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SYNTHESIS AND CHARACTERIZATION OF GRAPHENE: ITS ELECTRICAL PROPERTIES FOR APPLICATION IN SOLAR CELL

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Abstracts

Graphene, a one-atom thick layer of graphite with a two-dimensional sp^2-hybridized carbon network, has recently attracted tremendous research interest due to its peculiar properties such as good mechanical strength, high thermal conductivity, superior transparency, large specific surface area and exceptional charge transport properties. Accelerating global energy consumption makes the development of clean and renewable alternative energy sources important. This research focuses on beneficiation of graphite using flotation separation techniques, and synthesis of graphene via electrochemical exfoliation technique, its characterization and measurements to investigate electrical properties of graphene. The obtained films of graphene were characterized using scanning electron microscopy (SEM) to examine the size, shape and morphology of graphene, while X-ray Diffraction (XRD) will be used to determine the degree of orientation and interlayer spacing of graphene layers and four point probe to determine the resistivity and electrical conductivity, also preparation of thin film electrodes for solar application.

Keywords: Synthesis, Characterization, Graphene, Solar Cell.

1.0 INTRODUCTION

The development of clean and renewable energy is vital to meet ever-increasing global energy demands arising from rapid economic expansion and increasing world population, while minimizing fossil-fuel depletion, pollution, and global warming (Doung, 2002). The increasing consumption and the rapid depletion of fossil fuels has given room for major research on exploitation and utilization of renewable energies such as geothermal, biofuel, tidal, wind and solar energy for past decades. To provide a wide spread usage of renewable energies, efficient energy storage and conversion technologies are required such as: lithium-ion batteries (LiBs), superconductors (SCs), and fuel cells (FCs) are termed representative of energy storage and conversion system as they rely on common electrochemistry principle (Manthiran et al, 2008).

Recently, nanotechnology opens up new frontiers in materials science and engineering to meet this energy challenge by creating new materials, particularly carbon nanomaterials, for efficient energy conversion and storage. With the rapid development of nanoscience and technology, nanostructured carbon materials, such as graphene (Nishihara and Kyotani, 2012), carbon nano fibers (Hammel et al, 2004) and nano tubes (CNTs) (Wei et al, 2011), have been explored to prepare hybrid materials with diversified morphology and intriguing properties. The remarkable physical properties of these nanocarbons especially graphene can be transferred to the frameworks of hybrid materials, leading to significant performance enhancement. To take advantage of its unusual properties, graphene has been widely studied in various energy conversion and storage applications. Amongst these applications, the development of efficient solar cells, which can convert sunlight into electricity, is in high demand in order to solve up-coming energy-related and global warming issues.

Graphene atom-thick graphite has attracted intensive interest due to its two-dimensional and unique physical properties, such as high intrinsic carrier mobility (~200 000 cm^2/V s), excellent mechanical strength, elasticity and superior thermal conductivity (Bolotin, et al, 2008). Modern electronic devices, including touch screens, flexibledisplays, printable electronics, light emitters, and solar cells, rely on transparentconducting oxides (TCOs), such as indium tin oxide (ITO) and ZnO/Al(ZnO) (Gordon, 2000). In addition, the indium (In) is expected to be depleted in a few years; therefore, it is urgent to seek promising replacements.Graphene is an optically transparent material, absorbing only 2.3% of visible light, and highly conducting material. Hence, it is considered as highly promising for replacing ITO materials.

Recent works on CVD methods using catalytic metal substrates have shown the capability of growing large-area graphene, greatly encouraging their applications in highly transparent and flexible conducting films,(Li et al 2009) although more efforts should be made to lower the production costs, particularly those associated with the high-temperature process and expensive substrates. Chemical exfoliation methods based on the Hummers' method, oxidation of graphite into thin graphene oxide (GO), followed by chemical or thermal reduction, have recently drawn much attention due to the advantages of potentially low-cost and solution-processed fabrication (Cote, et al, 2009.). However, the oxidation process severely damages the honeycomb lattices of graphene.

Also, the subsequent reduction of GO sheets typically involves high temperature to recover the graphitic structure (Li, et al. 2009). Moreover, the resistance of the films obtained from reported reduced GO (rGO), ranging from 1k to 70k Ω/sq (<80% transmittance) (Wu et al. 2008) or from 31k to 19M Ω/sq (at 95% transmittance), (Zhu, et al, 2009) is still much higher than that of ITO. Several other exfoliation methods attempting to obtain highly conductive graphene sheets have also been realized: (1) liquid-phase exfoliation of graphite by extended sonication, (Gu, et al, 2009) and (2) intercalation and expansion of graphite with volatile agents (Lee, et al, 2009).

