# Interaction of plasma with Graphene Oxide: A review

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***Abstract*—*Reduced Graphene Oxide (RGO) is a promising two dimensional material which has got the potential of replacing the graphene. The zero band gap of the graphene is itself a big problem which needs to be resolved. The reduced graphene oxide is almost graphene having band gap. Out of a lot methods and techniques of synthesizing the RGO, one is the reduction of graphene oxide (GO) in the presence of plasma. Various plasma techniques for GO reduction, present in the existing literature, have been reviewed here. Ammonia plasma reduction, hydrogen plasma reduction, methane plasma reduction and argon plasma reduction with some more gases is presented in the report. Combination of two gases for plasma creation is also looked upon. Apart from gas plasma, the liquid plasma and plasma plum created by laser are also presented. Some literature reports on nanomaterial composite formation by plasma sintering are also discussed.***

Keywords —*Graphene Oxide (GO), Reduced Graphene Oxide(RGO)*

## I. INTRODUCTION

Graphene has attracted the attention of a lot of researchers all over the world. Graphene is a single sheet of carbon atoms detached from graphite, and we all know that graphite is an allotrope of carbon. So graphene exist in two dimensional space. The attraction toward graphene has a reason. Graphene has extraordinary electronic and electrical properties. The mobility of electrons are also very high in graphene. But the synthesis of pure graphene is still a challenge. A lot of methods have been used to synthesize graphene. Some of them give low yield and some of them gives impure graphene. So purpose is not fulfilled. Here in this study we are going to study the chemical route to make graphene. First an intermediate material graphene oxide (GO) will be synthesized. Then it will be reduced to make reduced graphene oxide (RGO). The whole purpose of studying RGO is that it shows some interesting properties like graphene. The making of RGO is not as tough as that of graphene. So interest in RGO is not a bad deal. It can be an alternate to graphene.

After a long time of wait ultimately we got graphene in the laboratory. Earlier graphene was a two dimensional abstract material having no real existence. It was just a theory derived very first time by Prof. P. R. Wallace in 1947[1].Interesting fact about the theory is that it was not derived in the name of graphene. The name graphene came after long time. The material was only in theories till 2004[2]. But A. K. Geim and K. S. Novoselov succeed in synthesizing graphene. They got Nobel Prize also for this. The other interesting fact about graphene is that More than 7 decade ago, Landau and Peierls proposed that two dimension crystals were thermodynamically unstable and could not able to exist in reality. That theory pointed out that a divergent contribution of thermal fluctuations in low dimensional crystal lattices as in case of graphene here, would lead to displacements of atoms in such a way that they will become comparable to interatomic distances at any finite temperature.

To make graphene oxide there are a lot of methods but most famous one, followed by a plenty of researchers all over the globe, is Hummer's modified method[3][4]. After getting the grapheneoxide(GO) the next step was to reduce the oxide part of the material and people did that with a number of methods. One of those methods is the thermal annealing of the GO with or without the presence of other gas and solid species. Research world got a lot of success in the reduction of GO by thermal annealing. Getting inspired by the thermal annealing/heating researchers developed some advanced techniques. One of them is the reduction through plasma. Plasma treatment is a green, mild and highly efficient method for the reduction of graphene oxide and hybrid nanomaterial synthesis. Here in this report, the reduction of graphene oxide (GO) with the help of plasma, is reviewed.Thevery idea of plasma is not new at all. Soon after the discovery of graphene. J. Wang et al. oxidized the graphene sheet by plasma and found that multilayer graphene did not get affected and showed no luminescence. But the monolayer graphene got affected by the plasma and became photoluminescent[5].

