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Graphene and Its Derivatives: Properties, Synthesis, and Applications in Wearable and Transparent Sensors

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Abstract - **Graphene is a 2-D carbon-based material consisting of a single layer or few layers that has revolutionized the scientific world with its potential applications in various types of sensors, especially wearable and transparent devices. This material possesses astonishing thermal, electrical/electronic, optical, and excellent mechanical properties. It has the potential to replace carbon nanotubes (CNTs) due to its low cost and relatively similar properties. There is a need to work on different graphene synthesis methods, optimize various parameters, reduce costs, and achieve large-scale production. This study discusses in detail graphene, graphene oxide, reduced graphene oxide, their properties, various synthesis methods, and potential applications. Among these methods, we have found that Hummer's method is relatively better for producing low-cost graphene on an industrial scale. However, there is a need to optimize Hummer's method parameters, such as finding the right chemical to reduce graphene oxide and synthesizing few layers of graphene with high conductivity.**
Keywords: **Graphene, Graphite Oxide, CNT, Carbon, Sensors**

I. INTRODUCTION

The term "graphene" is derived from combining "graphite" with the suffix "-ene." Graphene refers to a flat monolayer of carbon atoms arranged in a tightly packed two-dimensional (2D) honeycomb lattice. It serves as the fundamental building block for graphitic materials of various dimensionalities. It consists of carbon atoms having 4 valence electrons and each carbon atom is bonded to three other carbon atoms by σ bonds forming a hexagonal close packed structure, leaving one free valence electron. The free electrons are also called pi bonding electrons or simply π electrons because each carbon atom forms π bond with the adjacent carbon atom through these free valence electrons, which helps to strengthen the σ bonds. Thus forming a hexagonal close packed sp^2 hybridized structure i.e. 2s and 2p orbital overlap with each other. Graphene is a light material due to stacking of carbon atoms in hexagonal form making it transparent with unexpected electrical and other properties, and even stronger than steel [1-2]. Graphene has potential to replace carbon nano tubes due to its low cost and almost similar properties as it is a single or few layers of carbon atoms. These days it is also modified by using various functional groups [3-4].

Graphene is relatively a new concept for the large family of material scientists with potential application in mechanical, electronics, medical and related field of sensor technology. This study is a small step in the direction of collecting data related to properties, application and synthesis techniques. Three million graphene sheets is just equal to the tip of the pencil this material will offer huge contribution in the field of dimensional physics [5][6].

II. GRAPHENE PROPERTIES

Carbon element has mainly three allotropes namely: Graphite, Diamond, and Charcoal, all having diverse chemical, electrical, physical and other properties. Amongst these single completely exfoliated graphite layer viz. graphene shows exceptionally different properties in the field of material science. It shows outstanding electrical, thermal, mechanical and optical properties as per detail below.

Graphene is rightly considered as an exciting material mainly because of its conducting property as the electrons move relatively easier way then most of the conducting metals such as copper. Speed of electrons through graphene may be one hundredth the speed of light if they carry no mass. The electron movement behavior is an interesting study conducted by Klein tunneling and quantum Hall effect by restricting their movement in two dimensions. Because of zero band gap and high charge carrier mobility of 10,000 $cm^2/v^{-1}s^{-1}$, this material shows exceptionally high electrical conductivity. It is also noticed in various experimental studies that in comparison to chemically reduced graphene oxide thermally reduced shows higher electrical conductivity because of low number of oxygen atoms as functional groups. Its application as a filler in various materials embellished their conducting property [7-9].

Application of this material is further improved due to its transparent nature. Transparent electrical and even thermal conductors have various applications in diverse fields including flexible wearable sensors. At room temperature it shows thermal conductivity of 3,000 Wm^{-1} K^{-1}. This property is mainly noticed due to the lattice vibrations observed in graphene, and thus results in thermally conducting and thermally stable behavior. Apart from this they are mechanical stronger (Modulus of 125GPa, 200 times better than steel), shows stretching properties ~ 30 %, and addition of 0.7 % can raise tensile strength by 76 % and Young's modulus by 62%. Vandar wall and hydrogen bond interaction are mainly responsible for their mechanical properties. Graphene layers with size lesser than 20nm (or upto 6000 atoms) is thermodynamically unstable and most stable in the form of fullerene for larger than 24,000 atoms.

Optically it is transparent thinnest material, however, due to its unique property of absorbing 2.3% of light passing through it we can see it in air even by naked eye. We can control its visibility color using the similar concept of rainbow and oil film. In this concept the color visibility is controlled by the wavelength and path length through which light is traveling. This can by controlled by using the deposition of layers of graphite flakes on material like silicon-dioxide. Light will be transmitted and reflected from different layers in different proportions results in control of its optical properties.

