An exploration on anti-corrosion performance of electrochemically synthesized graphene coating on commercially pure copper

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Abstract

Graphene, a path-breaking discovery of the last decade has opened up numerous research avenues with its superior properties. Endowed with qualitative physical, electrical and optical characteristics, this "wonder material" has set the tone for many futuristic applications. The present work focuses on electrochemical exfoliation of graphite in sodium sulphate (Na_2SO_4) aqueous solution of 4 different molar concentrations to synthesize graphene. Graphite rods were subjected to pre-cathodic exfoliation in 1M H_2SO_4 solution after which anodic exfoliation was carried out in the aqueous solution at a potential of 10 V. The supernatant is disposed, the residue is washed thoroughly with acetone, and double distilled water. Subsequently, the collected material is dried and used for further characterizations. XRD and EDS analysis substantiate the presence of graphene nano-sheets along with traces of oxygen. Raman studies pointed out that the graphene synthesized at lower molar concentration had less defects owing to slow and smooth exfoliation. UV studies point out to electronic transitions in C=C bonds present in the graphene sheets. Graphene with few layered structures and stacked onto one another, were obtained for all concentrations in SEM. Transparent, curled layers are depicted through TEM morphologies. FTIR shows minimum oxygenation occurring at lower molarities. Graphene nano-sheets are coated onto copper substrates via electrophoretic deposition at a constant voltage of 5 V and different timings. The coated samples are then made to undergo corrosion tests in 3.5 wt% NaCl solution. Graphene coatings prove effective in combating corrosion and it is worth noting that bi-layer coatings prove to be more efficient in comparison to single or multi-layer coatings.

Keywords: Electrochemical exfoliation, Graphene nano-particles, Electrophoretic deposition, Corrosion

1. Introduction

Graphene is a thin honeycomb structured, atomic level arrangement of carbon atoms having sp^2-hybridization that has sparked interest in the field of materials. Owing to its extraordinary electronic, thermal and mechanical properties, researchers are aiming for a scientific leap. Since its discovery in 2004, it has sparkled the attention of many researchers with focus being the development of a fabrication route to obtain defect-free, quantitative and superior quality graphene. Graphene has set a huge trend with the increase in demand for nanotechnology; however, the production of graphene on an industrial scale is a challenge. Graphene synthesis techniques like Hummer's method, chemical reduction are quantity oriented and they comprise of defects. CVD technique produces quality graphene; however, the quantity and costs stand compromised. Electrochemical exfoliation stands out among these difficulties as it promises high-grade graphene with a quantitative yield. The technique is advantageous owing its easiness, high efficacy, and low oxidizing phenomena [1–3]. Authors have documented the effect of ionic forms from various electrolytes on graphite/graphite modified electrodes. [4–8]. Simple protic solvents were employed initially for successful synthesis of GNPs (graphene nanoparticles) [1,9,10]. Other protonic acids such as perchloric, phosphoric, and oxalic acids also show promising electrochemical anionic intercalation and exfoliation leading single to few-layer graphene nanosheets formations [11,12]. Sahoo et al. [13–15] have been succeeded in the scalable synthesis of FLGPs

in cost-effective manner from various protic solvents.

Corrosion is a major aspect, which influences the longevity of a metal, thus its performance. Early failures due to corrosion have been quite impactful on industries and economies. Necessity demands better protection schemes, which can ne also cost effective. Graphene and graphene oxide (GO) have proven to be effective as an anti-corrosion barrier against various corrosive medium when provided as a coating on the substrates. [16,17]. The reason behind choosing graphene as a corrosion reducing material is due to its very low reactivity and high chemical inertness and adhesive nature towards any metal. Coating thickness, presence of multiple layers was notable in reducing corrosion [18–25]. R K Singh Raman et al. [26] coated graphene on copper and highlighted the protective ability of graphene. Electrochemical coating of the synthesized graphene onto thin copper foils is achieved in the present work and the corrosion behaviour of the coated sample is observed in LSV test. XRD, Raman, FTIR, SEM, FESEM have been carried out to characterize graphene.