## II. INTERACTION OF PLASMA WITH GRAPHENE

So people started thinking of the plasma application onto the graphene and graphene like materials and fortunately got some interesting results. Researcher took one step further and applied different gas plasma onto the graphene. Shao et al. subjected the nitrogen plasma to the the graphene sheets and made it N-doped[6]. Lin at al. exposed the graphene to the ammonia plasma and electron

doping happened[7]. Childres et al. did apply the oxygen plasma to the graphene and created defects in the graphene sheets[8]. Y. Kim et al. synthesized the graphene manolayer sheets by using the Microwave plasma chemical vapor deposition. A mixture of hydrogen and methane was used for plasma formation. The sheets obtained by this approach were very big in size compared to the previous ones[9]. Nourbakhsh et al. noticed the band gap opening in the graphene after oxygen plasma treatment which converted it from semimatallic to semiconductor[10]. Kolesov et al. reported that the imposement of infrared laser pulses creates the high density electron-hole plasma in graphene and affects it[11]. Lombardo et al. made the manolayer sheets of graphene photo active by applying the oxidative plasma etching[12]. Imran Jafri at al. did a study in which they exfoliated the graphitic oxide thermally and made graphene nanoplates. These nanoplates were subjected to the nitrogen plasma for N doping of graphene occured [13]. Dato et al. synthesized graphene sheets by passing liquid ethanol droplets into an argon microwave plasma[14].

## III. INTERACTION OF PLASMA WITH GRAPHENE OXIDE

Apart from applying the plasma onto the graphene sheets people also used the technique to reduce and functionalize the graphene oxide sheets and got succeed. Graphene oxide(GO), reduced graphene oxide(RGO) and their composites with other nanomaterials have been widely used to make sensors, supercapacitors etc. Different indivisual gases and the mixtures of gases have been used to create the plasma. People applied the plasma with a variety of temperature and pressure conditions. The effect of plasma has been observed to change the material in every aspect. It affected or enhanced all the properties like electrical, electronic, optical and mechanical etc. In some cases the application of plasma introduced some new properties into the GO. The plasma species also plays a very important role in the defect reparation of GO.

### *A. Hydrogen Plasma*

Various effects of plasma has been observed onto the graphene oxide monolayers/multilayers. Reduction in sheet thicknees with lowering of O/C ratio and hydrogenation of $sp^2$ carbon network happened after hydrogen plasma onto GO. Increment of few orders in the electrical conductivity also happened[15]. The microwave exfoliation of graphite oxide in the presence of hydrogen plasma earned a graphene like material[16]. The reduction process monitoring with the help of optical emission spectroscopy is quite helpful and tells about the process, how and when it should be controlled[17]. The electrochemical modification of plasma treated GO was carried out in ethanol containing $Na_2PdCl_4$and $LiClO_4$ as supporting electrolyte. This electrodeposition of palladium introduced hydrogen sensitivity into the RGO layers[18]. The absorption capacity of the GO was found to be enhanced after having interaction with plasma. The resulting product was reduced graphene sheets which is a better absorber of 4,4'-DCB compared to Graphene oxide, carbon nanotubes or any other nanomaterials[19]. The conductivity and the field effect mobilitiy values were found to be increased by 2 or 3 order of magnitude having values ranging between 0.05 - 2 S/cm and 2 - 200 $cm^2$/V-s respectively. The charge transport through variable range hopping between intact graphene islands with sizes on the order of several nanometers was also confirmed by Raman spectrometry[20]. The generation of atomic hydrogen by microplasma of $Ar/H_2$ reduced the oxygen content from the GO and lowered the sheet resistance two order more lesser, without creating any defects in the GO sheets[21]. The elastic modulas of Hydrogen plasma treated GO showed values close to that of graphene. This makes it a qualified candidate for nanoelectromechanical devices[22]. A carbon dioxide gas sensor was developed by reducing the GO with Hydrogen plasma. Plasma interacrion turned the GO into RGO and introduced sensing capability into it. The radicals and atoms contained by the hydrogen plasma provides the enough energy for the dissociation of oxygen functional groups[23]. The dried mixture of Pt/GO was catagorized into four parts and all the four parts were subjected to Hydrogen plasma for different time duration and power condition which reduced them to Pt/RGO, having different properties[24].