Chemically, graphene layers have ability of absorbing and desorbing different molecules such as NO_2/NH_3/OH, which further can act as donors/ acceptors. Thus, the change in their concentration will result in variation in its conducting properties. This is a remarkable property for future research in testing this material as sensors for chemicals. Number and type of chemical groups, monolayer graphene, or higher numbers of graphene layers can be studied in future by the researchers [10-12].

III. APPLICATIONS OF GRAPHENE

Graphene has garnered significant interest due to its exceptional mechanical, electrical, thermal, and optical properties. However, cost-effective and large-scale production of graphene remains a challenge. As a stable two-dimensional (2D) crystal discovered on Earth, graphene possesses a unique electronic structure and remains stable under ambient conditions. Due to amazing properties as the strongest and longest material and it has the conductivity, heat and electricity better than other nano based materials, so due to outstanding properties of graphene it is used in huge number of electronics and other applications. Initially, graphene will be used to enhance the performance and efficiency of existing materials and substances. In the future, it will be developed alongside other two-dimensional (2D) crystals to create even more remarkable compounds for a broader range of applications. To grasp the potential applications of graphene, it is essential to first understand its fundamental properties. One significant challenge with graphene is opening a band gap. High-quality graphene is an excellent conductor, but without a band gap, it cannot function as a switch, as the absence of a band gap prevents it from being turned off. Therefore, graphene is used in various future nano-electronic devices, but a band gap will need to be considered into it, because by reducing the electron mobility the conductivity of the materials can be controlled and used in silicon films. In the future graphene act as a wonder to replace the silicon based devices in electrical and electronics devices and vastly used for future research and development. The present study has revealed that dispersing a small amount of graphene in polymers can significantly improve various properties of the composites, including tensile strength, elastic modulus, electrical conductivity, and thermal conductivity. These enhancements hold great potential, making graphene a preferable alternative to conventional nano-fillers for applications in structural and functional materials. Potential uses include lightweight gasoline tanks, plastic containers, more fuel-efficient aircraft and car parts, stronger wind turbines, medical implants, LEDs, coatings for solar cell displays, and sports equipment. Graphene is a material that can be utilized in numerous applications like bioengineering, energy technology, composite materials and nanotechnology [13].

Graphene has potential application in solar cell which converts light to electricity. The amount of absorbed photons converted to current explains the photocurrent efficiency. The light conversion efficiency is $\eta = P_{max}/P_{in}$, where $P_{max} = FF \times V_{OC} \times I_{SC}$ and P_{in} is the incident power. I_{SC} is the maximum short-circuit current, V_{OC} is the maximum open-circuit voltage and FF is the fill factor, which is equal to $FF = (I_{max} \times V_{max})/(I_{SC} \times V_{OC})$, where V_{max} and I_{max} are the maximum voltage and current, respectively. The fraction of absorbed photons converted to current defines the internal photocurrent efficiency [13]. Solar cells for light absorption and charge transport depends on liquid electrolyte. This type of solar cells consists of a TiO_2, dye molecules and nanocrystalline photo anode. When light is incident then dye molecular capture the light, and generates electron hole pairs and electron are transfer from valance band to conduction band and generates large amount of current. Then ITO material use for both as a photo cathode and anode with a platinum coating, but ITO is brittle and makes solar cells expensive [14]. Subsequently, researchers aim to develop a new solar cell made from graphene and molybdenum disulfide. This solar cell is expected to be up to 1,000 times more efficient than silicon-based panels, as well as thin, flexible, and lightweight, with a conversion efficiency of up to 70%. The graphene-based organic cells are bending up to 5.2mm [14].

Graphene is used as a preliminary material for gas sensing applications for sensing gases like NH_3, N_2 gas etc. Since graphene is intrinsically insensitive, so it can not alone be used for gas sensing. For that we prepare the composites of graphene with polymers, which will add functional groups in the graphene's structure. These groups absorb the gas molecules introducing a change in the electrical resistance of the material. Graphene can sense a gas even at a nanoscale level. Graphene can also be used as body sensors to measure the heart-beat, breathing rate etc. The main principle of graphene based bio sensors and gas sensors depends on the graphene's conductivity due to absorption of molecules on graphene layer surface [15]. The change in conductivity is change the concentration of graphene layer carriers. Graphene has some properties which increase the sensitivity of material up to one atom. First, graphene has high conductivity and low thermal noise. Second, graphene has less structural defects. Firstly, in 2007, graphene revealed outstanding sensing properties near NO_2, NH_3, H_2O, CO. Then NO_2, NH_3 and dinitrotoulene compound was also detected [16]. The sensing principle of NO_2 was assigned to hole induced conduction which withdraws electron from graphene layer and NH_3 was assigned to electron induced conduction. Graphene based sensors also used to detect or sense high metal ions. Nafion graphene was used for the detection of cadmium [17]. Lithographic is the technique which is used for the fabrication of graphene but it because negative effects on sensing properties of graphene layer due to presence of residual polymers [18]. The main problem is to sense hydrogen gas but graphene has high temperature i.e. 2700°C, so easy to sense hydrogen gas by using graphene layer in sensing devices.

