

# Synthesis of Graphene Oxide from Hydrogenated Diamond Like Carbon and Protein Immobilization onto It: Characterization and Study of Practical Utility

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## Abstract

In the last few years, Graphene oxide material and biomolecules studies have increased. The various synthesis methods of graphene oxide are constantly pursued to improve and provide safer and more effective alternatives. Though the preparation of graphene oxide from Graphite powder or Graphite flake through Hummers method is one of the oldest techniques but still now it is one of the most suitable methods. Here, Graphene Oxide has been prepared from a tunable material Hydrogenated diamond like carbon (HDLC) which is an atomically smooth surface that can be deposited on high-surface area Silicon (100) wafer plate. The HDLC film was heated at a fixed temperature of 900°C for 30 min in high vacuum  $\sim 1 \times 10^{-6}$  torr and oxygenated at room temperature. A synthetic sequence is described involving Oxidation of annealed HDLC (A-HDLC). Raman measurements confirm the G and D peak by Oxidation of A-HDLC and FTIR confirms functional groups. Atomic force microscopy (AFM) images describe the surface of A-HDLC, Oxidized Graphene and BSA immobilized GO. This GO onto Silicon substrate offers many technical advantages than as oxidized graphene Synthesis from other Chemical methods.

## Keywords

Carbon, Diamond, Graphene Oxide, AFM, Raman, FTIR

## 1. Introduction

Preparation of Graphene Oxide and Immobilization of biomolecules onto the large smooth surface for biosensor applications has attracted significant attention in recent years around the Globe. Besides, immobilization of biomolecules [1] [2] [3] [4] onto surfaces of different material is essential in many fields of Chemical [5] [6], physical, biological sciences, including cell and molecular biology, medical diagnostics, analytical chemistry, tissue engineering, and bioprocess engineering. Since the discovery of the many advantageous properties of immobilized biomolecules, the challenges in this area have been to develop new support substrate materials with appropriate structures, surface morphology and functionality to widen the understanding of immobilized biomolecules. Recently, along with the development of nanostructured materials [7], a range of nanomaterials with different surface properties have been utilized as substrates for biomolecule immobilization [8]. In particular, oxidized graphene [9] has been demonstrated as a well-defined, ordered, and atomically smooth surface [9]. Graphene is comprised of a lattice of carbon atoms in a hexagonal array, creating an ordered structure against which morphology of immobilized biomolecules can be easily discerned. We have demonstrated here that the GO can be prepared from A-HDLC as large solid-state sheets to produce an ordered structure which can be used as a solid support substrate on which biomolecules can be immobilized, spontaneously adsorbed [10].

## 2. Abbreviations and Acronyms

Hydrogenated Diamond Like Carbon (HDLC), Graphene Oxide (GO), Oxidation of annealed HDLC (A-HDLC), Atomic force microscopy (AFM), Bovine Serum Albumin (BSA), BSA treated Graphene oxide sample (BGO), Transmission electron microscopy (TEM), Scanning electron microscopy (SEM).

## 3. Materials and Methods

### 3.1. Materials

Potassium Permanganate (extra pure AR, 99.5%, Sisco Research Laboratories Pvt. Ltd.), Potassium nitrate, (ACS reagent,  $\geq 99.0\%$ , Sigma Aldrich), Sodium Phosphate Dibasic Anhydrous, (99.5%, Sisco Research Laboratories Pvt. Ltd.), Bovine Serum Albumin (BSA) (96.96%, Sisco Research Laboratories Pvt. Ltd.). Ultra-pure water using Milli-Q system of Millipore Co. was used throughout the experiment.

### 3.2. Preparation Graphene Oxide from HDLC

We already have demonstrated earlier [11] a straightforward synthesis of HDLC film surfaces. The HDLC film was heated to get Graphene-like and graphite-like structure [12] (A-HDLC) at a fixed temperature of  $1000^{\circ}\text{C}$  for 30 min in high vacuum  $\sim 1 \times 10^{-6}$  torr and cooled to room temperature after heating. Graphene oxide (GO) was prepared from A-HDLC according to the

modified Hummers-Offeman method [13]. In brief, about 10mm diameter A-HDLC sample was taken in 50 mL beaker and cold concentrated sulphuric acid added to it (1 mL, 98 wt%, dry ice bath) and potassium permanganate ( $\text{KMnO}_4$ , 20 mg) gradually added with continuous vigorous stirring and cooling to prevent the temperature from exceeding 25°C. The dry ice bath was removed and replaced by a water bath and the mixture heated to 35°C for half an hour with gas release under continuous stirring, followed by slow addition of deionized water (2 mL), which produced a rapid increase in solution temperature up to a maximum of 90°C. The reaction was maintained for 10 min to increase the oxidation degree of the graphene oxide product.