### *B. Ammonia Plasma*

The another gas used to create plasma was ammonia which was a promising candidate to equipe the material with Nitrogen atoms. This decoration of N atoms, changed the properties of the material in several aspects.It has been observed that Nitrogen doping in graphene results in the shift of fermi level above the Dirac point followed by suppression of the DOS near the Fermi level. This opens up the band gap. Singh et al. developed a plasma system to reduce and dope the graphene oxide manolayer sheets. The electrical properties of N doped GO sheets were further investigated and an inhancement in the values was reported. Ammonia gets incoporated in the carbon network taking place of some carbon atoms which does functionalize the GO and repares its defects[25][26][27]. The removal of oxygen groups flattened the GO surface with 6% N doping and a lower oxygen content[28]. Kumar et al. reduced the graphene oxide using microwave plasma of a mixture of ammonia and hydrogen. The resulting material was Oxygen Reduction Reaction catalysis(ORR)[29]. Ammonia and hydrogen plasma treated graphene oxide was mixed with the solution

of $H_2PtCl_6$ and dried under nitrogen. This mixture was again treated with plasma and the Pt/GH and Pt/GHA catalysts got produced[30].

### C. Methane Plasma

Apart from hydrogen and ammonia some other gas plasma were also created. The application of methane plasma onto the GO sheets lowered the oxygen content and repaired the structure making it close to graphene which helped in enhancement of electrical conductivity[31]. Baraket et al. showed that electron beam plasma of argon and methane mixture reduced the graphene oxide. The study covers a spectrum of changes in the graphen oxide sheets at different flow rates of methane. Methane helps the argon in removal of the oxygen content. One more advantage of using methane is its carbon and hydrogen content which somehow helps the removal of oxygen and additon of carbon[32]. Oxygen plasma treated GO thin films were subjected to methane plasma. Thus reduced GO films were furthur used to fabricate micro-supercapacitors[33].

### D. Argon, Oxygen and other gas plasmaPlasma

Mechanicaly transferred GO onto silicon substrate was subjected to argon plasma in a RF plasma generator. This reduced GO showed characteristics very close to graphene[34]. The GO paper was exposed to the junction of negative-glow and Faraday-dark area in the glow discharge plasma of argon generated between two parallel plates of graphite[35]. The mixture of Graphene oxide and chloroplatinic acid ($H_2PtCl_6$) solution was dried under $N_2$ and then treated with argon plasma for 40 min under continuous stirring to grow Pt nanoparticles on graphene surfaces. This process produced Pt/GA catalysts which holds more tolerance toward carbonaceous species and is highly active towards methanol[36]. The Ni was dropped onto GO and resulting material was treated with the argon plasma which created NiO-NP-RGO hybrid nanostructure[37].Yu et al. synthesized the Pd/GO composite using the glow discharge of argon gas[38].To make improvement in the deposition of GO layer, the oxygen plasma was applied during spin coating of the GO solution onto the substrate. Plasma helped in thickness control of GO sheets. The GO was further used to make metal-insulater-metal structure[39]. A dc-pulse plasma jet of nitrogen at atmospheric pressure was used to synthesize the a flexible reduced graphene oxide (RGO) supercapacitor with polyvinyl alcohol & sulfuric acid gel electrolyte[40]. A low damage plasma of Argon and Hydrogen was exposed to the graphene oxide (GO) spin coated coating on a polymer (polyethylene terephthalate). The incorporation of atomic hydrogen was found to be increased, confirmed by optical emission spectroscopy. The reduction capability of plasma was calibrated with RF power and exposer time of the plasma[41]. The oxygen plasma treatment enhanced the surface reactivity of reduced graphene oxide (RGO) for better sensing of amyloid-beta peptides and the pathological hallmarks of Alzheimer's disease[42]. The reason, why Oxygen plasma is used for activation of surfaces is the content of the plasma itself, having high energy radicals, ions and electrons. All these species helps the plasma in the oxidation of the organic species present on the surface[43]. The hydrogen and oxygen plasma exposure to GO, used in organic light emitting devices, enhnaced the hole injection property[44]. The plasma exposure of argon and hydrogen increased the electrochemical stability of the GO[45].

## IV. Some Plasma Techniques other than glow discharge plasma

Apart from the glow discharge plasma of above mentioned gases some other plasma techniques have also been used in the synthesis and fabrication of RGO and its composites. Continuous reporting of all such techniques and methods is also available in the literature. These section includes all those techniques.