However, the size of graphene sheets obtained by liquid-phase exfoliation or intercalation of graphite is normally smaller than $1\mu m^2$. The transparent conducting (TC) film made by such small sheets exhibits

transparency from 83 to 90% and resistance from 8000 to 5000 Ω/sq.(Li, et al,.2008) The high resistance of these TC films is caused by the damage during exfoliation and large amounts of inter sheet junctions.

Here we demonstrate a novel one-step approach of obtaining high-quality graphene thin sheets by electrochemical exfoliation of graphite. The lateral size of the exfoliated graphene sheets ranges from several up to 30 μm, which significantly reduces the number of inter sheet junctions for making percolative TC films. The TC film made by these exfoliated graphene sheets exhibits excellent conductivity (sheet resistance is 210 ohm/sq at 96% transparency). Moreover, the effective field-effect mobility extracted from the single-sheet device is readily up to $17cm^2/V^3s$ (Su, et al, 2011)

Electrochemical synthesis of graphene is considered cheaper and greener method potentially capable of mass production of few-layer graphene (FLG). Using electrochemical exfoliation method Liu *et al.* peeled FLG off graphite at anode in ionic liquids (Su, et al, 2011). Here we demonstrate a novel one-step approach of obtaining high-quality graphene thin sheets by electrochemical exfoliation of graphite. This solution based method makes graphene amenable to be processed into the form of film, on most substrates including flexible type.

1.2 ELECTRICAL PROPERTIES OF MATERIALS

Forces are one of the basic means by which they characterize materials. When presented with a new material they immediately want to know two things: how the electrons in the material respond to electrical forces and how the atoms respond to mechanical forces. The first of these is summed up by Ohm's Law:

$V = IR$ ……………………………………………..1.0

Where V is the voltage difference across the conductor, I is the current, and R is the resistance. A useful way to express this resistance is in terms of a resistivity ρ defined as:

$R = \frac{\rho L}{A}$………………………………..………………1.1

Where L is the length of the material and A is the cross sectional area. The resistivity of a material is independent of its geometry making it a useful quantity to compare different materials.

Ohm's law is a general formula applicable to 3D, 2D, and 1D conductors. In a typical conductor charges are moving and scattering at random with no net movement of charge across the sample. This situation changes when a voltage difference, V, is applied across the conductor. The voltage difference creates an electric field, E, which gives these randomly scattered electrons a net force in one direction. Some of the possible scattering mechanisms are phonons in the material, defects in the lattice, or charge inhomogeneity in the material. The velocity with which the charges move in the direction of the applied field is known as the drift velocity, v_d and is related to the current density J by:

$J = nev_d$……………………………………………..1.2

Where n is the charge carrier density and e is the electron charge. When there is less scattering in a material, the charge carriers will travel farther with the same electric field. This ratio is defined as the mobility,

$\mu = \mathrm{v_d}/\mathrm{E}$ …………………………………………… 1.3

and is an important quantity that is used to characterize scattering in conductors. One can then express the resistivity of a material in terms of its mobility by:

$\rho = {}^{1}/_{ne\mu}$……………………………………………..1.4

1.3 THE GRAPHENE STRUCTURE

For many years, graphene was the missing allotrope of carbon, after the discovery of graphite, diamond, carbon nanotubes and fullerenes. Graphene was discovered in the late 2004 at the Centre for Mesoscopic and Nanotechnology of the University of Manchester in the United Kingdom, directed by A.K. Geim and K.S. Novoselov(Novoselov et al 2004., Novoselov et al 2005). Graphene was obtained by the cleavage of a single atomic layer from a sample of graphite using Electrochemical exfoliation technique (Neto et al. 2009). Graphene is a one-atom thick sheet of carbon atoms, arranged in a honeycomb (hexagonal) lattice and is two dimensional (http:doi:10.1016/j.vacuum). Graphene as one allotrope of carbon is black in colour and is a very soft material compared to hard diamond. The softness of graphene is due to the fact that it has out of plane vibrational modes (phonons) which are absent in three dimensional solids.

To date, graphene is the building block of all other modern allotropes. By rolling it in one dimension it becomes a carbon nanotube while by stacking in three dimensions, it becomes graphite and it can be wrapped to form a zero dimensional fullerene (Charlie et al, 2007). Most theoreticians doubted the existence of graphene thinking that it might be highly thermodynamically unstable until it was found that its stability is similar to that of graphite.