### 3.3. Immobilization of Protein onto the Surface of Graphene Oxide

0.2 M Phosphate buffers of pH 7.5 were prepared at room temperature. Graphene Oxide sample, cleaned by isopropyl alcohol and rinsed in ultra-pure water, dried with nitrogen and was immersed in BSA solution, prepared by dissolving 25 mg of BSA in 25 mL of 0.2 M Phosphate buffer, for 2 hours at room temperature under slow stirring was condition using a magnetic stirrer. The BGO was rinsed with ultra-pure water and dried with nitrogen.

### 3.4. Characterization of Samples

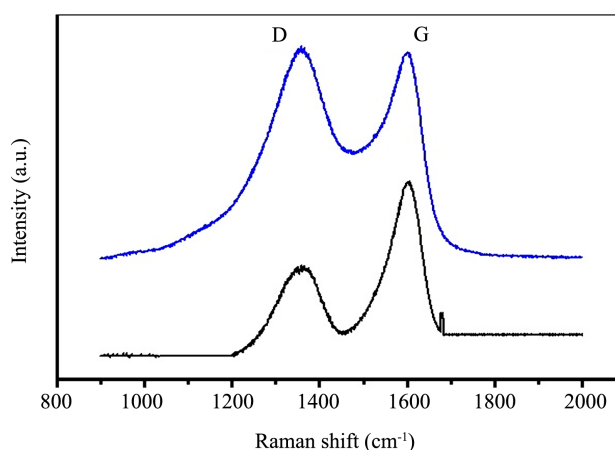
The cross sectional view and the morphology of the surface of the GO film surface have been analyzed by TEM, AFM and SEM respectively. Raman spectrum of the GO sample was obtained by the confocal Micro Raman spectrometer (LabRAM HR Vis, Horiba Jobin Yvon SAS France) which includes an 800 mm focal length Czerny-Turner type spectrograph equipped with mirrors (reflective optics) having high spectral resolution of 0.1  $\text{cm}^{-1}$ /pixel at 488 nm with an 1800 gr/mm grating. FTIR spectrum of the sample was obtained by Infrared Fourier Vacuum Spectrometer Vertex 70v (Bruker Optik GmbH Germany) with fixed 30° angle reflection with ATR crystal (Zn-Se) for solid GO sheet and sample. KBr beam splitter, DLaTGS detector, MIR source, Rock Solid Interferometer system, spectral resolution ( $>0.4 \text{ cm}^{-1}$ ) and contamination free vacuum ( $<0.2 \text{ mbar}$ ) are the key technical features of Vertex 70 V. AFM topography image of the sample was obtained in non-contact mode by Multimode Scanning Probe Microscope (Agilent AFM 5500 series, USA) having multipurpose small scanner with low coherence laser (1 mW power, 670 nm wavelength, length of coherence ( $<50 \text{ mm}$ ), scan range: XY: 0 - 10 mm; Z: 0 - 2 mm, noise level: XY  $< 0.1 \text{ nm RMS}$ , Z  $< 0.02 \text{ nm RMS}$ .

## 4. Results and Discussion

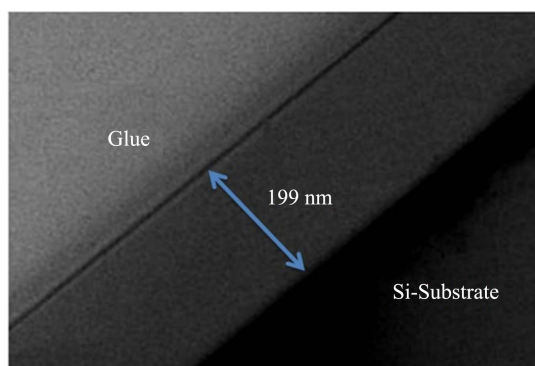
Raman spectroscopy is a fingerprint and non-destructive technique to characterize graphitic materials, in particular to determine the defects and the ordered and disordered structures of graphene. **Figure 1** shows the Raman spectra of the A-HDLC and GO sheets prepared from HDLC. The Raman spectra of GO sheet display two prominent peaks at 1358 and 1598  $\text{cm}^{-1}$ , corresponding to the