There are large numbers of sensor which are used in bio. Graphene based electrochemical DNA is the better choice for the detection of selected DNA metamorphose gens related with human disease because biosensors offer low cost and high sensitivity for the detection of selected DNA metamorphose gens related with human disease, and provide a simple, accurate and economical stage of patient diagnosis. Graphene has high electro catalytic properties of graphene near H_2O_2 and outstanding performance for electrochemistry action of glucose oxides, so it is used in enzyme based graphene sensors. Carbon nano-tubes are not successfully detecting serotonin and dopamine so we use graphene based sensors [19].

Graphene is used to replace silicon in many of the electronic devices like in transistors, conducting electrodes, frequency multipliers, optoelectronic devices etc. Graphene is an atomically thin crystalline film formed by sp^2- hybridized carbon atoms. It has a unique electronic structure and electronics properties like high conductivity which make it differ from solid-state electronics which are traditionally employed. Graphene is a basically a semimetal or semiconductor with a no band, overlap electronic states linear density and linear energy dispersion. The conduction and valence bands form conic shapes (called as "Dirac cones") in the low energy range related to the Fermi level, and these conic shapes meet each other at point so called Dirac points. One of the outstanding property of graphene is a strong electric field effect which tends to a carrier density which is electrostatically tuneable in the range of n is less than 1014 cm^{-2}. Together with high carrier motilities for both electrons and holes (as high as 104 cm^2/Vs at room temperature), this attracts a lot of attention to graphene as a possible material for a future high-speed field effect transistor (FET) [19-20].

Graphene can also be widely used in solar cells, photovoltaic cells etc because of its property of having high electrical conductivity and good optical transparency. It is also used in fuel cells as it enhances its durability and reduces its efficiency. Since graphene has a high surface area to mass ratio, it can be used in conducting plates of super capacitors.

IV. SYNTHESIS METHODS

Up to now, we have studied the properties and applications of graphene. Now we must know how to synthesize it. Graphene can be synthesized by various techniques. Pure graphene cannot be synthesized by any of the techniques and therefore we obtain only rGO i.e. there is always some content of oxygen present in it. Before proceedings towards methods of synthesis of graphene we need to understand that there are basically two approaches namely top down and bottom up. In the first approach larger material is break down and in the latter approach small building blocks are used to get the desired material. Therefore, graphite is used in top down approach and break down into few layers of graphene using chemical, mechanical and thermal techniques. In the bottom up approach there is layer by layer deposition of carbon or graphene on noble/transition metals such as cobalt or also on thin SiC wafers. The techniques used for these two approaches is discussed as below.

A. Micromechanical Exfoliation

This method is also called a *Scotch Tape* method. It had first been per- formed by Geim and Novoslev and they won *Nobel Prize* for their work in 2004. In this method, graphite in the bulk form is placed on a scotch tape. The tape is peeled off and we get graphene in microns. But we do not get a thin layer of graphene through this method and is not that useful, as it is very difficult to get a single layer graphene with a large surface area by this method. However, this method helps to provide researchers with pure and clean graphene which further can be used to measure the various mechanical and electrical properties. There are many draw backs of this method such as finding right kind of flakes, surface region and exact surface position to peel off the desired layer for required application of the like of solar cell in which large surface area needs to be covered [20].

The few layer of graphene was identified using an optical microscope and single layers using an SEM. By using this method, Geim and Novoslev were capable to produce single and few layer graphene flakes with dimensions of 10 μm [20]. The few layer of graphene was found to high mobilities like as high as 15000 $cm^2V^{-1}s^{-1}$ and have ballistic transport at room temperature.

With the scotch tape method, we can generate graphite flakes sides of 1mm in length [21], which have an outstanding quality and much suitable for fundamental research. This process is useful only for small scale production not for large industrial production [22-32].