2. Methodology

2.1 Electrochemical exfoliation

Electrochemical exfoliation is a low-cost technology for large-scale production of the high quality graphene nanoparticles (GNPs). The technique as shown in Figure 1 follows the intercalation of cations or anions from the electrolyte to the graphite electrode with a suitably applied bias. As a result, of ionic impacts as well as the electrochemical reactions at the electrode surface, exfoliation occurs. All chemicals, materials and solvents used were of analytical grade and have been used without further purification. The water solvent used for the experiments was collected from an installed Borosil, Model 3362 double distillation unit. High quality, isostatic processed super fine grain pyrolytic graphite sheet of thickness 3 mm (Asbury Graphite Mills, IPG15) was employed as both the working as well as counter electrodes in a voltage regulated DC bias system. A freshly cleaved graphite sheet (1.0 cm × 2 cm × 0.3 cm) was used as working electrode (WE) by blanketing the remaining portion by an adhesive tape. The counter electrode was taken the same one in none blanketed conditions. The separation between the two electrodes was maintained 1 cm gap facing vertically to each other. A voltage regulated, ammeter conjugated DC bias system of Aplab make, Model No. 7103 was used for the electrochemical synthesis. The electrolytes used were 0.5 M, 1.0 M, 1.5 M and 2.0 M aqueous sodium sulphate solutions. 1.0 M sulphuric acid solution was used for pre-cathodic exfoliation. Initially, a high negative DC bias of 10 V for 2 minutes was applied to the WE (cathodic treatment) to serve a dual purpose, i.e., to make the availability of surface as well as internal lattice pores of graphite electrode for efficient intercalation. Then the cathodically treated WE were swapped to the anodic terminal for the anionic intercalation and subsequent exfoliation with ramping applied bias from 0-10 V. Aftermath, the intercalation and exfoliation at a constant voltage of 10 V was carried out till the synthesis of the desired amount of exfoliated graphene nanosheets have been obtained.

2.2 Characterization of exfoliated few layer graphene nanosheets

X-Ray diffraction analysis has been done for few layer graphene nanoparticles (FLGPs) in the range of 2θ from 5° to 70° at the scan rate 10°/min with the time step 0.02 at CuKα radiation (λ= 0.154 nm) using Rigaku Model: Ultima IV. X-ray diffraction technique is based on Bragg's law, which is used to determine crystal structure, phases and orientation of planes. It can also provide information about unit cell dimension of crystalline material. Surface morphology analysis has been done by FESEM Model: Nova NanoSEM 450. Field emission scanning electron microscope is an analytical technique used to study the microstructure of material and their electronic properties. Jeol 6048 LV SEM and INCA software was used for elemental analysis or chemical characterization of the samples. TEM of FEI, Tecnai G2 TF30-ST was used to look at the morphology of FLGPs. FTIR Perkin Elmer IRL 1600300 wss used for detecting asymmetric molecular stretching, vibration and rotation of chemical bonds of the sample. Raman spectroscopy can provide information about crystallite size, defect, number of graphene layers etc. PL Micro Raman Spectrometer (WITec) was used for the same with a 532 nm excitation laser. In UV-Visible spectroscopy, the electromagnetic radiation ranging from 200 to 900 nm wavelengths have been incident on the sample. When these high monochromatic energies are incident onto the solid/liquid sample, it will absorb certain radiation, and the rest will be transmitted. The absorption of incident radiation by the sample has been recorded and reproduced in the form of the absorption spectrum. From this spectrum, the nature of the sample, functional groups, electronic transitions, and optical band gap can be known. Malvern Nano ZS model was used for particle size analysis, which was carried

out by dynamic study of speckle patterns of the particles.