### A. Plasma Sintering

The plasma sintering is good technique for the in situ reduction and the production of nanocomposites. The GO and $Si_3N_4$ mixed in an attrition mill. This prepared mixture was subjected to spark plasma sintering(SPS) in a chamber having argon or nitrogen atmosphere. The mixture was treated with the SPS at different conditions , a couple of time. This whole process created a composite of reduced graphene oxide and silicon nitride[46]. A mixtue was made by the dripping of GO suspension into the suspension of alumina and heated in the presence of hydrazine monohydrate followed by a pressing by vacuum SPS, turned the mixture into graphene nanosheeet GNS/alumina composite[47]. Porwal et al. synthesized the graphene oxide nanoplates with the help of spark plasma sintering[48].Graphene oxide (GO) with different weight % was sintered onto a biomaterial by spark plasma sintering (SPS) during which GO turned to RGO and around 130 % increament in the fracture toughness of the biomaterial was found[49].

### B. Arc Plasma

Lu et al. reported the making of noncovalant hybrid nano structure of graphene oxide and silver for electronics and optoelectronics application. they decorated the graphene oxide sheets by silver nanocrystals. The suspension of GO was drop casted onto the transmission electron microscopy (TEM) grids. On the other hand, the aerosol silver nanocrystals were synthesized by the evaporation of silver wire using a mini-arc plasma mentained between the tungsten and graphite electrodes having

graphite as an anode. Such produced Ag aerosol nanocrystals were carried by argon or nitrogen between the two electrodes. One electrode contained the GO sheets with TEM grids and the other one contained the aerosol nanocrystals. A direct current was applied to the conductive grid which assemebled the oppositely charged aerosol nanocrystals were assembled onto the surface of graphene oxide sheets[50].

### C. *Pulse laser plasma plum*

The appliaction of the CW(continuous-wave) laser in air and $N_2$ background or pulsed laser in $N_2$ background onto the graphite oxide reduced the material and introduced graphene features into it. The electron-hole plasma was produced by the initial excitations within the material which made the oxygen-containing groups to trap excitons and holes. All this lead to plasma plume expansion which caused the nucleation and growth of graphene sheets and nanoparticles[51].

### D. *Liquid Plasma*

The elecrodes were immersed into a solution of GO and acetonitrile and a dicharge voltage was applied between them. The plasma reactions happened in the solution and produced functionalized graphene oxide. The process was made to occur at ambient temperature and pressure. The nitrile and amine gets inserted into the GO after the treatment turning the GO into fluorescencent material of higher quality[52].

### E. *Miscellaneous Plasma*

There are some other studies also of the plasma interaction with GO and RGO. The atmospheric pressure jet plasma treated RGO was used as an electrode in an solar cell(Dye Sensitized Solar Cell) instead of platinum. The efficiency of the solar cell was found to be changed as a function of plasma duration. The SEM pictures of the jet plasma treated RGO shows a modication in the sructure reducing its conuctivity and enhancing its catalytic activity simultaneously somehow. And the removal of organic part from RGO by jet plasma enhances the efficiency of the solar cell[53].The dry plasma treated hybrid nanostructure of RGO and NiO nanoparticles, used as counter electrode, increased the efficiency of the DSSC (Dye-Sensitized Solar Cell) upto 7.5% [37]. The inductively coupled radio frequency plasma of argon was applied to the $N_2$ dried mixture of GO and chloroplatinic acid with stirring of the mixture. This helped out in the growth of platinum nanoparticles onto the reduced graphene oxide. So reduction and composite formation happened simultaneously prducing the Pt/GA catalyst[36]. When GO is irradiated by a beam of argonions($Ar^+$),it gets reduced. A controlled reduction can be done[54].