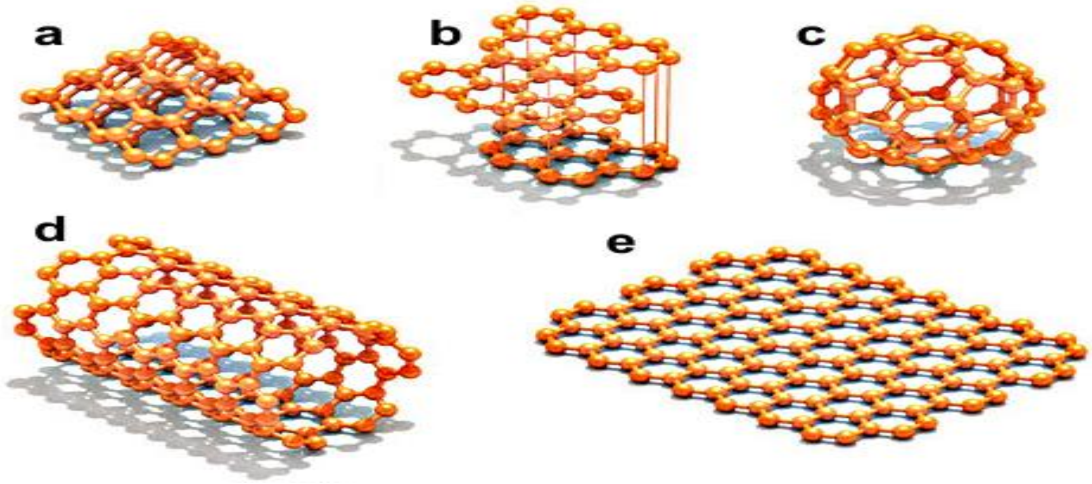

Figure 1.3: The carbon family and their considered material dimensionality. (a)Diamond (3D),

(b) Graphite (3D), (c) Fullerenes (0D), (d) Nanotube (1D), (e) Graphene (2D).

Whenever one writes with a pencil, it releases black pieces containing graphene layers. The first graphene membrane was produced with an area of $1mm^2$(Geim, 2009). Electrons in graphene behave as `Dirac Fermions' and mimic the dynamics of hyper-relativistic electrons (Neto et al., 2006). The Dirac Fermions move at a speed of 10^6m/s which is 300 times less than the speed of light. They behave differently to ordinary electrons especially when exposed to a magnetic field (Guysin et al, 2006). It was observed that graphene exhibits high carrier mobility of electrons which display unusual dependence on the concentration of impurities (Neto et al, 2006).

1.3.1 STRUCTURAL PROPERTIES OF GRAPHENE

Graphene has a two dimensional hexagonal structure with a space group of P6/mmm with the lattice vectors expressed as follows,

$$\bar{a}_1 = \frac{a}{2}(3, \sqrt{3}), \quad \bar{a}_2 = \frac{\bar{a}}{2}(3, -\sqrt{3}) \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots 1.14$$

The nearest neighbour distance (carbon-carbon distance) is represented by the letter (a) in equation above and is approximately $a \approx 1.42$Å. Graphene has lattice constants of $a_1 = a_2 =$ 2:46Å (Geim, 2009) and is a triangular lattice with a basis of two atoms per unit cell. When extending the graphene layer, we consider the other three next nearest neighbour vectors given by,

$$\sigma_1 = \frac{a}{2}\left(1, \sqrt{3}\right), \ \sigma_2 = \frac{a}{2}(1, -\sqrt{3}), \ \sigma_3 = -a(1,0) \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots 1.15$$

There are two types of graphene structure, namely the zigzag and armchair type.

These structures differ according to their orientations and the directions of the edges. By looking at figure (1.4) and considering the edge along the y axis, we see an armchair structure. Using the edge along the x-axis, we see the zigzag structure.

1.5 PROBLEM STATEMENT

Graphene, a single layer of graphite, has stimulated enormous scientific interest.

Its many unique properties are still not properly understood, and need further investigations. Electrical properties in the past few years have contributed to the understanding of graphene for solar cell applications. There is still more effort needed to understand the unique properties of this material. Hopefully, if such challenges are resolved, graphene will find more applications in energy conversion and storage.

1.6 AIM AND OBJECTIVES

The aim of this research work is to synthesis and characterizes graphene via electrochemical exfoliation, to determine its electrical properties for application in solar cell.

The objectives are as follows;

- To investigate resistivity and conductivity measurements of graphene
- Scanning Electron microscopy (SEM) will provide us with information about the size, shape and morphology of graphene.
- X-ray Diffraction (XRD) will be used to determine phase composition, crystal structure, Texture/Orientation, crystallite size and micro strain of the sample.
- Preparation of thin film electrodes for solar application.

