

Supporting Information

--for--

Green Approach for the Synthesis of Monolayer Reduced Graphene Oxide: One-Step Protocol with Simultaneous Iodination and Reduction

Pugalenthi Ramesh,^{*a} A. Alvin Kalicharan,^b Jayaraman Pitchaimani^{*c} and Ravindranadh Koutavarapu^{*d}

^a*Department of Chemistry, Vivekanandha College of Arts & Sciences for Women, (Autonomous), Tirchengode, Namakkal-637205, Tamil Nadu, India*

^b*Department of Chemistry, Panimalar Engineering College, Chennai-600123, Tamil Nadu, India*

^c*Department of Chemistry, Sri Sai Ram Institute of Technology, West Tambaram, Chennai-600044, Tamil Nadu, India*

^d*Department of Robotics Engineering, College of Mechanical and IT Engineering, Yeungnam University, Gyeongsan 38541, Republic of Korea*

**Correspondence: drramesh2608@gmail.com (P. Ramesh); pitchumani07@gmail.com (J. Pitchaimani); ravindra.physicist@gmail.com (R. Koutavarapu)*

1.1. Materials

Graphite (500 mechs) was purchased from Sigma-Aldrich. Concentrated H_2SO_4 (98.08%) and HCl (35%) were purchased from Merck Laboratory. NaIO_4 and m-CPBA were purchased from Sigma-Aldrich. In this study, we used all analytical-grade chemicals.

1.2. Characterization

The chemical properties, types of functional moieties presence, the number of layers and sizes, transparency, the thickness of the sheet, structural flaws, and thermal stability were all evaluated by XRD, TEM, SEM, XPS, AFM, FT-IR, Raman spectrum and TGA analysis in order to assess the prepared i-GO and i-RGO material. Structure-related data By using powder X-ray diffraction data from a Shimadzu XRD-6000 equipped with $\text{Cu-K}\alpha$ radiation ($\lambda = 1.5406 \text{ \AA}$) as the source and 40 kV and 30 mA as the applied voltage and current, respectively, the produced materials were analyzed. On a JOEL JEM-2100, SAED pictures and high-resolution transmission electron microscopy data were gathered to examine the morphology of i-GO material. 5 mg of i-GO in ethanol and 10 minutes of sonication, TEM material was developed. The dispersed solution was poured onto a Ni-coated grid and dried before the examination. Achromatic Al K radiation with a wavelength of 1486.6 eV was used as the radiation source for the XPS data on a Carl Zeiss X-ray photoemission spectroscopy apparatus. Using a TGA instrument, the thermal stability of several materials was measured in a nitrogen environment (NETZSCH, STA449F3, Jupiter, made in Germany). IR-Prestige-21-Shimadzu performed the testing for FT-IR spectra in the spectral region from 4500 to 400 cm^{-1} . The substance was prepared by mixing KBr with samples that had been formed into pellets. We used the NTMDT (NTEGRA Prima, Russia) semi-contact operating mode with Si (silicon) cantilevers (spring constant $\sim 0.3\text{-}6.6 \text{ N m}^{-1}$, resonance frequency range $\sim 97.0 \text{ kHz}$) to execute the atomic force microscopy technique and capture the image. Data from Raman spectra were collected using a Horiba-Jobin (Raman spectrometer) and a powerful 514 nm laser in an N_2 environment.

1.3 Experimental section

Synthesis of iodinated RGO through the one-step synthesis protocol

A novel one-step protocol was used to synthesize i-GO and i-RGO (Fig. 1). First, 1 g of graphite powder was mixed with 30 mL of concentrated H_2SO_4 through ultrasonication for 20 min, and the mixture was stirred for an additional 20 min at room temperature (RT). Subsequently, NaIO_4 (12 g) was added slowly and stirred for 24 h at RT. In stage two, m-CPBA was slowly added to the mixture and stirred for 8 h at RT. The mixture was then washed with distilled water several times to maintain a neutral pH to collect a pure product of i-GO. The produced i-GO and i-RGO were analyzed separately. A small amount was collected for analysis from stage one and washed with water several times to obtain i-GO. Subsequently, in stage two, a small amount was collected and washed with water several times to analyze i-RGO