## V. TABLE I

**DIFFERENT KIND OF PLASMA TREATMENTS APPLIED TO THE GO AND RGO**

| Sr. no. | Plasma Process | Gases Used | Refer ence |
|---|---|---|---|
| 1. | Glow discharge plasma at room temp. | Argon | [38] |
| 2. | High vacuum Glow discharge plasma (Faraday dark) | Argon | [35] |
| 3. | Atmospheric pressure and low temp. plasma | Methane, Hydrogen | [55] |
| 4. | Atmospheric pressure micro plasma at $150^0$C | Hydrogen, Argon | [56] |
| 5. | High vacuum and high temp. plasma | Oxygen, Methane | [33] |
| 6. | Room temp. plasma at 0.8 mbar | Hydrogen | [20] |
| 7. | Atmospheric pressure microwave plasma | Argon | [14] |
| 8. | Room temp. plasma at 1 Pascal | Ammonia | [25] |
| 9. | Liquid plasma at ambient condition | Acetonitril e | [52] |
| 10. | Plasma at 0.8 mbar | Hydrogen | [18] |
| 11. | Low temp. plasma at 10 Pascal | Hydrogen | [19] |
| 12. | CW laser pulsed plasma plum | | [51] |
| 13. | Steaming at $200^0$C | Oxygen | [57] |
| 14. | Low temp. plasma at 25 Pascal | Argon | [36] |
| 15. | Plasma at 50 Pascal &$120^0$C | Hydrogen | [15] |
| 16. | Room temp. microwave plasma | Hydrogen, Ammonia | [29] |
| 17. | Atmospheric pressure dry plasma at room temp. | Argon | [37] |
| 18. | Plasma | Hydrogen, Ammonia | [26] |
| 19. | Atmospheric pressure plasma jet at $400^0$C | Oxygen | [53] |
| 20. | Plasma at 15 Pascal and $30^0$C | Hydrogen, Ammonia | [30] |
| 21. | Mini arc plasma at $100^0$C, $200^0$C &$300^0$C | Argon | [50] |
| 22. | Spark plasma sintering at $1200^0$C & 50MPa | | [58] |
| 23. | Spark plasma sintering at $1300^0$C & 50MPa | Argon | [47] |

| | | | |
|---|---|---|---|
| 24. | Spark plasma sintering at various temp. and pressure | Argon | [46] |
| 25. | Radio frequency plasma at 0.2 torr and $560^0$C | Methane | [31] |
| 26. | Plasma at $750^0$C | Hydrogen | [31] |
| 27. | Plasma at 90 m Torr | Methane, Argon | [32] |
| 28. | Room temp. plasma at 20 mTorr | Hydrogen | [17] |
| 29. | Radio frequency plasma at 500 mTorr and $150^0$C | Ammonia | [28] |
| 30. | Radio frequency plasma at 4.7 Pascal | Hydrogen, Argon | [45] |
| 31. | Microwave plasma at 1-20 mbar | Hydrogen | [16] |
| 32. | Discharge plasma at 0.2 Torr | Hydrogen, Oxygen | [44] |
| 33. | Sputtering | $N_2^+$ ions | [59] |
| 34. | Ion beam irradiation | $Ar^+$ ions | [54] |
| 35. | Spark plasma sintering at $1400^0$C and 50 MPa | | [60] |
| 36. | Low energy ion beam at $10^{-7}$ mbar | Argon | [54] |
| 37. | Room temp. plasma at 100 mTorr | Ammonia | [27] |
| 38. | Radio frequency plasma at $10^{-2}$Torr | Argon | [34] |
| 39. | Spark plasma sintering at $750^0$C | | [61] |
| 40. | Spark plasma sintering at $550^0$C | | [49] |
| 41. | DBD glow plasma | | [62] |
| 42. | Plasma | $SF_6$, $CF_4$ | [63] |
| 43. | Screen plasma at 4 mbar pressure &$100^0$C-$200^0$C | Hydrogen, Nitrogen | [64] |
| 44. | Discharge plasma at 4.7 Pascal | Hydrogen, Argon | [65] |
| 45. | Atmospheric pressure plasma jet sintering | | [66] |
| 46. | Discharge plasma at 0.2 Torr | Oxygen, Hydrogen | [44] |
| 47. | Plasma at $800^0$C & 0.2-0.34 Torr | Methane, Hydrogen | [67] |
| 48. | RF room temp. plasma at 50 Pascal | Nitrogen | [68] |
| 49. | Glow Plasma | Oxygen | [39] |
| 50. | DC-pulse atmospheric pressure plasma jet | Nitrogen | [40] |
| 51. | Glow Plasma | Hydrogen | [24] |
| 52. | Low damage plasma | Hydrogen, Argon | [41] |
| 53. | Microwave plasma | Oxygen | [42], [43] |
| 54. | Electric discharge plasma at 0.1-0.01m bar & Room temp. | Air | [69] |
| 55. | RF plasma at room temp and 0.6-0.3 m bar | Hydrogen | [23] |
| 56. | Glow discharge plasma at 0.2 torr | Hydrogen, Oxygen | [44] |
| 57. | Microwave plasma | Hydrogen | [16] |
| 58. | RF plasma | Argon, Hydrogen | [45] |
| 59. | RF plasma at 20 m Torr | Hydrogen | [17] |
| 60. | Laser plasma plum | $CO_2$ | [73] |

