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### Microwave Irradiation on Synthesis of High Quality Graphene Nano Sheets

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#### Abstract

Graphene, a single atom thick monolayer sheet of hexagonally arrayed sp<sup>2</sup> bonded carbon atoms in honeycomb crystal lattice has a lot of attraction and tremendous attention from the both in experimental and theoretical scientific communities in the recent world due to its enormous physical and chemical properties. Here in this present research work, the synthesis of Graphene Oxide (GO) and Graphene (G) nano sheets are prepared and developed on large scale by using chemical exfoliation method by simply controlling the oxidation and the exfoliation procedures (Improved Hummer's method) to produce a high and fine quality of graphene. The graphene formed is microwave irradiation to give pure and layered graphene. The material characterizations are confirmed by X-ray diffraction (XRD), Raman, FT-IR and Scanning Electron Microscope (SEM) was reported.

**Keywords:** Graphene Oxide, Graphene and Microwave Irradiation

#### Introduction

Graphene is thinnest and thickest material known today to the world of science and technology is the mother of all other carbon nano materials, it can be rolled up into 1D carbon nano tubes, formed to 0D buck ball fullerenes and stacked into 3D to form graphite[1]. Graphene has increasingly attracted the attention of owing its fascinating physical and chemical properties, including its unique electronic, thermal and mechanical properties [2]. These distinctive properties hold great potential applications in many fields of technological, such as nano electronics, sensors, nano composite, batteries, super capacitors and hydrogen storage [3]. In order to turn graphene applications into reality, one must fabricate and synthesis the material on large scale. Graphene can be prepared by various techniques and methods, but often been made by chemical vapour deposition method (CVD) [4], by which graphene films can be made on different metal substrate like (Ni, Si, Pt, etc.,). However, for ultra capacitor, transparent conducting electrodes and energy storage applications powder form is preferred and hence alternative chemical routes are required to produce in large. The chemical exfoliation strategy from natural graphite powder been suggested the best and effective general way to obtain the desired graphene layers in large and bulk quantity at very low cost[5].

#### Experimental Procedure

Synthesis of graphene is two-step process in which (i) synthesis of Graphene Oxide from graphite and (ii) Graphene from graphene oxide by controlling the temperature or water solvent in chemical route.

#### Synthesis of Graphene Oxide

In brief, 1g of natural graphite powder and 0.5g of NaNO<sub>3</sub> taken in 1L conical flask and mixed properly under stirring. Then slowly and gradually add 23 ml of 99% Conc. H<sub>2</sub>SO<sub>4</sub>, pre cooled down to below 0°C in ice bath condition for 30-45 min. separately and kept on stirring continuously for 30 min. Then after, 3g KMnO<sub>4</sub> slowly and gradually poured in to the solution. The solution color turns immediately to greenish yellow from black by maintaining the temperature of solution at 25°C under constant stirring conditions for an hour. Temperature of solution rises to 100°C. Now allow the temperature to fall down to room atmosphere and add the mixture of 120 ml of DI water and 10 ml of 30% of H<sub>2</sub>O<sub>2</sub> to terminate the reaction and kept on stirring for another 30 min. Next, the solution is washed several times with the solution 5ml of HCl and 100ml DI water to remove the impurities and sulphates ion and filtered to obtain a black cake. The cake is now micro wave irradiated at 450W for 60s and kept overnight to decantation. Finally, the obtained solid is rinse with water for 3 to 5 times and then wash with ethanol. Then the product is dried in vacuum oven for 24 hours at temperature of 80°C. A

fine dark black powder particle of Graphene Oxide is obtained.

**Synthesis of Graphene**

For the Graphene preparation, adding equal amount of graphene oxide and DI water taken in a flask and stirred for an hour at a temperature of 60°C. Then 1:5 volume ratio of hydrazine hydrate and DI water solutions are slowly and gradually poured to the above mixture at temperature of 90°C and continuously stirring for 4 hours to have complete reduction process. The color of the solution turns completely into dark black. Finally, the solution is washed with water and filtered to obtain a black paste. Then, the paste is microwave irradiate for 60s at 450 W and kept overnight to be settled down and dried in vacuum oven for 24 hours at a temperature of 80°C. A fine pure black powder particle of Graphene is obtained.