well-documented D- and G-band. The G line represents the in-plane bond stretching motion of the pairs of C  $sp^2$  atoms (the  $E_{2g}$  phonons); while the D line corresponds to breathing modes of rings or j-point phonons of  $A_{1g}$  symmetry [14]. Usually, the graphene-like sheets synthesized by annealing of HDLC approach show two clear D and G band in the Raman spectrum with the intensity ratio of  $I_D/I_G \approx 1$  due to the defects and partially disordered crystal structure of graphene-like sheets. It is noted here that for the GO, the intensity of the G band is significantly higher than that of the D band, suggesting that the prepared graphene Oxide has low defect content [15] [16]. **Figure 2** shows the thickness of the HDLC thin film.

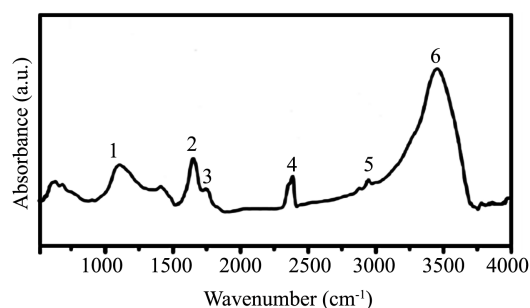
The FT-IR analysis [17] of GO in **Figure 3** shows the existence of H-bonded OH stretch at  $3415\text{ cm}^{-1}$ , C-H stretch at  $2915\text{ cm}^{-1}$ , C=O in carboxylic acid group at  $1725\text{ cm}^{-1}$ , C=C in conjugated ketones at  $1628\text{ cm}^{-1}$  and primary alcohol C-O stretch at  $1086\text{ cm}^{-1}$ . The presence of oxygen-containing groups on GO is observed while Graphene-like/Graphite-like spectra have no significant peak for these groups [18] [19]. FTIR analysis [20] [21] [22] in **Figure 3** and



**Figure 1.** Raman spectra of the A-HDLC and GO sheets prepared from HDLC. (Black = GO, Blue = A-HDLC; Excitation wavelength 488 nm, laser power 7.5 mW, grating 1800 gr/mm, objective 100 $\times$ , aperture 100  $\mu\text{m}$ ).



**Figure 2.** Typical TEM cross sectional view of the GO film measured with a Philips CM200 kV TEM machine operated at 200 kV with LaB6 filament; the sample of the cross section was prepared by the TEM sample preparation equipments of Gatan, USA.



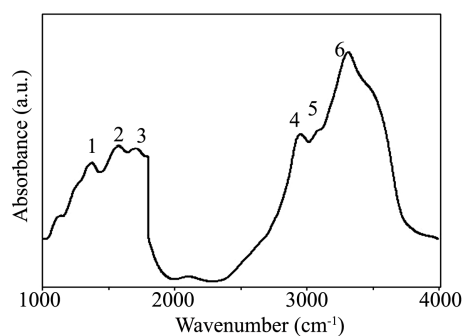
**Figure 3.** Typical FTIR Spectra of GO sheet sample surface.

**Figure 4** revealed that, after immobilization BSA onto GO, the peak intensity of GO at  $1725\text{ cm}^{-1}$  (carboxylic acid),  $1628\text{ cm}^{-1}$  (primary alcohol) and disappeared implying the reaction of such groups in GO. Characteristic peaks [19] [20] [21] [22] of the functional groups present in the GO and BGO samples, as shown in **Figure 3** and **Figure 4**, are assigned, and are given in **Table 1(a)** and **Table 1(b)**, respectively. **Figure 4** shows the immobilization of BSA, where the protein is immobilization on GO based on physical immobilization via  $\pi$ - $\pi$  stacking and other molecular interactions. The physical immobilization onto GO occurs via different types of interactions, such as Van der Waals forces, electrostatic or hydrophobic  $\pi$ - $\pi$  stacking interactions and hydrogen bonds between the oxygen functional groups of GO and nitrogen/oxygen groups of protein and chemical interaction *i.e.* covalent bond may among C=O of GO and  $\text{-NH}_2$  of Protein [23] [24].