## VI. EXPERIMENTAL OBSERVATIONS

The GO solutions obtained from Hummer's modified method can be transferred onto the substrates by Langmuir Blodgett(LB)[70][71] or Modified Langmuir Blodgett (MLB)[72]. Monolayer sheets were transferred onto the $Si/SiO_2$ substrates by Modified Langmuir Blodgett (MLB) technique. GO sheets reduced by ammonia plasma and hydrogen plasma can be seen in Fig.2.

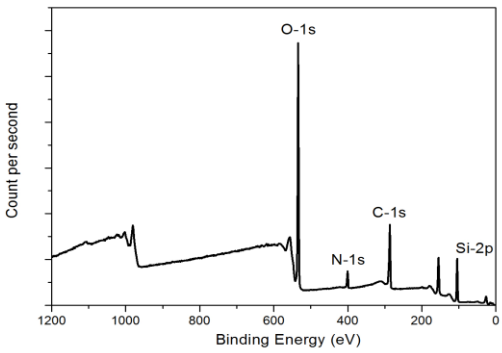


***Fig. 1***Survey scan of ammonia plasma treated GO

No morphological changes were observed in scanning electron microscopy (SEM) images after the plasma treatment. Sometime sheets get broken by plasma treatment but that is not the case here. The thickness and more clear measurement of the morphology of the GO and RGO sheets was done by Atomic Force Microscope (AFM). The thickness of the GO sheets got reduced from 1.2 nm to 1.0 nm after the ammonia plasma and hydrogen plasma treatments individually. This gives the rough estimate of the removal of the oxygen containing groups attached to the GO basal plane. Survey scan (Fig. 1), obtained from the X-Ray Photo Electron Spectroscopy (XPS) shows the incorporation of

ammonia atoms into the GO after 5 minute ammonia plasma treatment at room temperature. The de-convolution of the XPS spectrum states that the contribution from the $sp^3 - C$ containing functional groups is found to be decreased only marginally after the ammonia plasma treatment. So, plasma has led to the deoxygenation of GO with concurrent incorporation of nitrogen. This proves the N doping of GO.

