1. Introduction

The main focus of new age research is the achievement and preservation of clean environment. The world is facing numerous untold new environmental unpleasant situations every day which has lesser or greater impact on the topography. The swift technological development across-the-board building new window of opportunities for ordinary citizens is responsible to certain extent for this adverse impact. This is simply because financial profitability is at the heart of this crowd sourcing activities. Hasty industrialization is at the core of environmental devastation uttering a loud call for environmental protection[1]. The key component of any sustainable development is to cover climate change and environmental impacts and for that contribution from an individual, organization and from governmental echelon is very essential. As a part of this co-operation, research efforts should be made by chemical industries to develop and adopt a chemical process that minimize the waste with more number of recyclability which also trim downs monetary solvency.

In earlier times, when catalysts were not commercially available, industries had to depend on certain reaction stipulations, for instance, high pressure, temperature, concentration of reactant, increased time and additional chemicals to boost the production[2] which expend more capital and other sources. Hence, a need to develop such selective catalyst that increases the rate of reaction at diminished temperature and pressure was realized. As a result of this understanding, enzymes were used as a catalyst for the first time to produce wine[3]. The most antiquated catalyst used by Peregrin Philips[4] was platinum supported on Vanadia in the production of sulphuric acid. Now-a-days, as per report, larger than 90% of industries are using catalysts for the manufacture of countless precious products[5] and it is believed that this will continue to increase on a daily basis in the days to come. Though the existence of many catalysts, it is classified broadly into two category; homogenous and heterogeneous catalysts. However, homogeneous catalysts are experiences some perplexity such as stability only in mild conditions which make their separation from reaction mixture more difficult and expensive. Reusability of these homogeneous catalysts is thus limits to single use which in turn increases their production costs too. This anemic nature of homogeneous catalysts limits the use of sturdy reagents and ultimately its applications. It was these inherent limitations that lead to their heterogenization as the development of heterogeneous catalysts.

Easy controlling, ability to tolerate wide range of temperature and pressure, reusability, lesser degree of waste and adverse impact and improved innate process safety put them in a favorable position as compared to homogeneous one[6]. However, traditional heterogeneous catalysts are also suffering from low number of active sites which lessen the catalytic activity. To improve the catalytic activity of such heterogeneous catalysts, these active sites are supported on the inorganic inert solid support. A plenty of solid supports are available so far like silica, natural clays, heteropolyacids, cation exchange resin, molecular sieves, metal oxides and zeolites etc.[7]. These solid supports definitely surmount the number of difficulties cited above, yet they are incompetent due to deactivation[8], internal mass transfer[9], high mass transfer resistance and relatively high price. The quest of appropriate heterogeneous catalyst that surely overcomes the above stated intricacies was come to an end by the exploration of carbonaceous materials as solid supports[10].

The eminent materials like diamonds, fullerenes, carbon nanotubes (CNTs) and graphites were consider as the members of carbon family up to the end of the 20th century. Due to some key restriction of fullerene like toxicity, high price, etc., it was switched over with graphite and single layer graphene with ground breaking work of Brodie[11] and seminal report of Geim and Novoslow[12] in addition to the first isolation of 2D graphene by C. W. Bielawaski[13] in 2004. As a consequence, graphene and graphene oxide (GO) has gained much attention as a catalyst and catalytic support by research community[14]. Characteristically, graphene consist of a mono layer of carbon atoms has two dimensional (2D) stalked crystal structures with densely packed sp^2 carbon atoms hexagonal honeycomb shape lattice[15]. It also possesses the properties of both metals and non-metals in the form of thermal-electrical conductivity and inertness, high thermal resistance along with lubricity, respectively. On the other hand, oxidized graphene, GO is a layered structure with various oxygenated functional groups like hydroxyl, epoxy, carboxyl and carbonyl adorning its basal plane and edges. These oxidative debris make GO as peerless supporting material as they can be functionalized with numerous homogenous substances as active sites. A variety of heterogeneous catalysts can be formulated as per necessity by selectively modifying these functional groups with metals, non-metals, polymers, metal-organic frameworks (MOFs) etc. These stuffs can be grafted directly on to the oxidative debris or can be incorporated on to the surface of reduced GO (rGO).

Chemically modified graphene (CMG) possesses distinctive nanostructure with intriguing properties like high surface area[16] of ~2600 m^2/g, inherent low mass[17], elevated thermal stability up to 3000 °C[13], bounteous oxygen carrying functionalities[18], high tensile strength[18], exceptional mechanical strength[19], high mass transfer and greater resistivity in acidic and/or basic media and optical absorption[20]. Owing to all these characteristics, it attracted much attention of researchers and has been the cornerstone for various fields like sensors, energy-related materials, polymer composites, field-effect transistors (FET), as electrical conductors in alkaline batteries, as lubricants to shelter manufacturing tools, as compaction additives and paper like materials. It also becomes a host in lithium-ion batteries and biomedical applications etc.[21]

1.1. History of genesis

Ancientness of GO preserving an early stage of studies which involves archaic chemistry of graphite. It reveals the eminent potential and widespread interest of graphene as an essence of this exploration. In 1859, British chemist B. C. Brodie had investigated the reactivity of graphite flakes and explored its structure in order to come up with the molecular formula and molecular weight of graphite. For this intense, he made slurry of graphite in fuming nitric acid (HNO_3) followed by oxidized with “potash of chlorate” ($KClO_3$). As a result of this reaction, he observed excess mass of the resulting material composed of carbon, hydrogen and oxygen. He stated that successive four oxidative reactions enhance the oxygen content compare to graphite flakes. He proposed the net molecular formula of $C_{2.19}H_{0.80}O_{1.00}$ with C: H: O composition 61.04: 1.85: 37.11. He named this material “graphic acid” as it can be identified as dispersible in or basic water, but not in acidic media. At his best, he finally suggested molecular weight of this graphite to be 33 and termed as Graphon saying “this form of carbon should be characterized by a name marking it as a distinct element”.