EXPERIMENTAL WORK

3.1 GRAPHITE BENEFICIATION PROCESS

Graphite is an important industrial mineral that can be found in some Northern states of Nigeria. The graphite ore used for this research work was mined in Jalingo, Taraba state. The geographical coordinate of Jalingo, Taraba, Nigeria is $8^{0}53^{\prime}$N. Two types occur in nature, crystalline and amorphous. Crystalline graphite is used principally for crucibles and shaped structures. Amorphous graphite is used extensively in the steel and lubrication industries.In many deposits the two types grade into each other and the ore is of such low grade that beneficiation such as by flotation must be used.

PROCEDURE

The beneficiation of the graphite ore was carried out at department of mineralogy Kaduna state Polytechnic, Sample of lumps graphite as received were crushed and pulverized, sieved to -90um.

Concentration process adopted was froth flotation, on a 12D Denver laboratory type. Impeller speed of the machine was kept constant at 1500 rpm. 300g of graphite weighed using the weighing balance. Distilled water of 1000cm^3 was measured and mixed with the graphite sample, poured into the flotation cell and agitated for 3 minutes. During the agitation process, three drops of regulator (sodium hydroxide) was added to adjust the pulp pH to 8.5 and conditioned for seven minutes. Thereafter, 3ml of kerosene and pine oil added. Immediately after the conditioning/agitation, air was allowed to pass through the pulp at a reasonable rate and froth emerged and collected till barren froth surfaced.

Table 3.1: Shows fixed carbon content of raw graphite (as received)

Head sample	Content %			
	MC	VM	AS	FC
	14.51	19.76	39.48	26.25

Note: MC = moisture content, VM = volatile matter, AS = ash content, FC = fixed carbon

Table 3.2: Shows fixed carbon content of the concentrate

Concentration	Content %			
	MC	VM	AS	FC
	2.97	5.45	10.30	81.28

Electrochemical exfoliation

The Electrochemical cells consisting of two electrodes: an anode (the electrode occurs oxidation reaction) and a cathode (the electrode occurs reduction reaction). The concentrated HNO_3 /H_2SO_4 in 1:3 volume ratio was used to prepare graphene, so solution (0.69 gm of H_2SO_4 and 0.19 gm of HNO_3) to make its pH value around 3 using graphite as anode and cathode. We performed electrochemical exfoliation of the graphite in the H_2SO_4 and HNO_3 to obtain high quality of the graphene, as shown in fig.3.4 the experimental setup. The bias of 1 Volt was first applied of the graphite electrode for 5 minute, then by increasing the bias to 10 Volt for other 5 minute. The elementary low bias helps to moistening the sample, before implementation a high bias of 10 Volt, graphite still yet as a one piece. Once has been applied the high bias to the graphite, was quickly separated into small pieces and spread in the solution surface. The

graphite works as the electrode and source of the graphene for exfoliation electrochemical. The process of electrochemical exfoliation was conducted by applying constant current (DC) bias on the graphite electrode. Noted that 10 volt activated to the exfoliation and oxidized graphene. Remove undesirable of large graphite particles produced in the process, the solution is left for 3 or 4h to take it enough time to drop down to bottom, and it can then be used for more characterization and film preparation. Occur of all these experiments electrochemical exfoliation at room temperature. The application of high voltages on the anode resulted in the slow exfoliation of graphite through edges. During the exfoliation there are two types of graphitic flakes formed; one gets regimented at the bottom which consists of thick graphitic pieces. The second type of graphitic sample floats on the surface of electrolyte. These flakes are nearly transparent and have been found to consist of few layer graphene (FLG). To prepare the graphene sheet suspension, the exfoliated graphene sheets were collected with a 100 nm porous filter and washed with DI water by vacuum filtration. After drying, they were dispersed in DMF solution by gentle water-bath sonication for 5 min. To remove unwanted large graphite particles produced in the exfoliation, the suspension was subjected to centrifugation at 2500 rpm. The centrifuged suspension can then be used for further characterizations and film preparation.

All of these electrochemical exfoliation experiments were performed at room temperature (25 ± 3℃).