Fig. 1. Different stages of exfoliation

2.3 Electrophoretic deposition of graphene nanosheets onto copper strips

Electrophoretic deposition of FLGPs was carried out onto copper substrates with working area of 1 cm^2. In the EPD setup, copper substrate is designated as the anode whereas graphite rod is designated as cathode. The working distance between working and counter electrode was 1 cm and connected to a D.C. voltage using system (Aplab, Model No.7103). The solution for EPD was prepared by sonicating 2 g of graphene nanosheets in 1 litre of double distilled water. The stoichiometry of 2 mg/ml was maintained for the solutions. The coating procedure iterations have been presented in Table 1. The coated samples were dried at room temperature.

Table 1. Iterations for obtaining coatings

Iterations	Voltage	Time	Coating Layer
Iteration 1	5 V	10 mins	1 layer
Iteration 2	5 V	10 mins	2 layers
Iteration 3	5 V	10 mins	3 layers
Iteration 4	5 V	20 mins	1 layer
Iteration 5	5 V	20 mins	2 layers
Iteration 6	5 V	20 mins	3 layers
Iteration 7	5 V	30 mins	1 layer
Iteration 8	5 V	30 mins	2 layers
Iteration 9	5 V	30 mins	3 layers

2.4 Corrosion studies

Linear sweep voltametric tests were carried out for the graphene coated samples in 3.5 wt% NaCl solution. The corrosion test was conducted using a three-electrode Corrtest Electrochemical Workstation. The graphene coated samples were placed as working electrodes, platinum rod was utilized as counter electrode and the reference electrode employed was saturated calomel electrode. An OCP of 90 minutes was applied. The graphene coated samples were polarized from -1 V to +1 V at a scan rate of 5 mV/s.
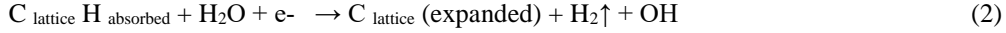
3. Results and Discussion

### 3.1 Exfoliation of graphene and characterization analysis

The production of FLGPs during exfoliation followed by two reactions:

Cathodic reaction: In this reaction, graphite sheet connected at cathode was given a voltage of 10 V for 2 min. The graphite crystal absorbed H$^+$ ions from electrolyte and released vigorously hydrogen gas at cathode. Therefore, surface

impurity of graphite lattice removed and produced more intercalated surface.

$$C_{lattice} + H^+ \rightarrow C_{lattice} H_{absorbed} \tag{1}$$
$$C_{lattice} H_{absorbed} + H_2O + e- \rightarrow C_{lattice} \text{ (expanded)} + H_2\uparrow + OH \tag{2}$$

Anodic reaction: In this reaction, graphite electrode connected at anode and applied voltage 0V to 10V with voltage step of 0.5 V increasing with time in an aqueous different solution. At low voltage range small bubbles and gases formed. The pretreated efficient intercalated electrode surface produced oxidation and carboxylation of exfoliated graphite compound.

$$C_{lattice} \text{ (expanded)} + Anions + H_2O \rightarrow C_{lattice} \text{ (exfoliated) } Anions + CO_2 + O_2 \tag{3}$$
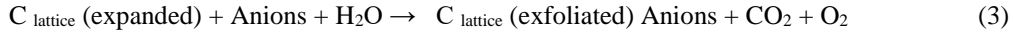
Due to electrochemical reaction at anode CO_2 and O_2 gases released and multilayer exfoliated graphite flakes settle down at the bottom.

XRD analysis was carried out in order to study the structural and any change in the inter-planar expansion of pyrolytic graphite (PGr) sheet and the exfoliated FLGPs. Sharp basal plane peaks (002) corresponding to 26.30° are visible in the XRD pattern depicted in figure 2 (a). The (002) plane corresponds to the basal plane of graphene with d spacing of 0.338nm. The slight broadening of peaks maybe attributed to a decrease in crystallite size.