L. Staudenmaier, nearly after 40 years of Brodie’s ground-breaking discovery to oxidize the graphite flakes, improved its $KClO_3$-fuming HNO_3 treatment by altering the introduction of chlorate as single dose with diverse aliquots together with concentrated sulphuric acid in order to increase the acidity of the mixture. This little alteration in the procedure resulted in analogous degree of oxidation (C:O ~ 2:1) to that of Brodie’s approach of multiple oxidations[22]. Hummers and Offeman, after 60 years of Staudenmaier, developed another different tactic for the oxidation of graphite. In this method, they employed a mixture of potassium permanganate ($KMnO_4$) and concentrated sulphuric acid (H_2SO_4) which offered similar degree of oxidation[23].

Other researchers have developed a new approach with a few modifications, but mentioned above three techniques were laid at its core. Hence, these three approaches were considered to be elementary route for the synthesis of GO. It has been examined that the yield of the reaction varied tremendously with oxidant used, source of graphite and reaction conditions. For instance, in Brodie and Staudenmaier methods of oxidation, they deal with $KClO_3$ and fuming HNO_3. Amongst them HNO_3 is potent oxidizing agent which vigorously oxidizes the aromatic carbon to carboxyls, lactones, and ketones[24], while $KClO_3$ was explosive used as an *in situ* source of dioxygen. For this treatment, reaction mechanism was also not known. On the other hand, to be acquainted with reaction mechanism, Hummers’ used admixture of $KMnO_4$ and Conc. H_2SO_4, so that reaction proceed through formation of an active species diamanganese heptoxide (Mn_2O_7) (Scheme1.1).

$$\mathbf{KMnO_4 + 3H_2SO_4 \longrightarrow K^+ + MnO_3^+ + H_3O^+ + 3HSO_4^-}$$

$$\mathbf{MnO_3^+ + MnO_4^- \longrightarrow Mn_2O_7}$$

Scheme 1.1. Formation of dimanganese heptoxide (Mn_2O_7) from $KMnO_4$ in the presence of strong acid (adapted from Ref. 25 with permission of American Chemical Society).

On heating an organic composites more than 55 ºC, this bimetallic heptoxide becomes more reactive[24,25] and selectively oxidize unsaturated aliphatic double bonds controlling reaction pathway and graphite structure[26]. Natural graphite flakes consisting of numerous single crystals are used for chemical reactions after obliteration of heteroatomic stains[17] and hence, particles are arranged arbitrarily in synthetic graphite creating a lot of defects in π-structure. As a result, oxidation process becomes very intricate. Since natural graphite is subjected to heat treatment, depending upon its source and ability to tolerate heat of natural graphite, many diversifications are seen in synthetic graphite. This heat is responsible for disorder in graphitic region and such graphite alters in crystallinity, electron mobility, surface area, thermal stability, purity, etc. The graphite produce by this method can be distinguished as primary and secondary synthetic graphites.

Sturdy heat treatment is used for the synthesis of primary synthetic graphite with controlled set-up to acquire consistent and static characteristics. PSTP characterization model were used to define the primary properties; where letter sign represents P: purity, S: structure crystallinity, T: texture, porosity and P: particle size[27]. The posterior properties were expressed as secondary properties. A range of spectroscopic methods such as inductively coupled plasma (ICP), emission spectroscopy and electrothermal vaporization combined with inductively coupled plasma (ETV/ICP) can be operated to investigate the degree of impurity. Degree of high electrical conductivity and lubricity in the form of crystallinity besides size of crystallite can be determined by XRD analysis classifying the graphite into macro-crystalline (>100 nm size) and micro-crystalline (<30 nm size). However, BET surface area employed to be familiar with pore-size distribution and gas adsorption indicates porous texture with characteristic aspects like true density, apparent and tap density. High purity of the product assessed by true density, whereas, apparent and tap density orients the porosity between the granules. To be aware of uniform distribution of particles, sieving method is commonly used.

1.2. Structural debate

Although a large number of methods are available for the oxidation of graphite, albeit exact chemical structure of GO is still matter of debate. Non-stoichiometric framework and the meager precise analytical techniques are the main cause of this structural vagueness. Regardless of these impediments, worth mentioning endeavors have been made intended to elucidate the precise chemical constitution of GO. An assorted group of researchers have been suggested diversified arrangement of elements in GO comprising multiple repetitious units as conversed below.