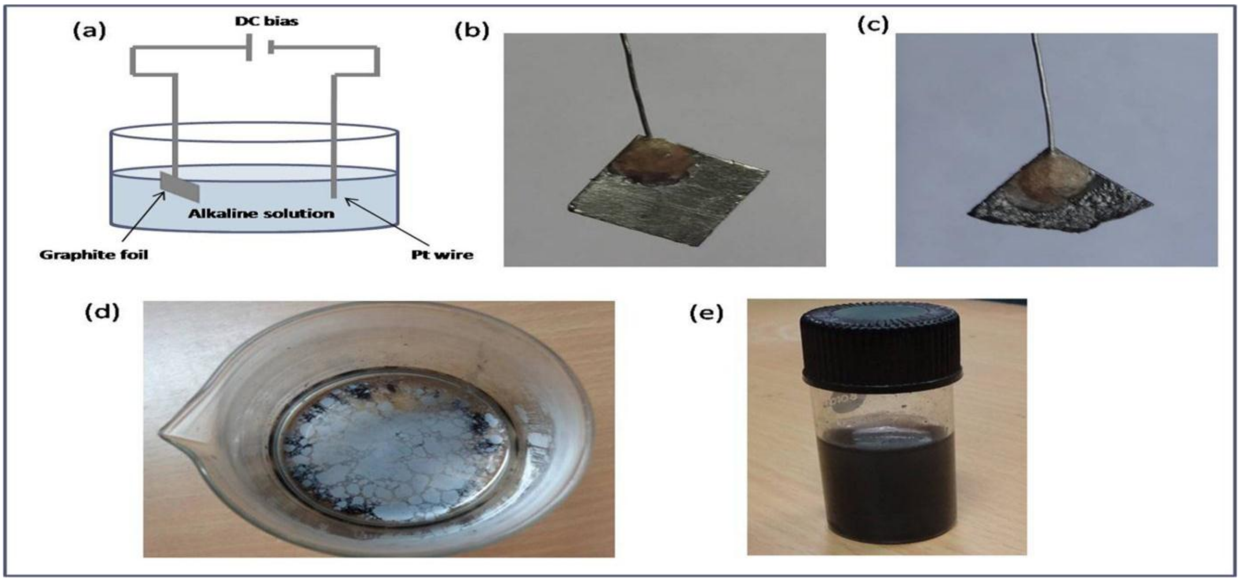

Fig. 3.4. (a) Schematic illustration of the electrochemical exfoliation setup. Optical images of (b) graphite foil before exfoliation (c) graphite foil after exfoliation (d) graphene sheets floating on the surface of electrolyte and (e) dispersed graphene sheets in DMF solution

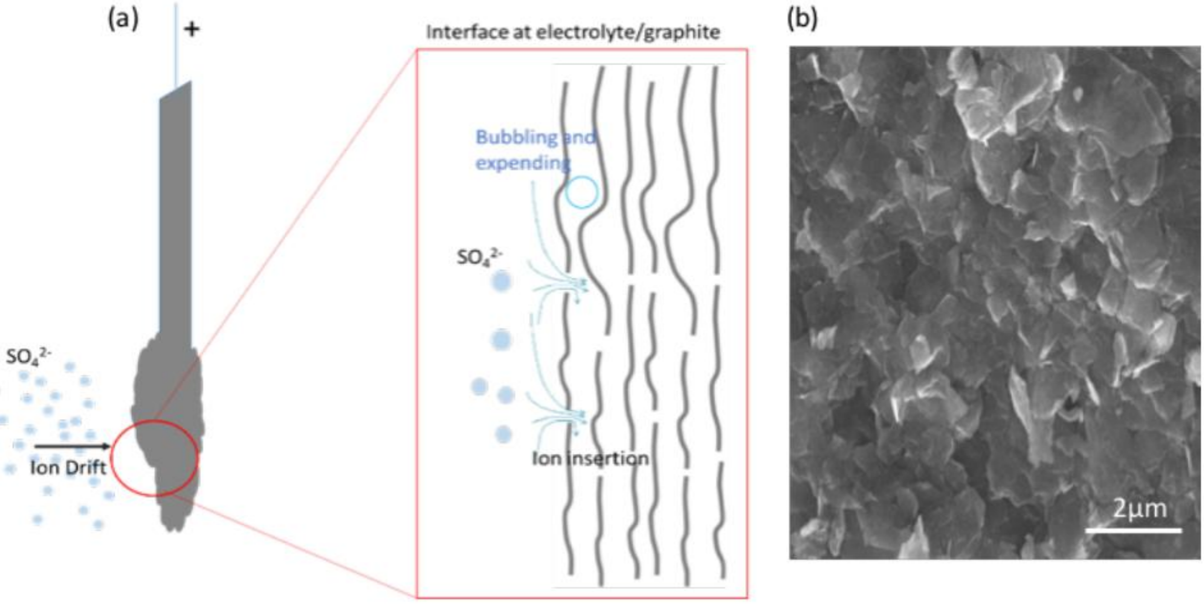

Figure 3.6: (a) proposed mechanism for electrochemical exfoliation of graphene, (b) SEM secondary electron image of graphite electrode surface following electrochemical exfoliation process.

