Supporting Information

For

Green Electrochemical Synthesis of Graphene Oxide for High-Performance Electromagnetic Shields

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Table S1: Comparison of reaction parameters for GO synthesis and characteristics: the current study vs the literature

No.	Raw Material	Electrolyte	EC reaction condition	Characteristics of the products	Ref.
*	**Industrial grade natural graphite**	**0.05 M Nitric acid**	**8 V for 250 min**	**No graphite peak: 100% efficiency XRD main peak 10° = high polarity**	**This work**
1	High-quality pyrolytic graphite sheet	1M nitric acid	Pretreatment: 10 V for 30 s Main treatment: Raising voltage from 0 to 8 V with increment of 0.5 V and interval of 3 min	Thickness: 3-5 nm XRD main peaks: stronger peak: 26.35º (related to graphite and FLG) and weaker peak: 13.56º (related to GO) This result is obtained after 30 min centrifuging at 4500 rpm and used of above colloidal GO containing colloid which makes the yield less than 1%	1
2	Graphite rod obtained from Nippo Battery	Dilute H_2SO_4 solution	Constant DC voltage = 10 V for 4 hr.	XRD main peak at 26.46º (characteristic of graphite and FLG rather than graphite oxide)	2
3	Commercial graphite rod	0.1 M oxalate acid and 0.05 M Na_2SO_4 + Illumination	Pretreatment: high potential: 10 V, low potential: -0.5 V, period: 2 seconds) for 20 minutes Main treatment: 15 V, time = few seconds whole system was illuminated with a Xenon lamp light source (Power: 450 mW cm^{-2})	C/O = 2.6 XRD main peak at ~ 10º related to GO Thickness = Single layer	3

4	Expanded graphite (Prepared via immersing graphite foil in 98% H_2SO_4)	1 M KCl	Main treatment = 4 V; time = 10 hr.	C/O = 2.77 Thickness = 1-3 layers There is just EDS result for confirming the existence of GO which is not scientifically approved	4
5	Pencil core	H_2SO_4 or H_3PO_4	Pretreatment = 1 V for 3-5 min, and 7 V for 5-8 min Main treatment = Alternating voltage between + 7 V and - 7 V every 5-8 min	XRD peak 26.4º (characteristic of graphite and FLG rather than graphite oxide)	5
6	Graphite flake	0.2 M Sodium Citrate	300 mA for 5 hrs.	C/O = 7.6 (near to graphene rather than GO) XRD main peak at 10.24º (characteristic of GO)	6
7	Graphite foil	0.5 M $LiClO_4$	Pretreatment = 2 V for 2 min Main treatment = 10 V	C/O = 4 Thickness = 1-4 layers	7
8	Graphite rod	$(NH_4)_2SO_4$	1 A for 2 hr.	XRD peak 26.2º (characteristic of graphite and FLG rather than graphite oxide)	8
9	Graphite disk obtained from graphite flakes	70% $HClO_4$	Current density of 50 µA/mg	Low optical band gap (0.88 eV) lower than GO synthesized through Hummer's method (2.8 eV)	9
10	Graphite foil	Pretreatment = 98% of H_2SO_4, V = 2.2 V for 10 min Main treatment = 0.1 M $(NH_4)_2SO_4$, V = 10 V for 5-10 min	-	C/O = 4.6 Thickness = 1-2 layers XRD main peak at ~ 11º related to GO and weaker peak: 22º related to FLG Low yield due to the repeatedly (5 times) sonication and centrifugation	10
11	Flexible graphite paper	Pretreatment = immersed 98% of H_2SO_4 for 20 min Main treatment = 50 % of H_2SO_4, V = 1.6 V for 20 min	-	C/O = 1.7 Thickness = 1-3 layers XRD main peak at 11.35º	11
12	Suspension of 160-200 µm of natural graphite	2.6 g of H_2SO_4 for 2 g of graphite	450-500 mA/g of graphite	XRD main peak at ~ 26.5º related to graphite and FLG, and weaker peak: 11.45º related to GO (low yield due to the much lower intensity of GO phase than graphite phase)	12
13	Graphite foil	42% HBF_4	Current density of 180 mA/cm^2	XRD main peak compatible with GO phase; C/O ratio = 0.99; Yield > 99%	13