Hofmann and Holst proposed GO with sp^2 hybridized having only epoxy groups throughout the basal plane (Fig. 1.1) acquiring a net formula of C_2O[28]. Quite opposite to this, Ruess, in 1946, suggested dissimilar structure of GO with sp^3 hybridized carbons possessing hydroxyl groups on basal plane[29] with 1/4th cyclohexanes having epoxides at 1,3 position at 4th position of hydroxyl groups. Another research group Scholz and Boehm in 1969 had recommended the structure of GO with quinoidal species into a sequence of collateral ridge backbone instead of epoxide and ether groups[30]. Afterwards, Nakajima and Matsuo had been proposed a significant model of graphite based on a intercalation concept of ploy $(C_2F)_n$ compounds[31,32]. There are some analytical techniques viz. elemental analysis and X-ray diffraction studies lies at the core of these projections. Solid state Nuclear Magnetic Resonance (NMR) spectroscopy was recorded to characterize GO by Lerf and co-workers for the first time[33]. Lerf and Klinowski[34] by isolation of structural features of series of GO derivatives, proposed a model (Fig. 1.2) which was widely accepted and cited too.

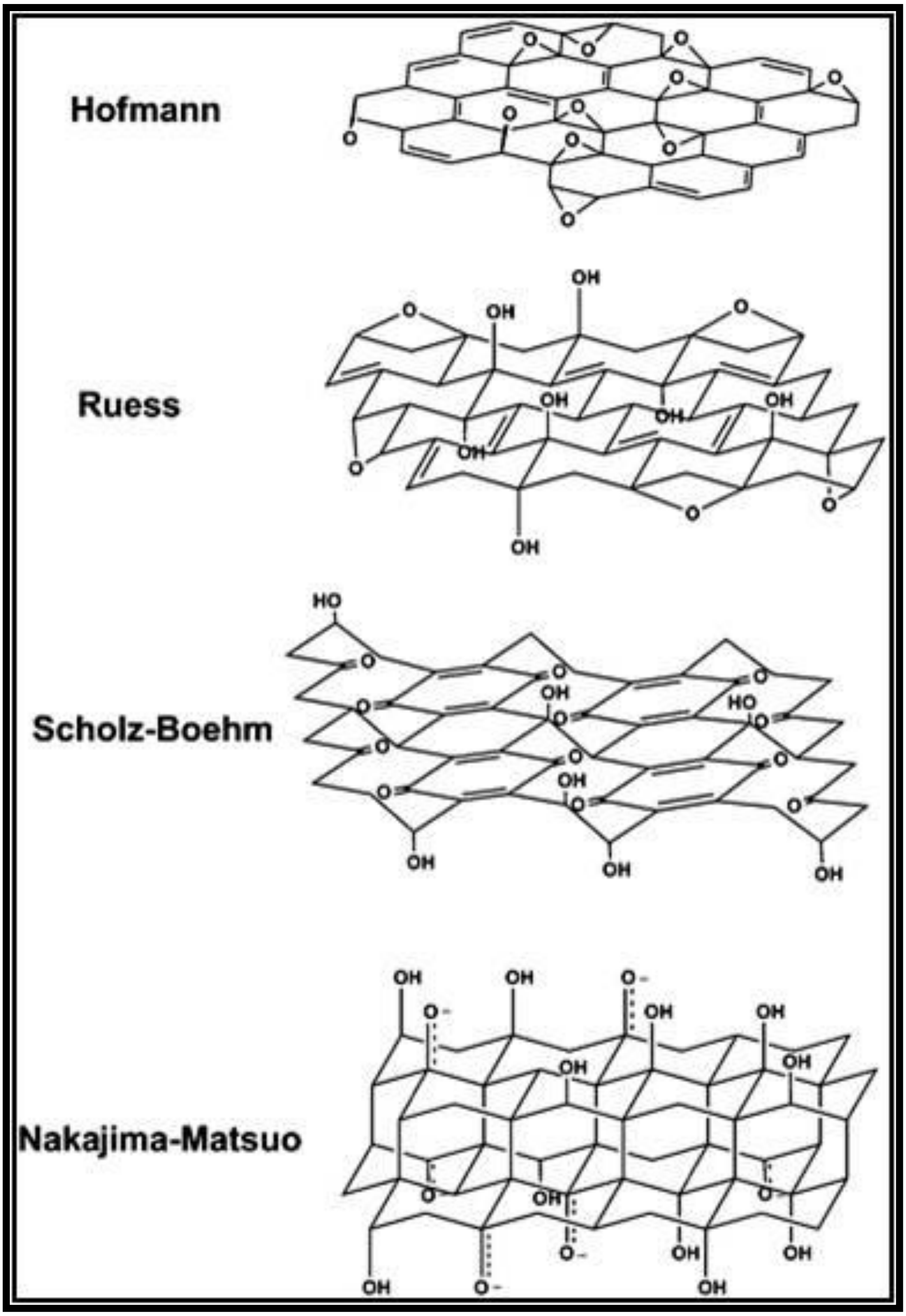

Fig. 1.1. A summary of some older structural models of GO (adapted from Ref. 42 with permission of American Chemical Society).