The average particle size of the collected FLGPs have been measured by dynamic light scattering (DLS) technique as shown in figure 2(b). We observe a non-linear behavior as the size of anionic intercalate (SO_4^{2-}) being larger than the C-C off plane distance in PGr, the exfoliation process is majorly initiated on the defect sites of the graphite surface.

Surface functionalization is a binding phenomenon that occurs with any chemical synthesis of graphene nanoparticles. Electrochemically exfoliated FLGPs are endowed with various oxygen functional groups with the carbon atoms that result out of the electrolyte when applied with high potential. Exfoliation process also leads to various carboxylation and hydroxylation functional group formation. Figure 2(c) depicts mild functionalization observed in the case of synthesized FLGPs. Stretching vibrations of C-H functional groups mostly observed at the edges of the graphene sheets are observed in the range of 2800 – 3000 cm^{-1}. Vibration arising from C=C bond at 1575 cm^{-1} confirm the presence of graphene. Also, oxidation of graphene sheets corresponding to C-O stretching vibrations is observed from 1108 cm^{-1} peaks.

In figure 2(d), most of characteristic bands for the FLGPs were observed at ~1356, ~1587 and ~2705 cm^{-1} corresponding to D, D and 2D bands respectively. The D band is due to the presence of defects and disorders in sp^2 network, defects at the edges, endowment of functional groups etc. The G band represents the vibration mode of sp^2 hybridized carbon atoms present in the graphene sheet. The 2D band is a second order overtone of D band, due to resonant scattering of two phonons. This band is useful in determination of layers in graphene structure [27]. There is a noticeable increase in the intensity of D band, which suggests an increase in defects or functionalization with increase of intercalate concentration. The defect density in the carbon sp^2 structure can be quantified by I_D/I_G. Due to ample amount of lattice defects and available pores in the PGr sheet, the I_D/I_G ratio was found to be 0.62, whereas after electrochemical exfoliation the defect density increased to 0.774, 0.819, 0.994 and 1.015 for FLGPs synthesized form 0.5, 1.0, 1.5, 2.0 M solvents. This linear increasing order of disorderliness in the FLGPs may be attributed to severe intercalation with increasing intercalate concentration. The observed broadness and lower intensity of the 2D band confirms the formation of few layered graphene sheets.

The UV-visible spectra of synthesized FLGPs has been analyzed as presented in figure 2(e). Absorbance peaks are noted in the range of ~ (250 – 280) nm. The peaks are attributed to π-π^* electronic transitions of aromatic C=C bond present in sp^2 network structure in the FLGPs. A broad carried over shoulder peak is seen at around 350 -370 nm, which is mainly due to the n-π^* transitions of carbonyl group (C=O) present in sp^3 region in the hexagonal network of FLGPs structure.
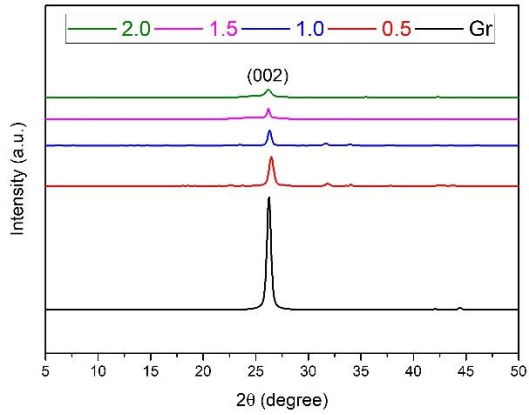
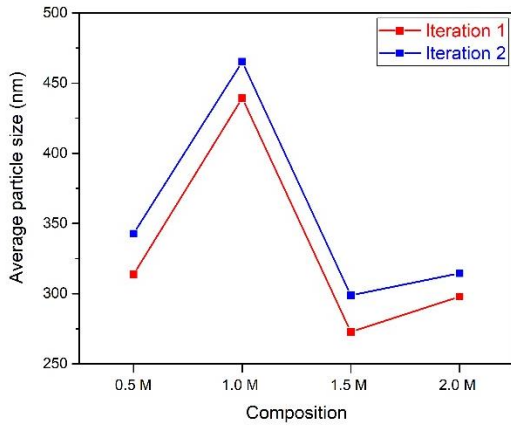Fig. 2(a). XRD Pattern of Na₂SO₄ synthesized FLGPs
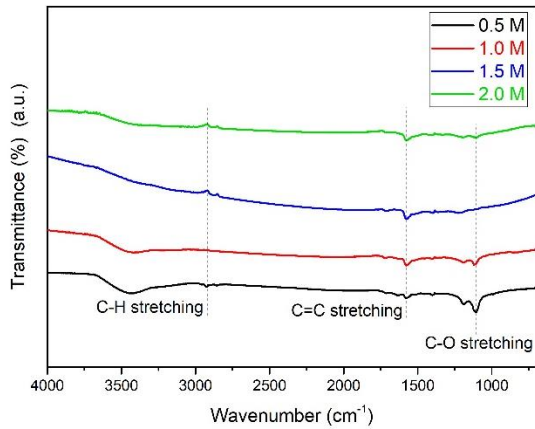