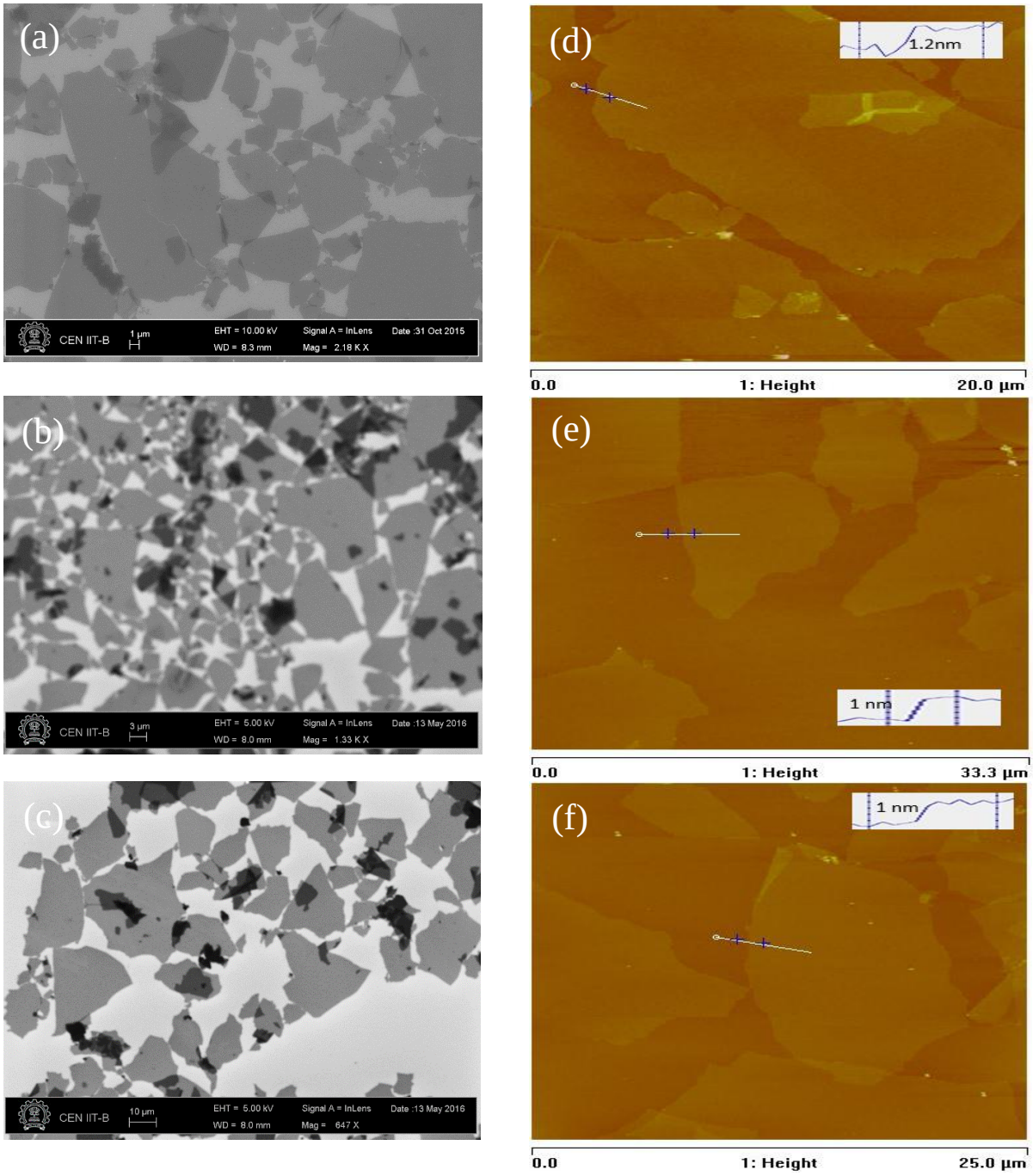


***Fig. 2***Scanning Electron Microscopy(SEM) image of (a) GO, (b) Ammonia plasma treated GO and (c) Hydrogen plasma treated GO. Atomic Force Microscopy(AFM) image of (d) GO, (e) Ammonia plasma treated GO and (f) Hydrogen plasma treated GO transferred onto $Si/SiO_2$ substrates.

## VII. SUMMARY

Plasma treatment is a green, mild and highly efficient method for the reduction of graphene oxide (GO) and hybrid nanomaterial synthesis. The properties of the final product can be tuned by tuning the plasma treatment conditions. Plasma treatment of GO in presence of different gas plasmas is presented in the report. Hydrogenation from hydrogen plasma, N doping from ammonia plasma is

discussed. Other gas plasma like methane, argon, nitrogen and oxygen are also found to change the GO properties like electrical conductivity, field effect mobility, optical activity, catalytic activity and chemical activity etc. These reduced GO films and sheets are also used for production of gas sensors. This method should also be suitable for the fabrication of other functional carbon based composite materials which can be used for various purposes. Sintering process helps in the synthesis of nanomaterial composites of GO and its reduced form (RGO) which can be used as counter electrodes in DSSC's(Dye Sensitized Solar Cells) enhancing its efficiency. Simultaneous reduction and composite formation happens in the sintering process. The plasma plum formation by the laser is something that needs to be explored more for reduction and functionalization purpose of the GO.

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