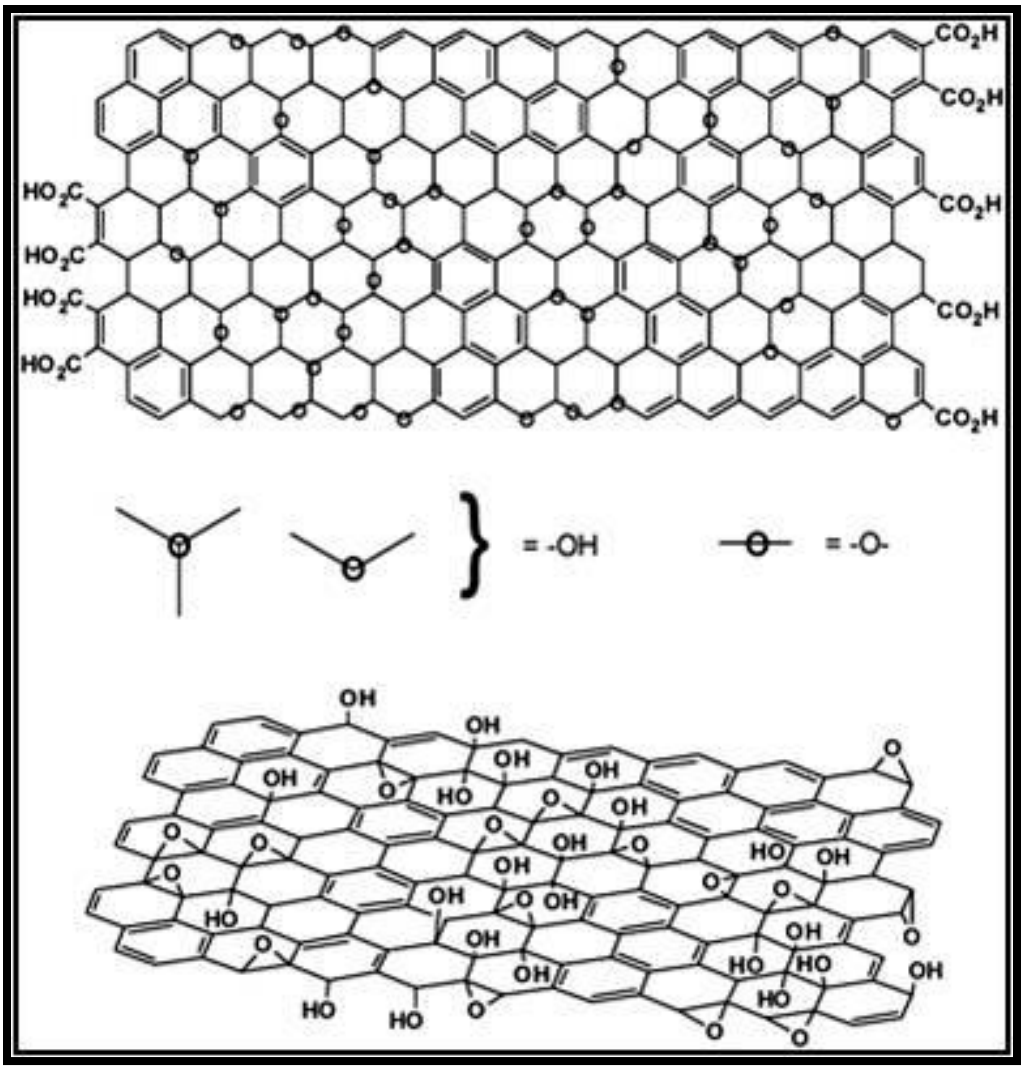

Fig. 1.2. Variations of the Lerf-Klinowski model indicating ambiguity regarding the presence or absence of carboxylic acids on the periphery of the basal plane of the graphitic platelets of GO (adapted from Ref. 34 (top) and Ref. 35 (bottom) with permission of American Chemical Society and Elsevier Science B.V.).

Carbon contests of GO can also be characterized by cross polarization/magic spinning (CP/MAS) experiment and short-contact-time spectra. The quaternary nature of all carbon atoms in GO can be identified by Mermoux's model[36] which allocated the peaks at 60, 70 and 130 ppm to tertiary alcohols, epoxy and mixture of alkenes, respectively. Inter-platelet H-bonding in GO through alcohols and epoxide groups with H_2O (Fig. 1.3) was corroborated by short-contact-time experimentation and neutron scattering study[37], which confirms the stacked layered structure of GO.