Fig. 2(b). Particle size of synthesized FLGPs

Fig. 2(d). RAMAN spectra of synthesized FLGPs
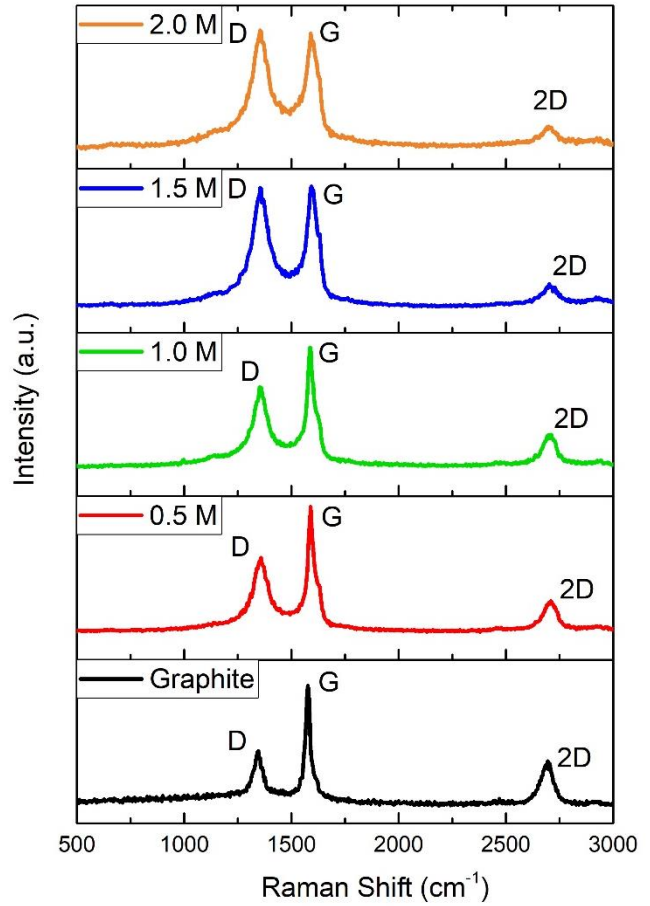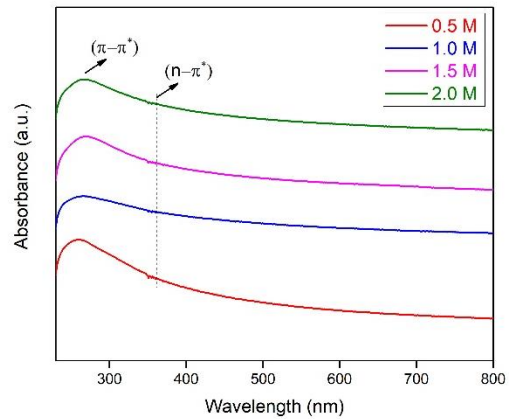