**Result and Discussion**

**X-ray diffraction analysis**

The XRD studies are carried for Graphite shows a major sharp high intensity peak value at  $2\theta = 26.4^\circ$  for corresponding (002) plane and small minor peaks at  $2\theta = 42.5^\circ, 43^\circ$  and  $55^\circ$  respectively for the corresponding planes (100), (101) and (004) for an “d” spacing of 3.37Å, 2.12Å and 2.03Å with an average particle size of 23 nm [11] are shown in fig1(a) and for that of graphene oxide the major predominant peak value is observed at  $11.6^\circ$  (002) plane with average particle size of 7.9nm for the “d” spacing of 7.63Å are shown in fig.1(b) [12]. when the annealing temperature of GO increases the major predominant peak value at  $2\theta = 11.6^\circ$  disappears giving rise to new high index and strong peak which is the transitional stage between GO and G, as r(GO) is obtained with a peak value at  $2\theta = 23.1^\circ$  for (002) plane and a small index peak of graphene existence is observed at  $2\theta = 43^\circ$  for (100) plane with an average particle size of 11nm [11] shown in fig.1(c). Finally for that of graphene crystal a broadened strong and major peak value are observed at  $2\theta = 26.4^\circ$  for (002) plane with “d” spacing of 3.40Å for an average particle size of 8.3nm is also shown in fig.1 (d) [13].

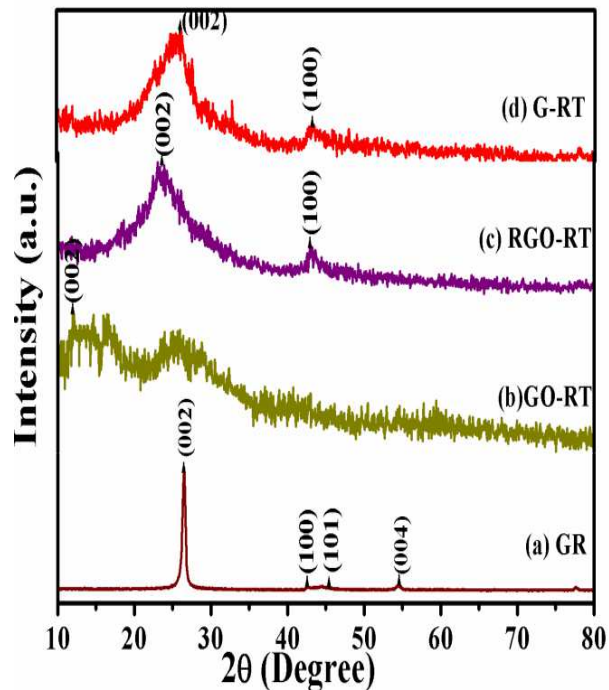


FIGURE 1. XRD of (a) Graphite, (b) Graphene Oxide (c) Reduced Graphene Oxide and (d) Graphene

The calculation of particle size, dislocation density and strain constraints nature of synthesized materials given in the tabular form for graphite, graphene oxide and graphene are given in table 1.

Materials	2θ	Size	“D” $10^{15}$ lines $m^{-2}$	Micro strain $10^{-3}$ line $^{-2}m^{-4}$
GO	11.6	7.9	0.15	4.33
rGO	23.1	8.1	0.14	4.24
	42.9	14.0	0.13	4.04
G	26.4	8.1	0.15	4.24
	43.0	8.5	0.13	4.05
GR	26.4	23.0	1.81	1.44

**Raman Spectroscopy studies**

Raman study of graphene oxide shows a high intensity major and strong D band peak value at  $1361\text{ cm}^{-1}$  which is the result of plane optical vibration and is due to first order resonance and a strong broadened G band is observed at  $1580\text{ cm}^{-1}$  [14] are the prominent peaks indicating the reduction in the functionality and decreases in particle size of

$sp^2$  plane domains, possibly due to the extensive oxidation of graphite shown in figure 2b and for that of graphene the major strong and high strength peak value of D and G bands are observed at  $1340\text{cm}^{-1}$  and  $1567\text{cm}^{-1}$ [12], indicating the presence of high purity carbon hybridization of atomic arrangement existing in multi channel layer and stacking of atoms makes the graphene different in strength and in layer formation shown in Fig.2(a)