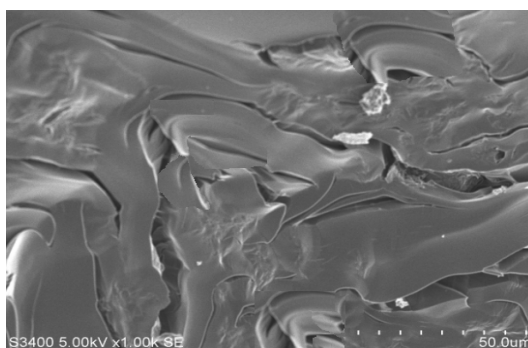
**Figure 5** and **Figure 6** show the SEM images of Graphene-like A-HDLC and GO sheets composites, respectively. The sample shows plate-like forms without any amorphous or other kinds of crystallized phase particles. Morphology of GO [25] [26] is observed as flaky texture reflecting its layered microstructure as shown in **Figure 6**. The larger interspaces of the layer and the thinner layer edges of GO can be clearly seen in **Figure 6**.

AFM was employed to characterize the degree of exfoliation of graphene-like sheets. As shown in **Figure 7**, a flat graphene-like sheet was selected for further investigation using the three-dimensional view.

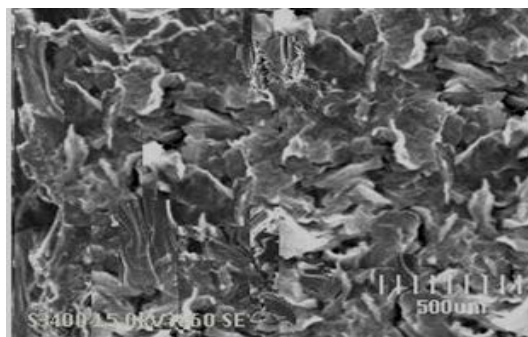
From the profile of the atomic force microscopy (AFM) image, the roughness of the GO film in the GO device was estimated to be approximately 10 nm. We performed AFM to investigate the morphology of the GO sheets (**Figure 8**). The well-ordered surface improves resolution of immobilized protein by AFM. The root mean square roughness was 0.1 nm. It seemed that whole area was covered with GO sheets. We have shown that GO can serve as a substratum for immobilization of proteins. Since BSA protein is a complex molecule having a large variety of functional groups, at this stage it is very difficult to identify the exact route of reaction pathways for GO sheets with BSA protein. There were two reactive groups namely C=O and -OH groups in GO sheets. The more possibility for the reaction of BSA protein with GO sheets is electrostatic interaction/H-bonding/Vander waal's force of attraction and covalent bonding. The



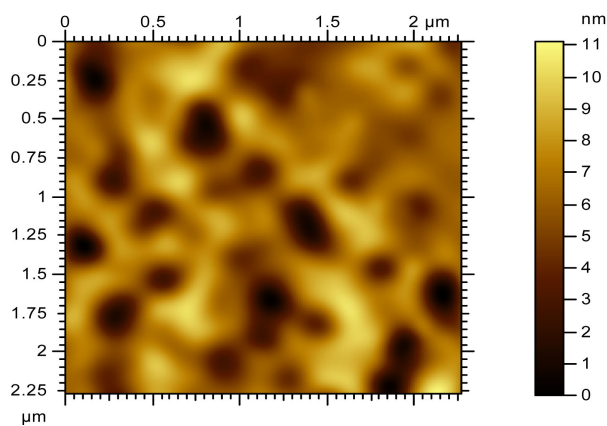
**Figure 4.** Typical FTIR Spectra of BGO sheet sample surface.