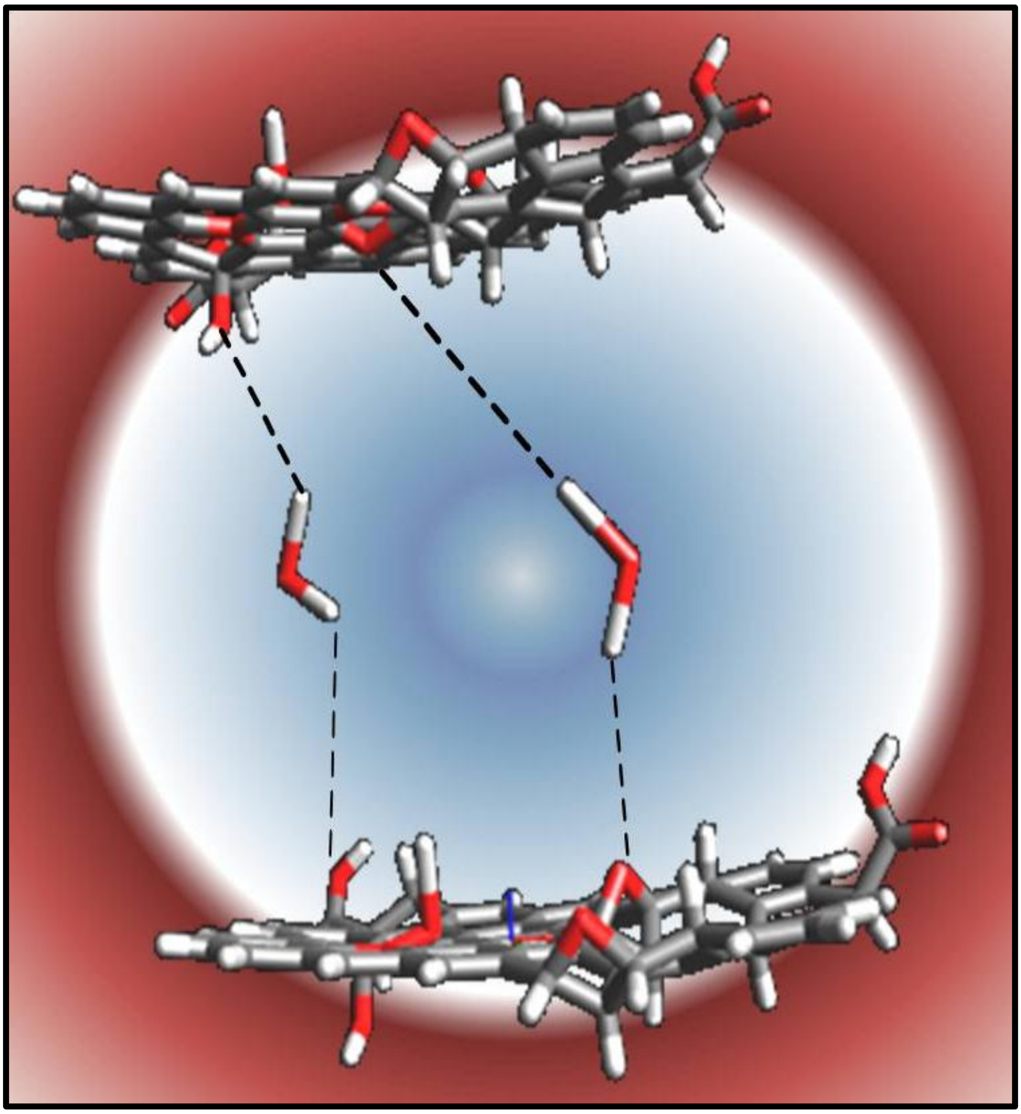

Fig. 1.3. Hydrogen bonding between oxygen functionalities of GO and water (adapted from Ref. 38 with permission of Nova Science Publishers)

These findings has resulted in identification of all the functional groups presented in GO, however, the specific site of these functional groups was remain unfocused. Lerf and co-workers[34] tried to resolve this problem by estimating and measuring the similarity and/ or dissimilarity of ^{1}H and ^{13}C NMR spectra of maleic anhydride treated GO with pristine GO. The results suggested the absence of aliphatic alkenes in GO as both the spectra of maleic anhydride treated GO were analogous to pristine GO. If true, these alkenes should promptly get reacted with maleic anhydride but no such reaction was seen.

In the ^{1}H NMR spectrum of D_2O tainted GO exhibited two characteristic peaks at $\delta = 1.3$ ppm and $\delta = 1.0$ ppm. Amongst these, first peak corroborates the existence of tertiary alcohol on GO surface; while another peak was associated with magnetically uneven more than one alcohol species. However, these species were not recognized accurately as stated which raises the question on its veracity. Now the question arise that why these peaks was reflected in ^{1}H NMR spectra? The reason behind this was might be strong H-bonding with water between platelets. In order to authenticate the existence of epoxide groups on the surface of GO, it was allowed to react with ether (sodium ethoxide). It was examined that in this reaction, epoxide groups act as electrophilic centres for surface functionalization[39].

All the information available so far cannot provide crystal clear structure of GO. The keenness to get more clear structure, encouraged Lerf and his research group to inspect the reactivity of GO with other a choice of reactive groups so as to seek out existence of other functional groups on GO[34]. They observed that the double bond present on GO was capitulated in harsh conditions and hence they thought these double bonds either to be aromatic or conjugated. This gave accordance to their faith of existence of another functional groups on the surface of GO. Deconvolution of IR data recorded before a decade substantiates the presence of carboxylic acid groups at the edges but in small extent[40]. With the help of Brodie's observation regarding highly reactive surface of GO, they noticed the evolution of CO and CO_2 instead of O_2 during thermal decomposition. This resulted in extremely chaotic blending of graphitic carbon with oxygen which is intricate to characterize.

Exposure of probability to isotopically labelled GO by Cai and co-workers makes the study of GO more simple enhancing the horizon of spectroscopic techniques[41]. However, Ruess and Scholz–Boehm model suggested the regular quinoidal system arranged into every second ridges and grooves interceded by trans-linked cyclohexyl domains functionalized by tertiary alcohols and 1,3-ethers. In the IR spectrum of GO, the band exhibited at 1714 cm^{-1} indicated the presence of ketons and/or quinines rather than carboxylic acid which was validated by DRIFT study and reassessment of the FTIR spectrum. Contrarily, the presence of acidic site on the basal plane of

GO was confirmed by potentiometric acid–base titration[42]. This inconsistency can be explained by keto-enol isomerization of *in situ* generated α, β-unsaturated ketones in Lerf - Klinowski model. Amongst this, if enols exist in the aromatic region, they promote the proton exchange as they generate the thermodynamic product, phenoxide. On the other hand, keto forms do not found to be favorable for enolization and acidic proton exchange.