3.3 MATERIAL CHARACTERIZATION

3.3.1 APPLICATION OF XRD TO GRAPHENE LAYER

This is the basic analysis to know the crystal configuration and orientation of the nano crystalline material. A diffraction pattern results, when the X-rays comes in contact with a crystalline phase and show the different orientations and the inter layer spacing of atomic layers. The grain size can be calculated by

Scherrer's Formula,

$$D = {}^{0.9\lambda}\!/_{\beta cos(\theta)} \cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots 3.1$$

D is the mean grain size; λ is the wave length of X ray radiation used, β is the line broadening of full width at half maxima (FWHM) in radians and θ is the Bragg's diffraction angle in degrees.

3.3.2 APPLICATION UV-VIS SPECTROMETER

This is used to measure absorption or transmission in transparent or opaque solids and liquids. In this technique, a beam of light is passed through sample and the remaining light is monitored in a detector. The range of wavelength is 200-800 nm in the case of UV-VIS spectrometer. As the light falls on the sample the light which is being passed through sample gets absorbed by some of the molecules present on the sample depending upon there structure and chemical bonding giving peaks at various wavelengths in this range. The first figure 3.7 shows the absorbance peaks at various wavelengths.

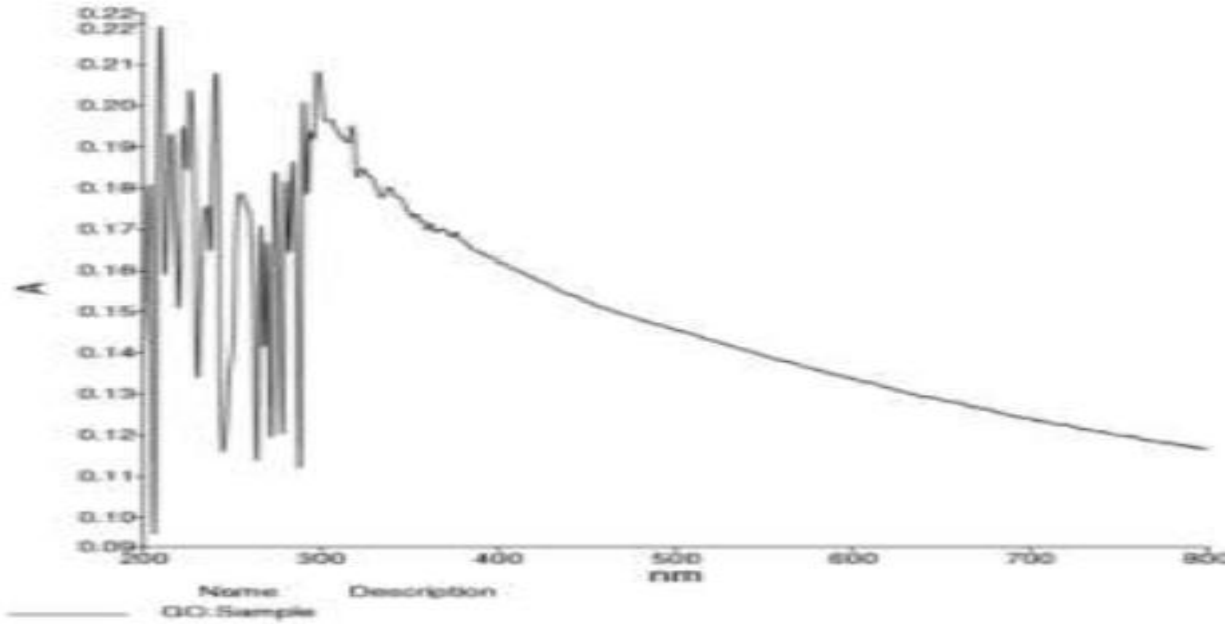

Figure 3.7 UV-Vis Spectrum on glass

3.3.3 APPLICATION OF SEM TO GRAPHENE LAYER

For morphological and structural analysis scanning electron microscope images using SU8000 series in Lens mode are provided here at different magnifications. Scanning electron microscopy (SEM) is a visualization tool for imaging on the order of 30 nm to 1 µm. The surface details of the exfoliated graphite samples were analyzed from the SEM images.