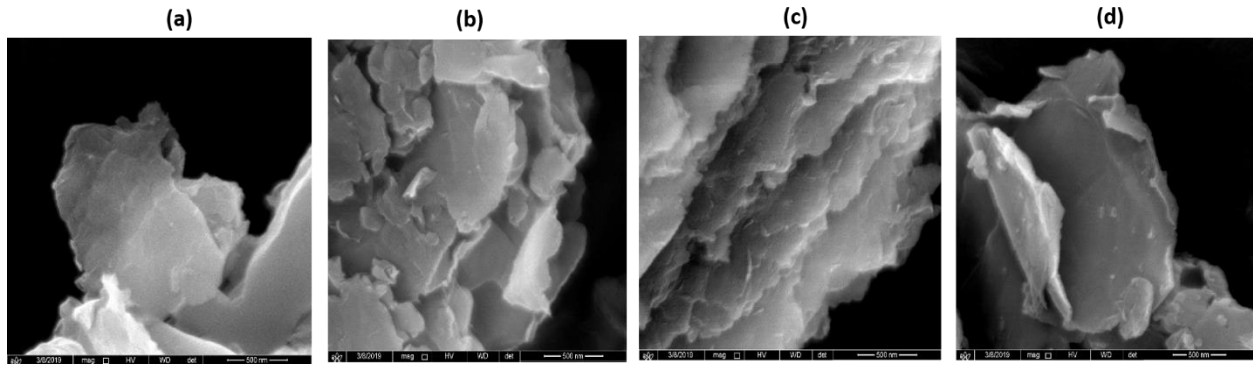Fig. 2(c). FTIR spectra of FLGPs

Fig. 2(e). UV spectra of FLGPs

Fig. 3. SEM micrographs of FLGPs (a) 0.5 M, (b) 1.0 M, (c) 1.5 M, (d) 2.0 M

Figure 3 shows the SEM images of electrochemically exfoliated graphene samples. A stacked layered structure at the edges is visible for all the samples. TEM images in figure 4 show partial transparency indicating the few layers of graphene sheets stacked together as well as the crumpling of the edges. With an increase in intercalation, FLGPs are formed with less number of layers.
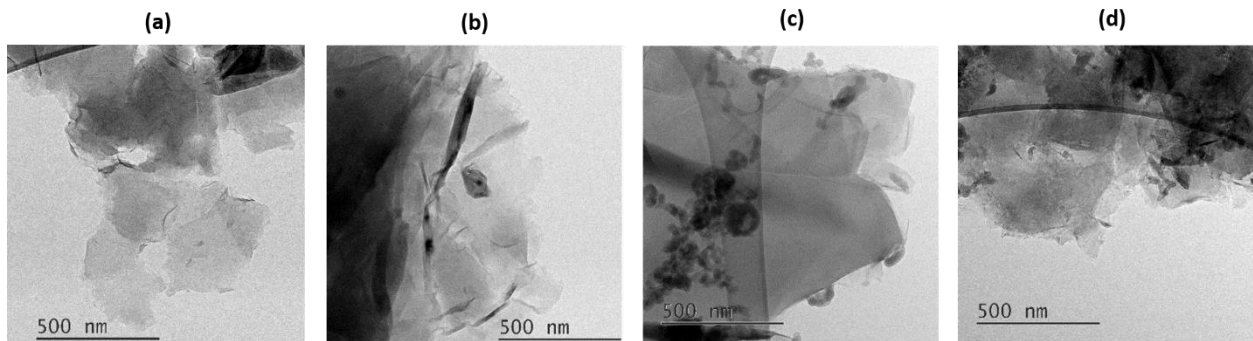