Aforementioned explication was endorsed by Dekany model (Fig. 1.4) comprises keto/quinoidal groups arranged into network shaped every second ridges and grooves and trans-linked cyclohexyl groups which scattered between tertiary alcohols and 1,3-ethers. Both these model are clear enough to distinguish. That is, carboxylic acid groups are thought to be absent in it. The aromatic sac conserved during oxidation would be subjected to oxidize further and alkenes of quinones get converted to 1,2 ethers. The presence of quinones was concluded by the macroscopic wrinkles found in TEM images. Besides this, rigidity and boundaries of the platelets were also thought to be due to quinone's role[42].

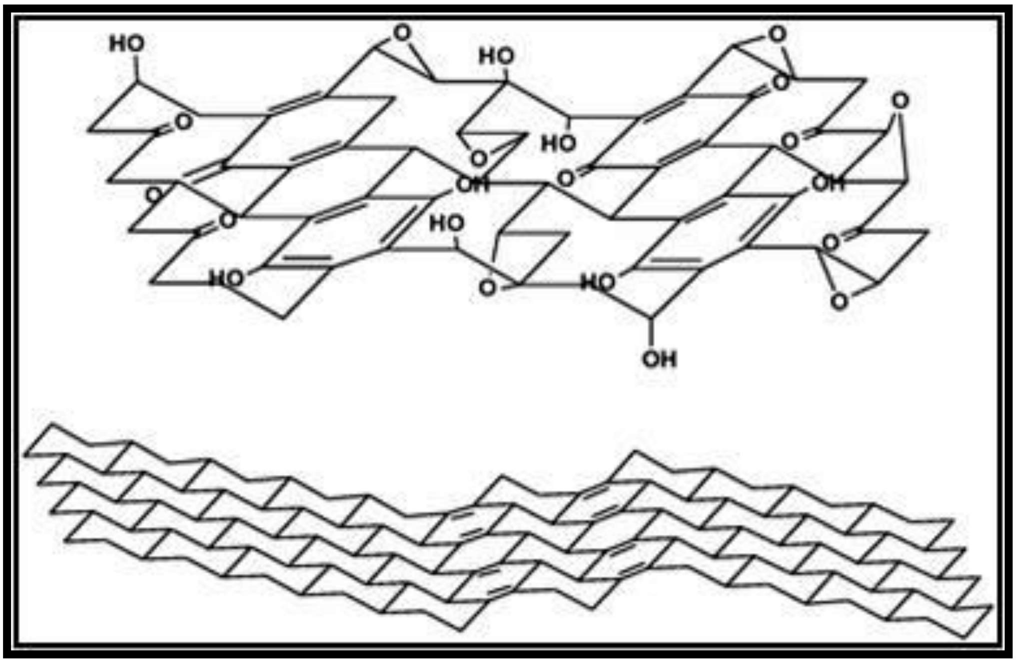

Fig. 1.4. Structure of GO proposed by Dekany and coworkers (adapted from Ref. 43 with permission of American Chemical Society).

It can be concluded from the discussion so far, graphite possesses various structures with diverse properties and the oxides formed are vary with source of “pristine graphite” and/or modus operandi use for oxidation. DFT calculation was performed against the experimentally obtained observations and it can be predicted that partial oxidation is thermodynamically more favored[44]. The possibility of conversion of epoxides to alcohols is increased with enhanced oxidation and it is envisioned theoretically. Hence, it can be said that specific conversion depends on the sharing of precise oxidative functional groups over the platelet[44].

1.3. Reduced graphene oxide (rGO) and synthesis strategy

“Bottom up” and ‘Top down” are two approaches for the synthesis of graphene that arouse much curiosity and hold the attention of the entire research community. Amongst these, “bottom up” techniques comprises organic synthesis[45], chemical vapour-deposition (CVD)[46], epitaxial growth on SiC[47], and solvothermal reaction[48] employed for the production of graphene. Although this technique offers high-quality of pristine graphene, it is limited to small scale production. Whereas “top down” approach hold various methods such as mechanical exfoliations (scotch tape method)[46], liquid-phase exfoliation[49], thermal reduction of GO[50], photothermal reduction of GO[51], electrochemical reduction of GO[52] and chemical reduction of GO[53] affording high yield, solution-based process ability and painless implementation.

The extensive technique used for large-scale fabrication with high potential is chemical reduction of GO. With the implementation of this technique along with reducing agents, the electronic properties that were disintegrated have been reinstated, but all the functional groups and several defects cannot be removed completely (Fig. 1.5). A huge range of reducing agents are available, for instance, hydrazine monohydrate[54], sodium borohydride[55], hydroquinone[56], strong alkalies[15], sulphur-containing compounds[57], etc. Hydrazine monohydrate (N_2H_4) is universally employed reducing agent having potential to reduce more number of oxygen containing functional groups. However, including hydrazine monohydrate all these reducing agents are found to be noxious and detrimental to ambiance and biological systems as well. As a remedy, l-ascorbic acid[58], amino acid[5], reducing sugar[60], alcohols[61], hydrophilic acids[62] etc. have been exercised in these days for the simple and green bulk synthesis of graphene. These environmentally benign reducing agents are gentle, cheaper and accommodate atypical ion environment.

