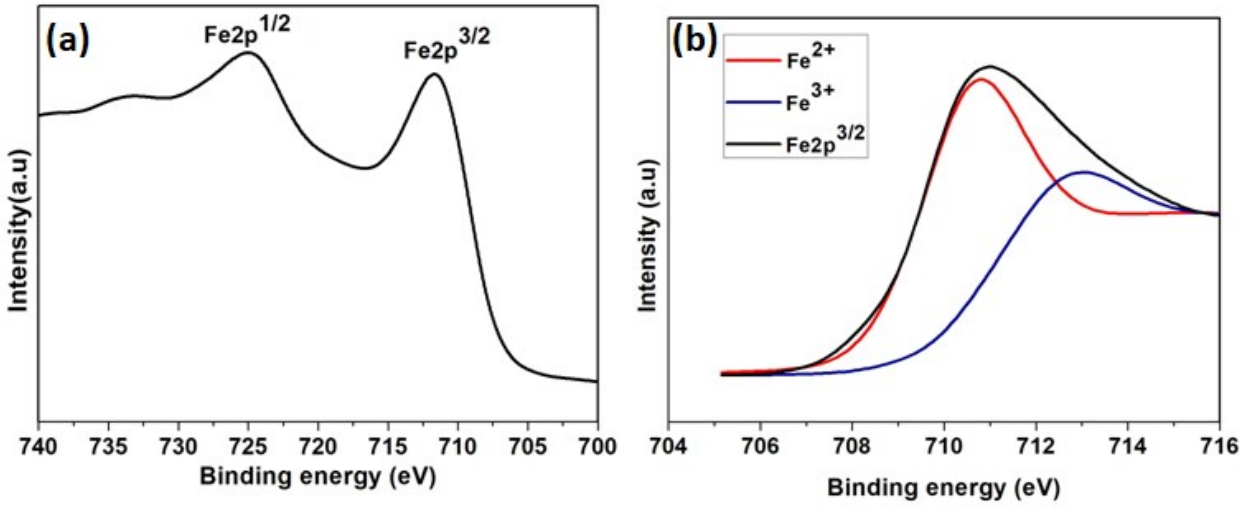

Figure S5 (a) Fe $2p^{1/2}$ and $2p^{3/2}$ peaks in the XPS spectra of Fe_3O_4 nanoparticles. (b) The deconvoluted Fe2p $^{3/2}$ spectra

XPS spectra given in Figure S5 display bands assigned to Fe $2p^{1/2}$ at 724 eV and Fe $2p^{3/2}$ at 710 eV in Fe_3O_4 . The spectral deconvolution of Fe $2p^{3/2}$ gives two signals of Fe^{2+} at 710.4 eV and Fe^{3+} at 712.5 eV.