Fig. 4. TEM micrographs of FLGPs (a) 0.5 M, (b) 1.0 M, (c) 1.5 M, (d) 2.0 M

3.2 Characterization of Graphene Coated Copper and its Corrosion analysis

Figure 5 presents the elemental mapping of the graphene coating on copper substrate. From the area of observation, it can be noted that the top part clearly points out a significant amount of graphene (around 51 %) whereas the bottom part indicates the presence of copper (around 39%). A small amount of oxygen (around 9%) is also detected. Figure 6 shows the optical micrographs of graphene coated samples before and after corrosion. We can there is no effective changes, thereby suggesting that the coating has acted a decent prevention against corrosion.
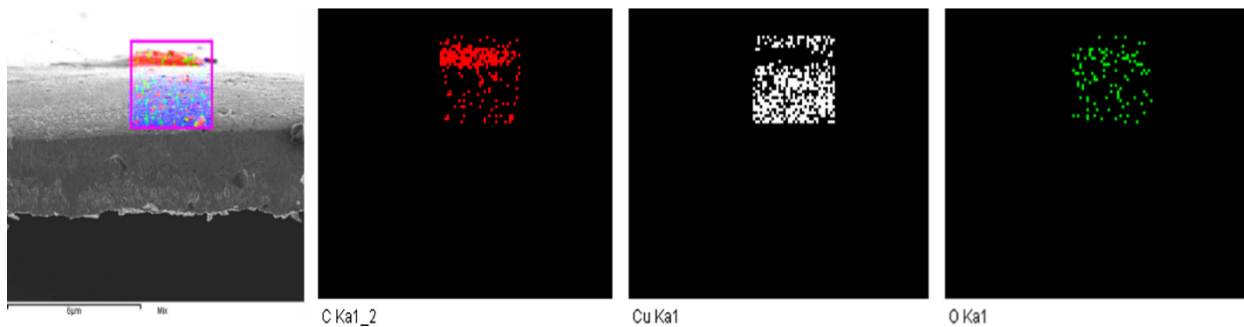

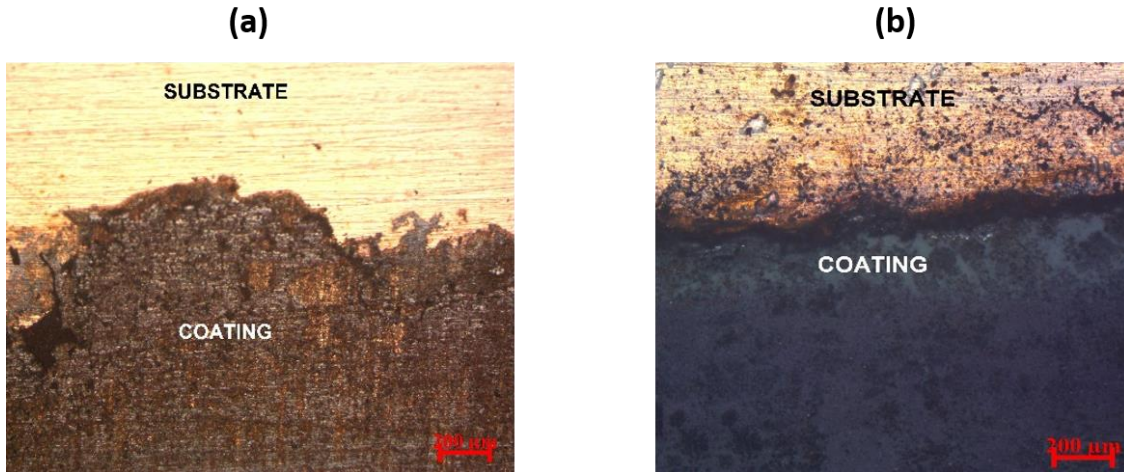Fig. 5. Elemental analysis of the coating

Fig. 6. Graphene coated copper (a) before corrosion and (b) after corrosion

The efficiency of graphene as an anti-corrosion barrier has been detailed out in the figure 7 and table 2. It is observed that corrosion performance for bi-layers was better than tri-layered coatings. This may be due to high defect density and accelerated corrosion rate in the latter case. Defects on graphene induced the permeation of corrosive medium and also promoted the lateral diffusion of chloride ions.