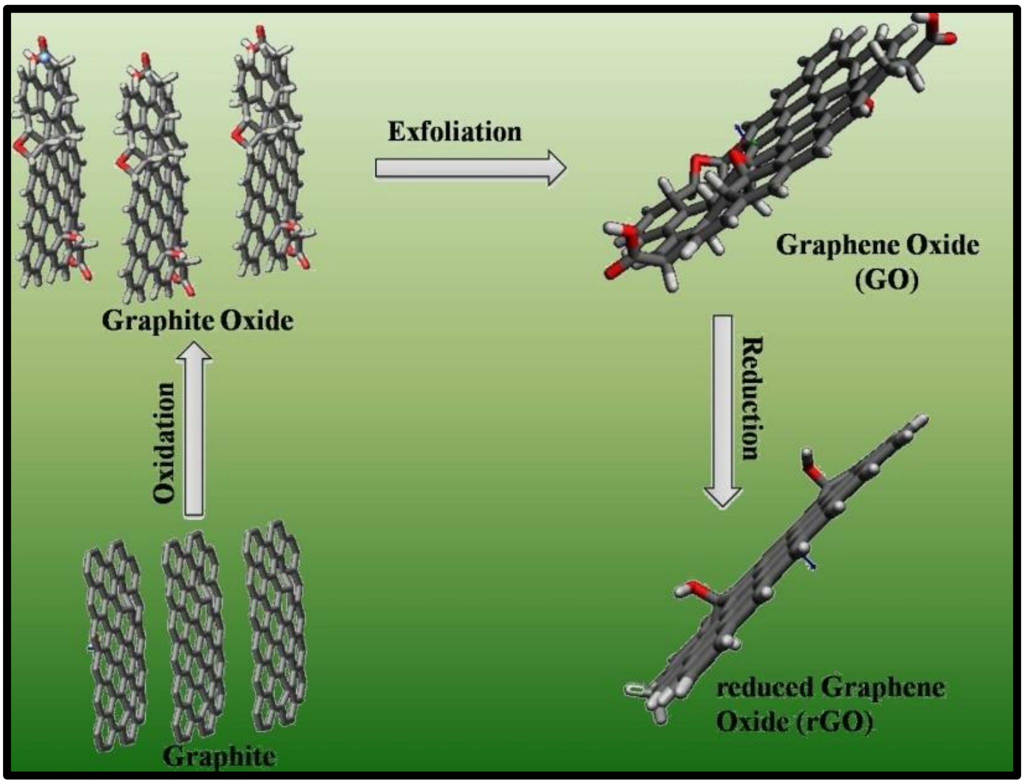

Fig. 1.5. Preparative steps of reduced graphene oxide (rGO) (adapted from Ref. 38 with permission of Nova Science Publishers)

1.4. Chemical reactivity of GO

It is very well known that water molecules are intercalated between the layers of GO and as a result GO preserves the spacing of ~6-12 Å but hold opposing views in humidity[63]. Graphite oxide and GO both are chemically similar but structurally quite different; however, it is promising to distinguish them. Graphite oxide holding stacked layered structure while hydrophilic surface functionalities in GO causes very weak platelet–platelet interactions and hence prone to differ with exfoliated single layered or few-layered structure.

Amongst all the available methods, aqueous sonication of graphite oxide is the most adoptable method. It is faster than other method and has its drawback also that creates an adverse situation. There is considerable destruction to GO platelet ensued[46] which trivialize the average size from micrometer to nanometer. Likewise, oxidation route destructs the GO structure in small extent not in nanometer size[64]. Exfoliation rely on dispersion of GO which depends on the nature of solvent and extent of functionalization. More polar surface enhances the dispersibility of GO. Exfoliation of rGO synthesized by ultra-sonication was validated by Becerril et al.[65] using AFM technique. For researchers, center of attention is to cast a method for up-scale production of GO and hence they are unremittingly focusing on advancement of synthesis technique. The presence of more numbers of oxygenated functional groups onto the surface of GO, makes its substitution more easily through covalent or non-covalent attachment. One can make the GO more advanced by two ways; through modification of available oxidative functional groups or via reduction of these functional groups. Both these ways are discussed here.

1.5. Modification of oxidative functional groups

The chemical reactivity of these oxidative functional groups makes the attachment of numerous groups more promising through addition reactions, nucleophilic and electrophilic substitutions reactions. One can modify more than one oxidative functional group at a time; however, selective modification of precise functional group is carrying great significance. At the present time, available modern analytical techniques make the expression of the selective modification more clear and uncomplicated.

1.5.1. Selective modification of epoxy groups

Under the diverse reaction conditions, it is likely to cleave the ring structure of epoxy groups. Ring opening by nucleophilic attack at α-carbon on reaction with substituted amines is one of the more precise tactics. During this reaction, carboxylic acid group may affect alongside. Differentiation of both these reactions is quite important. Numerous researchers have employed various compounds for ring opening of epoxides. Wang and co-workers[66] besides Y. Xue et al.[67] have used octadecylamine. However, H. Yang et al.[68] utilized ionic liquid 1-(3-aminopropyl)-3-methylimidazolium bromide (IL-

NH_2) for epoxide ring opening reaction using hydrochloric acid as an acid catalyst in presence of KOH (10 mg). Being polar, this substance is extremely dispersible in solvents like water, DMF and DMSO. H. Yang and his research group[68] have attempted 3-aminopropyltriethoxysilane (APTS) with *N*,*N′*-dicyclohexyl-carbodiimide (DCC) for the selective modification of epoxide groups of GO. For the successful functionalization, 10 mg of GO and 5 mg of DCC was dispersed in 10 mL of APTS using ultra-sonication and refluxed at 70 °C for 24 h. The resulting mixture was easily dispersible in water, DMF, DMSO and APTS, but its dispersibility is high in APTS due to presence of silane moieties. Form the discussion so far, it can be concluded that the selective modification of epoxy group required a catalyst.