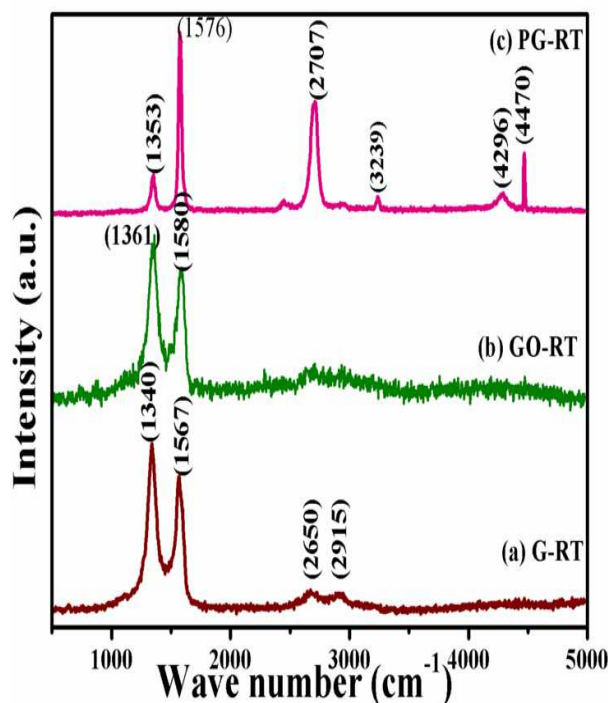


FIGURE 2. G and D bands of (a) graphite, (b) graphene oxide and (c) graphene

It is observed that, a small hump showing the presence of single layer stacking of carbon atoms in high crystalline nature in graphene, but the extent of layer formation is too small and not stable which is curbed at 2600 to  $2950\text{cm}^{-1}$  known as 2D band. The pure graphite G and D peaks and other sub micro peaks also shown in fig. 2(c) in comparison with G and GO but it consists of more number of impurities and multi layer formation are conformed. The peak value,  $I_D/I_G$  ratio observed to decrease from Graphene oxide to graphene shows the presence of more purity of carbon  $sp^2$  atoms arranged in well crystalline in nature.

#### FTIR analysis

The functional group of the synthesized materials has been confirmed by FTIR analysis which shows the presence of main compounds of graphite gives a strong peak at  $1059\text{cm}^{-1}$  (C-O),  $1120\text{cm}^{-1}$  (C-

O),  $1390\text{cm}^{-1}$  (C-OH),  $1580\text{cm}^{-1}$  (C=C),  $1720\text{cm}^{-1}$  (C=O),  $2350\text{cm}^{-1}$ ,  $2920\text{cm}^{-1}$  (C-H) and  $3530\text{cm}^{-1}$  (OH)[15]. For the Graphene Oxide, the functional groups are observed at  $1059\text{cm}^{-1}$  C-O (alkoxy/alkoids),  $1205\text{cm}^{-1}$  C-O (epoxy) and  $1390\text{cm}^{-1}$  C=C (skeletal vibrations);  $1580\text{cm}^{-1}$  C=C (stretching vibration);  $1720\text{cm}^{-1}$  stretching vibrations of C=O and  $3120$ ,  $3190$  and  $3420\text{cm}^{-1}$  at O-H (Stretching vibration), as the graphite converts to graphene oxide[16]. Some of the major functional group remains as of graphite, but new vibration of stretching modes and bending are formed due to high hydrophilic in nature due to the separation of oxygen functional group are shown in fig. 3(b).

Graphene shows a broad and high strength group absorption at  $1130\text{cm}^{-1}$  C-O (alkoxy),  $1580\text{cm}^{-1}$  C-C (carboxy)  $1720\text{cm}^{-1}$  C=O (carbonyl/carboxy) stretching vibrations and  $3035\text{cm}^{-1}$  (attributes to O-H stretching vibration)[17] are due to reduction of graphene oxide on thermal treatment makes the removal of some oxygen functional group and some loose bonding of  $\Pi$  and  $\sigma$  due to thermal effect and only the main carbon/carboxyl functional group shows the existing of graphite in nature in singular plane sheet are shown in the fig.3©