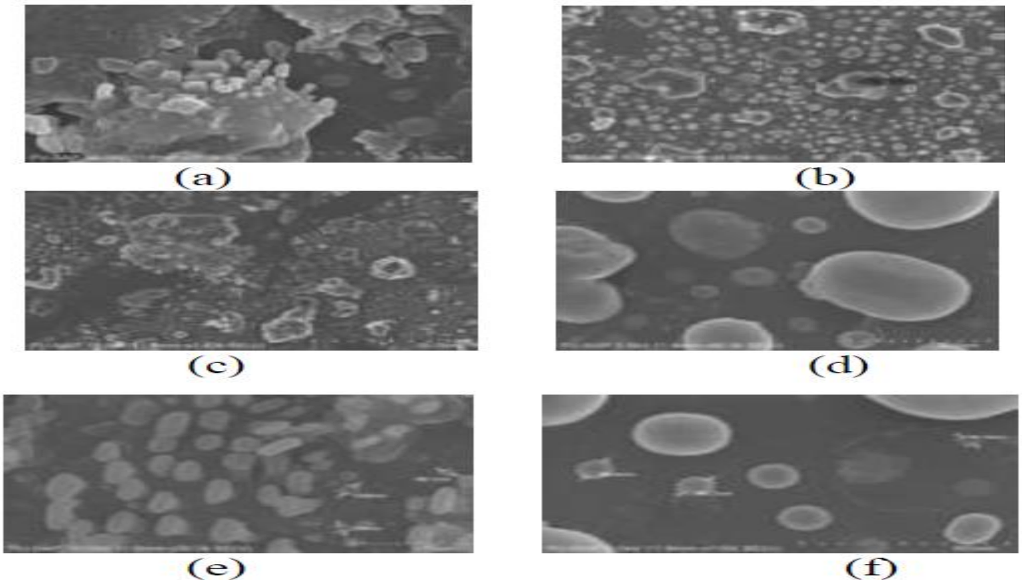

Figure 3.8: SEM results for Graphene at different magnifications.

3.3.4 RESISTIVITY AND CONDUCTIVITY MEASUREMENTS

Table 3.3 Parameters for calculation of the electrical conductivity of graphene film.

Parameters	L(mm)	R(Ω)	W(mm)	D(μm)
Value	3.79	33.14	3.34	7 ± 1

In measuring the electrical conductivity, the graphene film was cut into a smaller piece with 3.14 x 8.00 mm^2 in dimensions. After fabrication of four electrodes using platinum wires at two sides of the film, of which two electrodes were used to input linearly changing direct current and another two electrodes were used to detect the output voltage, the I-V plot of the film was obtained using PPMS as shown in figure 3.9. From this plot, the electrical conductivity of the graphene film was calculated to be 4990±710 Sm^{-1}according to the following equation:

$$\sigma = L/RWD \ \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots 3.2$$

Where L is the distance between the two electrodes outputting the voltage, R is the resistance obtained from the slope of the I-V plot, and W and D are the width and thickness of the graphene film respectively. The values of $L, R, W\ and\ D$ are given in the table 3.3. Such a high electrical conductivity is attributed to the fact that few oxidation reactions and little destruction to the graphene occurred in our method of preparation and process of graphene.

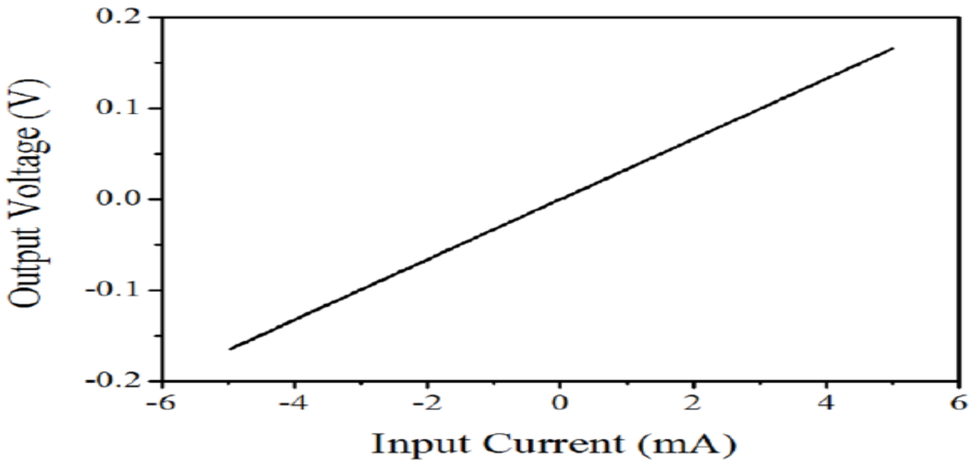

Figure 3.9: I-V plot of graphene film obtained by PPMS. The resistance of the film is 33.14Ω, which leads to an electrical conductivity of 4990±710 S/m for the graphene film.

3.4 FABRICATION OF FIELD-EFFECT TRANSISTOR DEVICES. The exfoliated graphene sheets were deposited onto the silicon substrates with a 300 nm silicon oxide layer by the dip-coating method, followed by a baking at 190 ℃ to remove solvent. The field-effect transistor device was fabricated by evaporating Au electrodes (30 nm thick) directly on top of the selected, regularly shaped graphene sheets using a copper grid (200 mesh, 20 μm spacing) as a hard mask. The typically obtained channel length between source and drain electrodes was around 20 μm. The electrical measurements were performed in ambient conditions using a Keithley semiconductor parameter analyzer, model 4200-SCS.