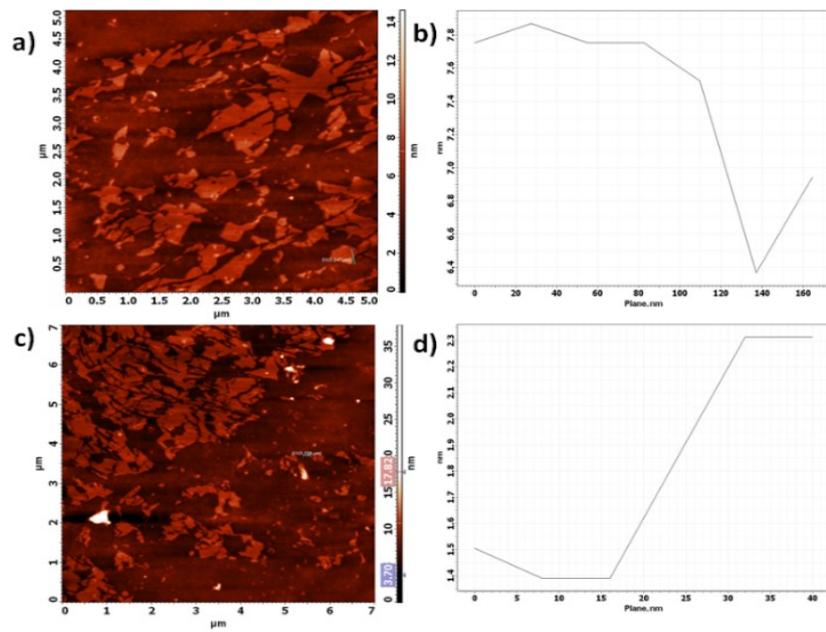


Fig. S1 AFM images for i-GO and i-RGO

The intercalation of C-I, oxygen functionalities, and the degree of reduction on the surface of graphite sheets were confirmed by Fourier-transform infrared (FT-IR) spectroscopy. The FT-IR spectra provide data on the type of functional moieties available in the i-GO and i-RGO materials. Figure S2 shows the FT-IR spectra of i-GO with several characteristic absorption peaks after the chemical oxidation of graphite. The peaks observed at 1012 and 1615 cm^{-1} are attributed to the (O-C-O) epoxy and (C=C) stretching vibrations of the sp^2 aromatic network. The peaks at 1729 and 3431 cm^{-1} correspond to secondary alcohol (-OH) and carbonyl (C=O) stretching vibrations, respectively. However, in the FT-IR spectra of the i-GO and i-RGO materials, a new intercalation peak is observed at 751 and 758 cm^{-1} owing to the stretching vibration band (C-I), which confirms that iodine is present on the surface of the graphite layers. After the chemical reduction of i-GO, there is a significant change in the intensity of the peaks corresponding to oxygen functional moieties. The intensities of the epoxy and secondary alcohol peaks decrease in the i-RGO material, such as those of the stretching vibration peaks at 1040 and 1725 cm^{-1} . Moreover, similar peaks are obtained at 3435 and 1622 cm^{-1} , ascribed to the stretching vibrations of the -COOH and C=C peak bands, respectively. The FT-IR results confirm that the removal of oxygen functionalities and the formation of iodine functional moieties on the graphite surface during reduction are successful.