Table 2 Corrosion rates of blank copper and different coating iterations

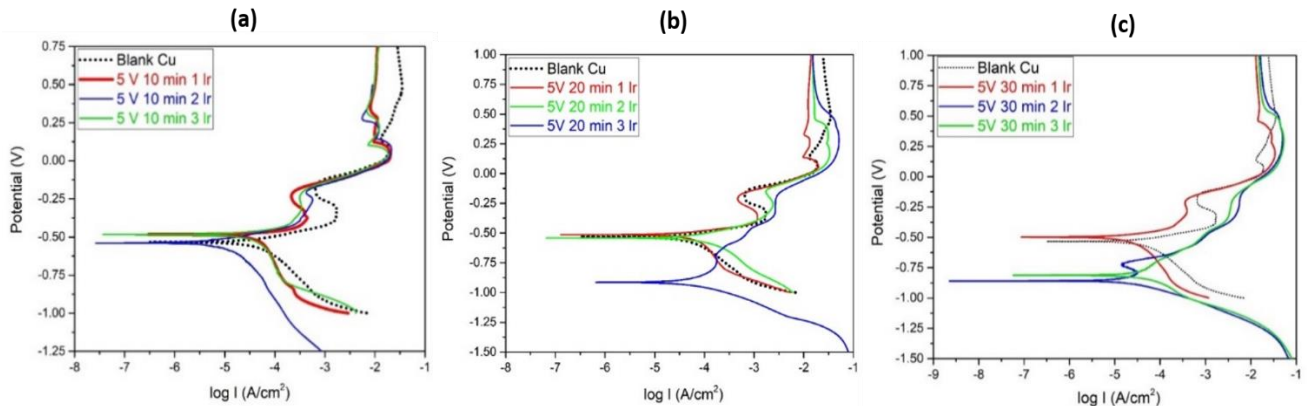| Sample	Corrosion rate
Blank copper	1.61 mm/a
5 V 10 mins 1 layer	0.64 mm/a
5 V 10 mins 2 layers	0.21 mm/a
5 V 10 mins 3 layers	0.63 mm/a
5 V 20 mins 1 layer	0.89 mm/a
5 V 20 mins 2 layers	0.30 mm/a
5 V 20 mins 3 layers	0.77 mm/a
5 V 30 mins 1 layer	0.40 mm/a
5 V 30 mins 2 layers	0.13 mm/a
5 V 30 mins 3 layers	0.42 mm/a

Fig. 7. Polarization curves for blank copper and different coating iterations (a) 5V 10 min, (b) 5V 20 min, (c) 5V 30 min

4. Summary

Successful exfoliation of graphene nano-sheets through an inorganic salt electrolyte. XRD and EDS studies substantiate the presence of graphene. SEM and TEM depict the morphology of graphene highlight the stacked as well as crumpled nature. Raman gives an idea about the quality of graphene and number of layers present. FTIR analysis show mild functionalised behaviour of the FLGPs. UV studies reveal the absorption peaks of $\pi \rightarrow \pi^*$ and n $\rightarrow \pi^*$ electronic transition of aromatic C=C and carbonyl C=O functional groups, respectively. SEM images showcases the multi-layered graphene coating on copper substrate. EDS analysis gives an idea about composition of the coating. Optical images shed light on the surface changes after corrosion. Tafel curves present the performance of the graphene coatings, which were found to be substantially effective in combating corrosion.

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