3.5 PREPARATION OF THIN-FILM ELECTRODES. For preparing electrodes, quartz or glass substrates were first cleaned with a Piranha solution to remove undesired impurities and to make the surface hydrophilic. The graphene solution with the concentration of 0.085 mg/mL in DMF was dropped (~500 μL) onto the cleaned substrate, followed by adding a drop (100-600 μL) of deionized (DI) water. The thin graphene film was then self-aggregated at the solution surface. After that, the substrates were heated on a hot plate at 190 ℃ for 30 min to evaporate the residual DMF. To treat the thin-film electrode with HNO_3, the as-prepared samples were dipped in a 69% of HNO_3 solution at 80 ℃ for 1 h. For the thermal annealing process, the samples were loaded into a quartz tube in a furnace, where a mixture gas of H_2/Ar (20 sccm/80 sccm) was directed into the quartz tube at 450 ℃ for 30 min (pressure fixed at 500 Torr).

4.0 Results and Discussion. The X-Ray diffraction (XRD) is employed for the identification and understanding the crystalline growth nature of grapheme prepared by the electrochemical exfoliation method. In Fig. 3.10, shows the natural graphite (graphite electrodes), the graphite has a powerful and sharp peak at 26.6^0 of 2θ corresponding to the highly organized layer structure with an interlayer distance of 0.33 nm along the (002) orientation, also has diffraction peak at 54.6^0 of 2θ corresponding to the

structure of layer with interlayer distance of 0.16 nm along the (004) orientation. The diffraction pattern of graphite electrode shows two different located peaks (002) and (004) at ($2\Theta = 26.6066\ ^0$ and54.6870^0),

respectively. Using data from X-ray diffraction (XRD), and fig. 3.10 and fig. 3.11 of the X-ray shows the patterns of XRD for graphene and was seen clearly (002) peaks, which have been mapped also to the distance layer to layer (d spacing = λ / 2 sin θ).The diffraction pattern of graphene as prepared shows different located peaks (002) at ($2\Theta = 26.7192^0$), see fig.3.11. The X-ray diffraction patterns of graphene show high diffraction peaks showing good crystallinity. The diffraction peaks agree with those given in JCPDS data card of graphite (002) and (004) plane. X-Ray diffraction (XRD) was used to verify these steps during the reaction and also to investigate if any other defects would be introduced to the lattice during this process.

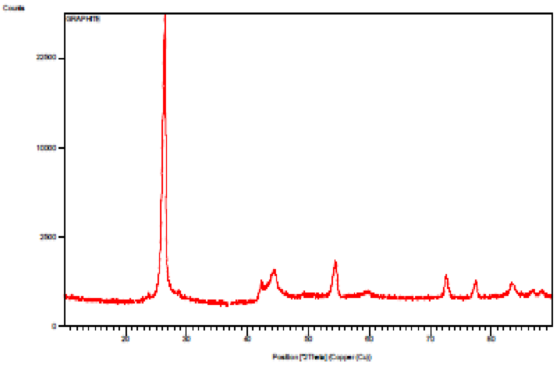

Figure 3.10: XRD results of natural graphite powder

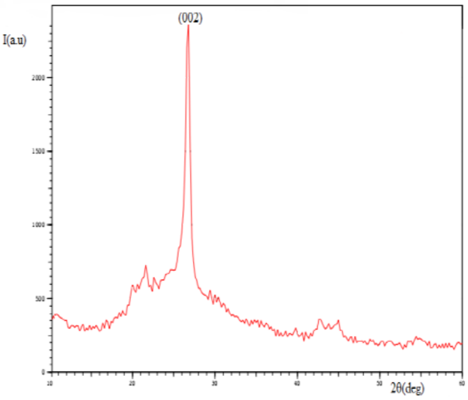

Figure 3.11 XRD pattern of graphene

Conclusion

In conclusion, a one-step method of obtaining high quality graphene sheets is demonstrated by electrochemical exfoliation of graphite. The exfoliated graphene sheets exhibit lateral size up to 30 μm.

Most (>60%) of the obtained sheets are bilayeredgraphene with A-B stacking. The field-effect mobility is up to $17cm^2/V^3s$, and the TC film made by self-assembled graphene sheets exhibits excellent conductivity (sheet resistance is ∼210 ohm/sq at 96% transparency). This work provides an efficient approach to obtain high-quality, cost-effective, and accessible production of "graphene ink", which may pave a way toward future applications in flexible electronics. As no oxidation reactions and little destruction occurred, the obtained graphene has few defects and functional groups. This property makes the graphene possess a high electrical conductivity of about $5000\mathrm{S}m^{-1}$.

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