**Figure 5.** SEM image of Graphite-like sheet (A-HDLC) and Graphene-like sheet.

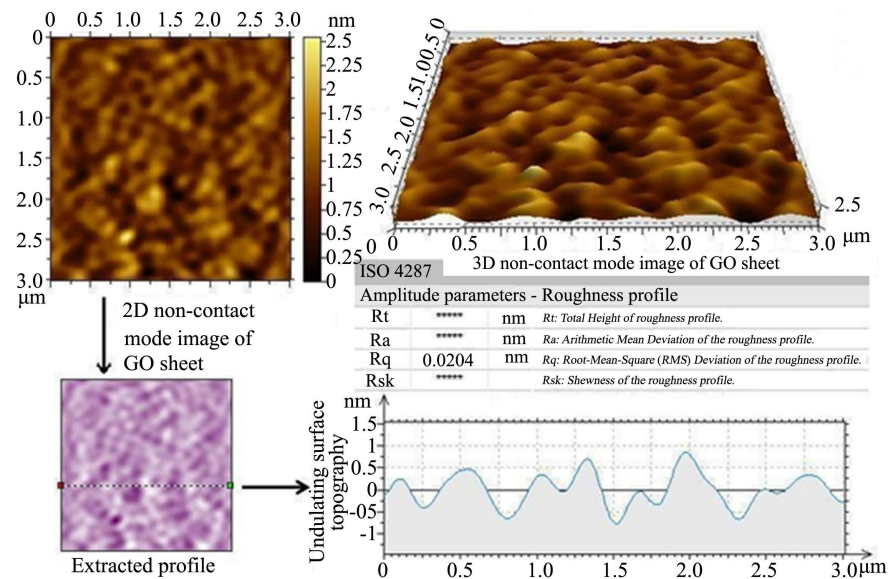


**Figure 6.** SEM image of GO sheets.



**Figure 7.** Non-contact mode AFM topography images of A-HDLC surface: scan size 2.25  $\mu\text{m} \times 2.25 \mu\text{m}$ .





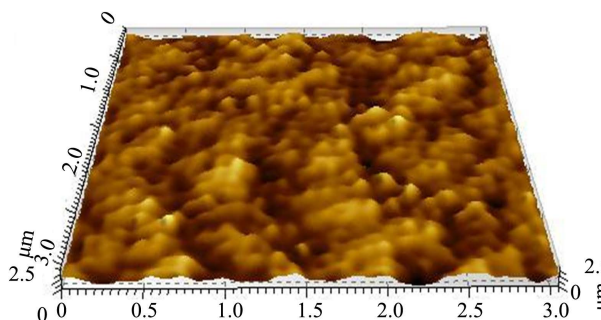
**Figure 8.** Non-contact mode 2D and 3D AFM topography images of GO sheets: scan size  $3 \mu\text{m} \times 3 \mu\text{m}$ .

**Table 1.** (a) Tentative assignments of peaks in the FTIR spectra of GO sheet; (b) Tentative assignments of peaks in the FTIR spectra of BGO.

(a)		
Peak No.	Wave number ( $\text{cm}^{-1}$ )	Tentative assignment
1	1086	C-O stretching
2	1628	C=C stretching
3	1725	C=O
4	2362	CO <sub>2</sub> stretching
5	2825 - 3050	Aliphatic and Aromatic C-H stretching
6	3100 - 3600	Broad band for O-H stretching and H-bonding

(b)		
Peak No.	Wave number ( $\text{cm}^{-1}$ )	Tentative assignment
1	1368	Amide-III N-H in solid state also for water absorption and H-bonding
2	1575	RCO <sub>2</sub> <sup>-</sup> , N-H bending for secondary Amide-II in solid state
3	1688	Amide-I
4 and 5	2900 - 3100	Aliphatic and Aromatic C-H stretching
6	3100 - 3600	Broad band for O-H stretching N-H stretching and H-bonding

AFM image (**Figure 9**) shows the topography of the surface of BGO sample. Its roughness value (0.5 nm) is higher, by an order of magnitude, than that of the GO sheets surface. The immobilized proteins are globular in nature and they are aggregated as shown in the image **Figure 9**.



**Figure 9.** Non-contact mode 3D AFM topography images of BGO sheets: scan size  $3\ \mu\text{m} \times 3\ \mu\text{m}$ .

## 5. Conclusion

We have demonstrated that GO films comprised of an ordered structure of carbon atoms can be prepared on large-area-substrate surfaces. We further demonstrated that GO is an atomically smooth substratum useful for protein immobilization. Good AFM imaging of BSA protein onto GO Sheet and retention of the conformation of the immobilized protein are due to the atomically smooth surface with roughness  $< 0.10\ \text{nm}$ . The ability to coat macroscopic surfaces presents GO Sheet as a potentially useful coating for biomedical devices, Spontaneous protein adsorption on Graphene Oxide nanosheets allowing efficient intracellular vaccine protein delivery [27]. HDLC materials have been shown to exhibit biocompatible surface properties [8] [9] [10]. It can be anticipated that GO sheet will also be biocompatible [9] [28] and may offer improved properties by harvesting [29] related to the highly ordered structure.

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## Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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