1.5.2. Selective modification of carboxylic acid groups

Carboxylic acid groups also adorn the edges of GO besides the chemically reactive epoxy groups at the basal plane. This reaction is little different than direct functionalization of epoxy groups. This selective modification requires the activation of carboxylic acid groups using diverse activating agents such as thionyl chloride ($SOCl_2$)[69], 1-ethyl-3-(3-dimethylaminopropyl)-carbodiimide (EDC)[70], N,N′-dicyclohexyl-carbodiimide (DCC)[71] and 2-(7-aza-1H-benzotriazole-1-yl)-1,1,3,3-tetramethyluronium hexafluorophosphate (HATU)[72].

Various methods have been developed using these reagents together with nucleophilic reagents like amines or hydroxyls which produces amide or esters via covalent attachment. In order to activate the carboxylic acid groups through 1-ethyl-3-(3-dimethylaminopropyl)-carbodiimide, diamine was also projected to embedded the quality, performance and reliability to the same. These amine groups are specific and varied with the reagent used for the activation of carboxylic acid groups. For example, polyvinyl alcohol (PVA) is used to modify the surface of GO if carbodiimide employed for the activation of carboxylic acid groups[71]. However, aliphatic diamine is used if carboxylic acid groups activated by EDC.

Dispersibility of GO in many polar aprotic solvents can be increased by eliminating the hydrophilic groups such as –OH groups as it forms strong H-bond, not in favour of dispersion in non-aqueous solvents. It can also be modified simultaneously with –COOH groups. Some researchers provided many examples, for instance, S. H. Lee with his co-workers[73] attempted to modify terminal amine and hydroxyl groups at the same time with atom transfer radical polymerization (ATRP) initiator, (a-bromoisobutyrylbromide). S. Stankovich and co-workers[74] also attempted modification of carboxylic acid groups together with hydroxyl groups using aryl and alkyl isocyanates and increased the dispersion of GO. They espouse new route and converted the carboxylic acid groups to other groups rather activating with substituted amide or ester. They made homogeneous solution by mixing 50 mg of GO in 5 mL of DMF followed by addition of 2 mmol of isocyanate. It was then allowed to stir for 24 h under nitrogen atmosphere. Final product was collected and washed with methylene chloride. By this reaction almost all hydroxyl and carboxylic acid groups can be removed and as a result this product shows complete dispersion.

1.5.3. Selective modification of hydroxyl groups

Hydroxyl groups of GO are responsible for hydrogen bonding between platelets which makes GO more hydrophilic. However, reduction in the number of these groups lessens the hydrophilicity which resulted in weakening of interlayer hydrogen bonding. This increases the dispersability of GO and make more favorable for the fabrication of ample of composite materials. Recently, S. Zhou et al.[75], endeavored the functionalization of hydroxyl groups by (3-Mercaptopropyl)trimethoxysilane (MPS) using hydrothermal reduction. Z. Li et al.[76] modified the surface hydroxyl groups using 3-aminopropyltriethoxysilane (APTES). As-prepared amino functionalized GO nanosheet was then reacted with CM–Salen H_2 metal complexes to fabricate GO supported heterogeneous catalysts. However, Q. Zhao and his co-workers[77] have employed APTMS to covalently functionalize the surface hydroxyl groups of GO followed by grafting with salicylaldehyde and copper complex forming tetrahedral chelate [Cu(salen)-*f*-GO]. A. Kumar et al.[78] have synthesized GO@AF-SB-Cu by tethering of Cu(II) Schiff base complex onto the amino functionalized GO sheet and inspected over Chan-Lam coupling reaction of amines and phenylboronic acid.

Herein, we have functionalized hydroxyl and carboxylic acid groups of GO one by one using APTMS and $SOCl_2$, respectively. Our aim for these modifications is to fabricate the heterogeneous catalysts using GO as a solid inert support. For this purpose, we have grafted 2-hydroxyacetophenone (Schiff base) onto–OH functionalized GO nanosheet followed by grafting of varying metals viz. metal salts, for instance, $Cu(NO_3)_2•6H_2O$, $VOSO_4•2H_2O$ and $Co(NO_3)_2•6H_2O$. However, for –COOH functionalized GO nanosheet, N,N'-bis(4-hydroxysalicylaldehyde)ethylenediamine (salen) and diverse metal-salen complexes, i.e. Cu-salen, VO-salen and Co-salen were prepared using metal salts like $Cu(NO_3)_2•6H_2O$, $VOSO_4•2H_2O$ and $Co(NO_3)_2•6H_2O$. These metal-salen complexes were then allowed to graft onto –COOH functionalized GO nanosheet as described in chapter 2. The successful fabrication of aforementioned solid heterogeneous catalysts was corroborated by various physico-chemical techniques as described in chapter 3. The catalytic aptitude of the as-synthesized catalysts was weighed against a variety of oxidation such as selective oxidation of benzyl alcohol, epoxidation of styrene, epoxidation of norbornene and oxidation of glucose by optimizing various parameters like varying mole ratio of substrate to oxidant, catalyst amount, diverse solvents, solvent amount, oxidants, time, temperatures as described in chapters 4 and 5. Finally, we have compared the proficiency of all the heterogeneous catalysts to give best suited oxidative site for the fabrication of GO supported heterogeneous catalyst for above mentioned oxidation reactions (described in chapter 6).

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