1. Characterization

X-ray diffraction (XRD) measurements were carried out using a PANalytical X'pert Pro MPD diffractometer with CuK_α radiation. The Bragg's ($\lambda=2d\sin\theta$) and Debye-Scherrer's laws ($t=0.89\lambda/B\cos\theta$)

were utilized to calculate the interlayer distance (d) and the crystallite size (t). In this formula, λ is the wavelength of CuK_{α}, θ is Bragg's angle, and B is the corrected full width at half maximum (FWHM). The number of layers (n) is obtained using a single equation (n=t/d). Attenuated total reflection (ATR) spectra were recorded using a Thermo Nicolet Avatar 380 in the 400-4000 cm^{-1} range. Thermo-gravimetric analysis (TGA) was conducted using a TGA TA Q600 in a nitrogen atmosphere in the 18-600 °C temperature range with a heating rate of 10 °C/min. Raman spectra were obtained using a Teksan TakRam N1-541 using an Nd: YAG laser with an excitation wavelength of 532 nm for all the samples.

UV-Vis spectral analysis was carried out using a JENWAY 6705 UV / Vis spectrometer in single-cell holder mode with the sample dispersed in type I water at a concentration of 0.2 mg/mL and compared with type I water as the reference sample. Scanning electron microscopy (SEM) images from as-synthesized GO nanosheets and graphite were recorded using the TeScan – Mira III machine. Other images were recorded from the intersection of the freeze-dried, vacuum-filtered, and solution-cast GO_{1M} samples.

Atomic force microscopy (AFM) images and surface roughness diagrams were recorded using a Veeco AUTO PROBE – CP_Research in tapping (non-contact) mode. Sample preparation was optimized at one concentration of GO sample in DI-water (0.25 mg/ml), and a small drop was dried on a mica substrate, which was cleaned via scotch tape Beforehand. X-ray photoelectron spectroscopy (XPS) was used to measure the surface composition of the GO nanoplatelets using a Bestec XPS instrument equipped with a monochromatic Al source and a multi-channeltron detector.

2. Intercalant agent matching size

Interlayer distance of graphite = 3.54 Å [14]

Nitric acid spatial size [15] = Pure nitric acid density = 1.51 g/cm^3

Molecular weight of nitric acid = 63.01 g/mol

1 mol nitric acid + free space between molecules = 41.72 cm^3

Considering simple cubic packing: PF = 0.524

Volume of 1 molecule of nitric acid = 36.3×10^{-30} m^3

Spatial size of nitric acid considering simple cubic crystals = 3.31 Å

Spatial size of nitric acid considering closed-pack solid crystals = 3.71 Å

Spatial size of nitric acid considering other crystalline packing factors < 3.31 Å

3. Cost analysis

To compare the cost of the present work with other available same materials, a table (Table S2) of the price of the used graphite and other usual graphite sheets in other works is presented; also, to show the potential of this synthesis to be applied in large scale, the total price of the synthesis of 1 g of GO powder is presented in Table S3.

Table S2: Comparing the cost of convenient HOPG graphite with used graphite

Company	Graphite type	Relative cost ($/gr)
Jiangxi DASEN [16]	Natural flexible graphite sheet	0.016
Alfa Aesar [17]	Highly Oriented Pyrolytic Graphite (HOPG)	4518

In each synthesis of GO powder, 0.2527 g of graphite was used; according to XRD results, 100 wt.% of initial graphite is converted to GO; so, for synthesizing 1 g of GO, 1 g of graphite is needed, which means five times synthesis in 75 ml electrolyte. Of course, with increased synthesis bath volume, this amount can be prepared at one step. All of the expenses to produce 1 g of GO is presented in Table 3:

Table S3: Expenses of GO sample production in this method

Materials	Expenses ($)
1.04 g of DASEN natural, flexible graphite sheet Model NO: DSN503	0.017
5 ml of Merck nitric acid	0.148
1 gallon of DI-water	0.01
Aggregate expenses	**0.175**

Comparing the cost of synthesizing 1 g of $GO_{0.2M}$ in the present work ($0.175) with 1 g of GO from Sigma-Aldrich Co., which is $155.5, proposes the current study as a promising route for large-scale production [18]. Also, by comparing the specification of the Sigma-Aldrich product with the current work, it can be understood that GO powder in the current study contains two layers; however, Sigma-Aldrich's products have 15-20 layers according to their datasheets [18].