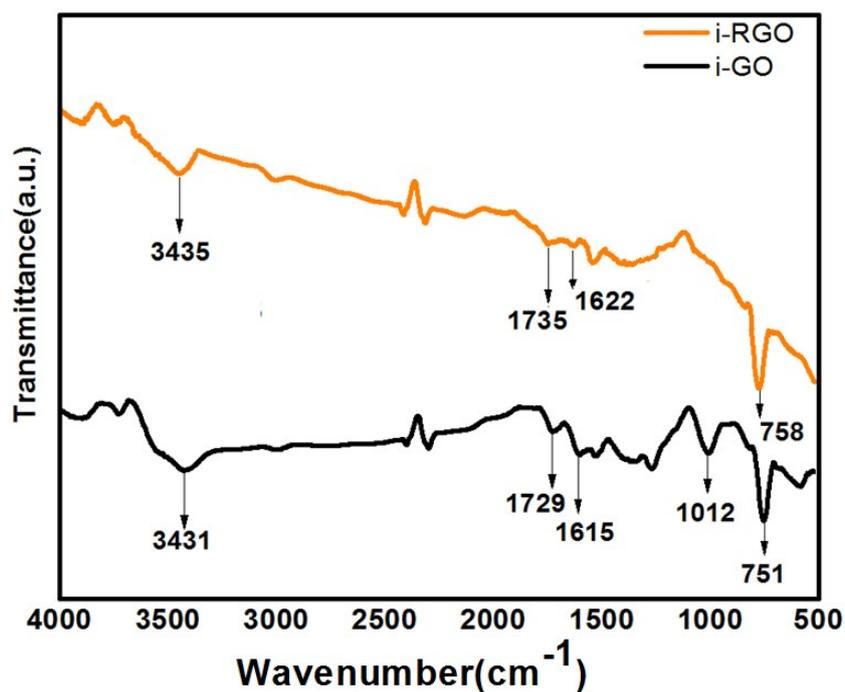


Fig. S2 FT-IR spectra of i-GO and i-RGO

Thermogravimetric analysis (TGA) was used to differentiate the thermal stabilities of i-RGO and i-GO. The TGA curves of i-GO and i-RGO show slight and similar weight losses between 50 and 230 °C. During this time, approximately 15–20% weight loss occurs because of the removal of the volatile species, trapped water molecules, and oxygen-containing functional groups (Fig. S3). Generally, modified GO shows a sudden major weight loss at temperatures above 230 °C owing to the decomposition of oxygen functional groups such as epoxide, –OH, and –COOH. However, i-GO shows a gradual weight loss above 250 °C because of the elimination of stable and iodine-type functional moieties. Furthermore, in i-RGO, slow degradation is observed above 250 °C and 30% weight loss is noted owing to the decomposition of the sp³-hybridized carbon network. The TGA data show that the iodine-intercalated surface of the carbon network (i-RGO) withstands higher temperatures than the i-GO material.

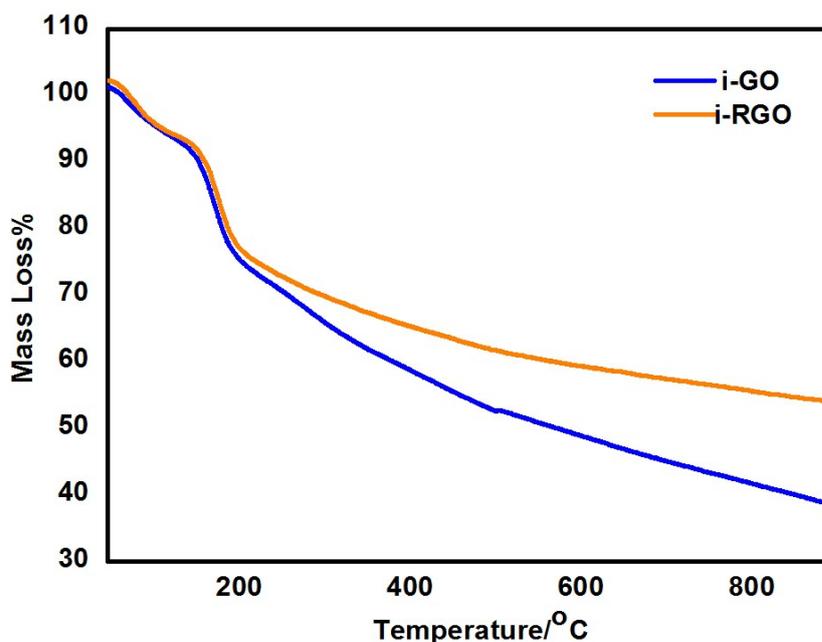


Fig. S3 TGA analysis of i-GO and i-RGO

Table: S1

High resolution XPS was shown to further examine the elemental composition of the surface of the GO and RGO, and the presence of iodine atoms is exhibited in Table 1.

Sample Name	Wt% of Carbon	Wt% of Oxygen	Wt% of Iodine
i-GO	63.2	28.7	8.1
i-RGO	81.4	10.6	8.0