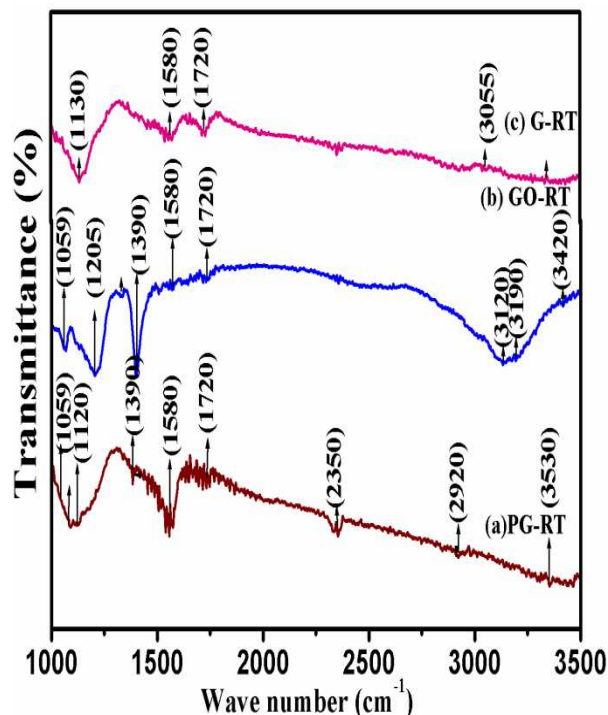


FIGURE 3. FTIR of (a) graphite, (b) Graphene Oxide and (c) Graphene

**SEM analysis**

Scanning Electron Microscope images reveals the clear information of morphology and structural nature of the synthesized particles as wrinkled nano buds of paper like structure for both the graphene oxide (fig 3(a)) and graphene (fig 3(b)) are shown which resembles like a jasmine flower in nature[17]. As the annealing temperature increases the particles size also increases having a well crystalline in nature and morphology.

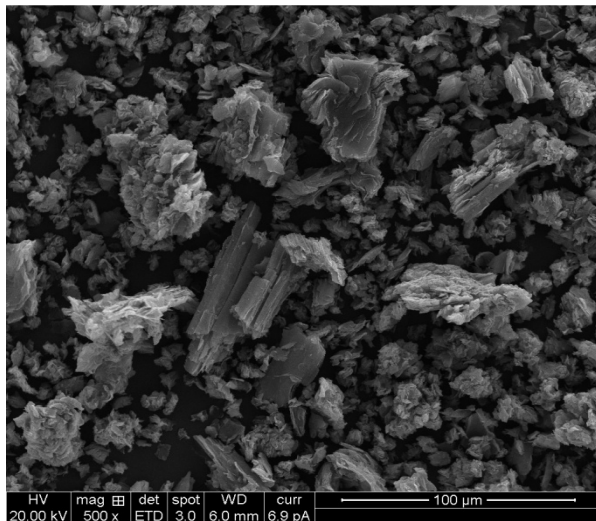


FIGURE 4(a). SEM image of Graphene Oxide

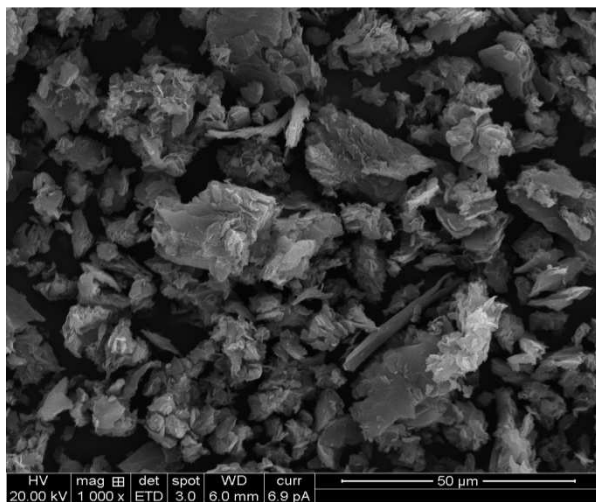


FIGURE 4(b). SEM image of Graphene

**Conclusion**

The XRD data reveals that the materials prepared are of high quality in nature and of very small particle with low crystalline formation for all the materials. The purity of the materials are confirmed by the Rama spectrum shows the perfect and absolute G and D bands and also predict the structural information of layers formed. The

identification of functional compounds groups by FTIR shows the presence of bending and stretching of carbon combinations in accurate manner. The SEM image gives a clear idea about the structural and morphology and the particle size, which matches with the XRD data.

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