Figure S1: Mass production of GO in 1M nitric acid: 4.2 g of graphite results in 4 g of GO sheets in just 15 min.

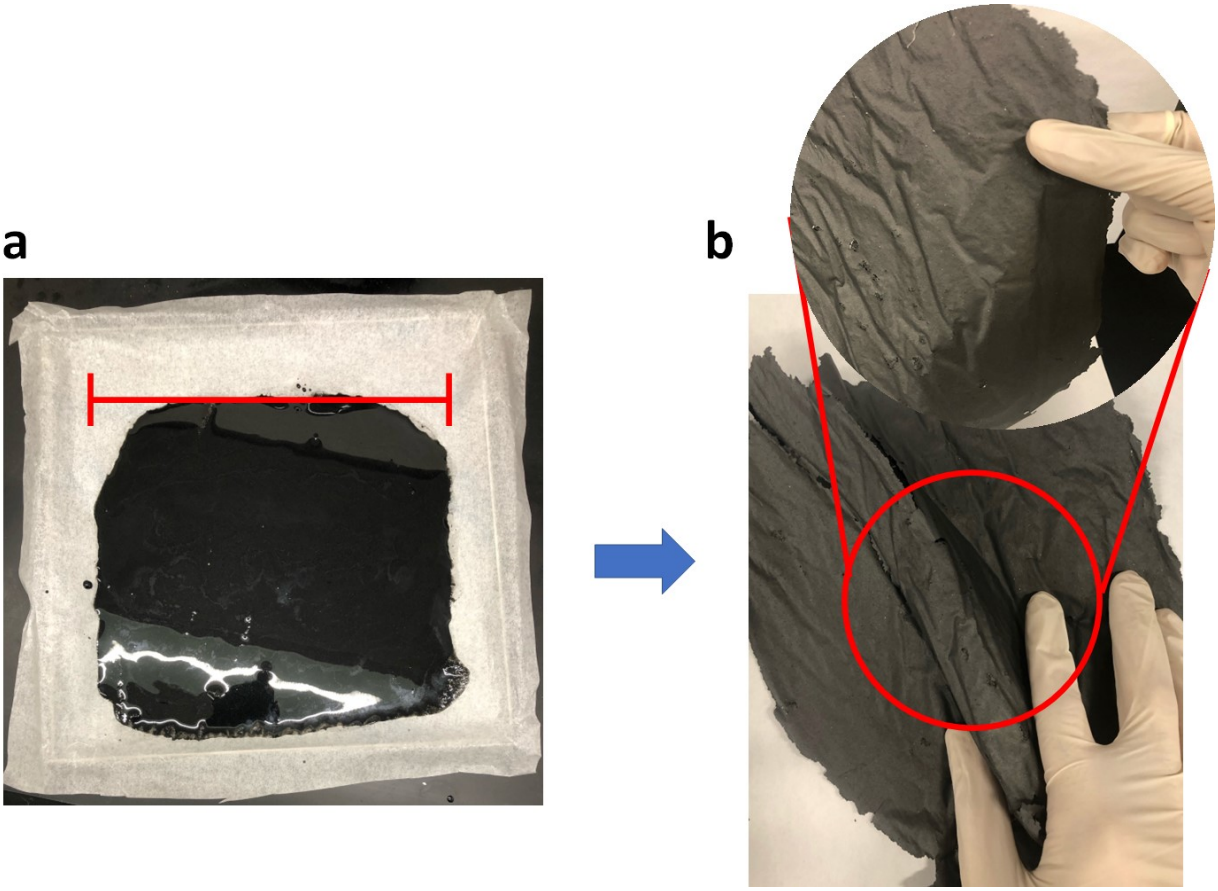

Figure S2: Cast-dried GO paper. (a) Casted GO suspension; (b) Dried GO paper. Red scale bar corresponds to 28 cm.

4. References

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