B. Epitaxial Growth on Metal Substrates

For making the disadvantage of Scotch tape method to be advantageous, we use the exfoliated graphene synthesized by this method. We form a suspension of graphene obtained from Scotch Tape method in an aqueous solution. This solution is filtered by a filter paper of grain size 25 microns. Graphene is obtained from the filter paper and it is dried in an oven. This method makes the graphene which is perfect, but this method is not that useful, as it is very difficult to get a single layer graphene with a large surface area by this method.

C. Chemical Growth Method

Exfoliation method generates a very pure and perfect single field graphene with approximately ideal thermal, electrical and mechanical properties; it has one of the biggest disadvantages. That is in the exfoliation method resultant graphene which is generated the flakes of graphene are randomly scattered on a substrate. Every flake of graphene has size in the order of only microns and much of the substrate remain sun covered.

A continuous layer of graphene is required for many applications discussed earlier, including transparent conducting electrodes for organic solar cells. To produce adjoining graphene films, exfoliation method cannot be usable and for large scale growth we use chemical methods instead to generate graphene from carbonators.

In the hummer method firstly we generate graphene oxide then generate reduced graphene oxide. Hummer method is most economical method and top down method. Common methods to generate graphene at large scale by chemically growth of graphene include hummer method in which reduced graphene oxides are generated through different reducing agents like hydrazine and sodium borohydrate etc. The hummer method technique by which reduced graphene oxides is formulated is the intersection of exfoliation and chemical growth methods. The graphene flakes which are produced through exfoliation method are oxidized, sanctioning them to be dangled in aqueous solution. After that solution is passed across a membrane act like a filter and which pores around 25 nanometers. Until the whole surface of filter is concealed with graphene sheets, the graphene oxide flakes get captured by the membrane. The end is the formation of graphene oxide instead of graphene and further chemical treatment of graphene oxide films is required to make them electrically conducting from insulating.

D. Chemical Vapour Deposition

Under the bottom up approach chemical vapour deposition (CVD) is quiet reliable and scalable synthesis method of large area graphene (Fig. 1). Among synthesis techniques, chemical vapor deposition (CVD) has proven promising for developing high-quality, high-purity, and fine-grained graphene. Analysis of carbon atom diffusion reveals that higher temperatures within the Ni film accelerate the diffusion process. The CVD process is typically conducted under atmospheric pressure and at relatively lower temperatures. (Fig. 2).

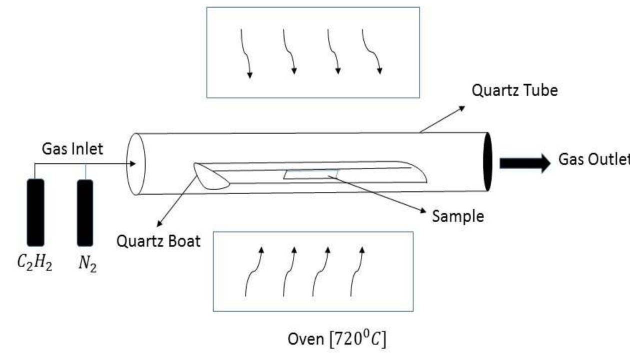

Fig. 1 Chemical Vapour Deposition setup [23]

In chemical vapour deposition technique, a metal substrate is heated in a furnace at 10000°Celsius temperature and at low pressure conditions. Due to heating or annealing, the domain size of the substrate becomes large. After a certain time of heating, methane and hydrogen gases are passed through the furnace. Methane reacts with the metal substrate and hydrogen acts as a catalyst to enhance the reaction between them. During the reaction, carbon atoms from the methane get deposited on the metal surface and the substrate is rapidly cooled down in order to freeze the layer of carbon atoms. this cooling should be so rapid in order to prevent the turning of layer of graphene into a bulky form. Thus a single layer of graphene is synthesized. Among synthesis techniques, chemical vapor deposition has shown great promise in producing high-quality graphene films.

The different kinds of substrate are used to growing graphene films on those appropriate transition metals. One of the examples is nickel (Ni). At the high temperature carbon atoms are decomposed and diffused into nickel, and during the cooling process carbon atoms are precipitation on the surface of the nickel. In the CVD method, single-layer graphene formulation and to keeping the quality of the graphene film can be very challenging because of the jumble of the growth conditions. In the University of Arkansas during research perceived the necessity to understand the growth phenomena as well as appropriate conditions for graphene production.

During the research study, the team found that if the temperature is high within the nickel, the diffusion of the carbon atoms is high and accelerates the diffusion process. From their results, they also found that more time was required for the carbon atoms to reach their penetration or saturation state in a broad Ni film. Additionally, the researchers modeled the super saturation by the cooling process.

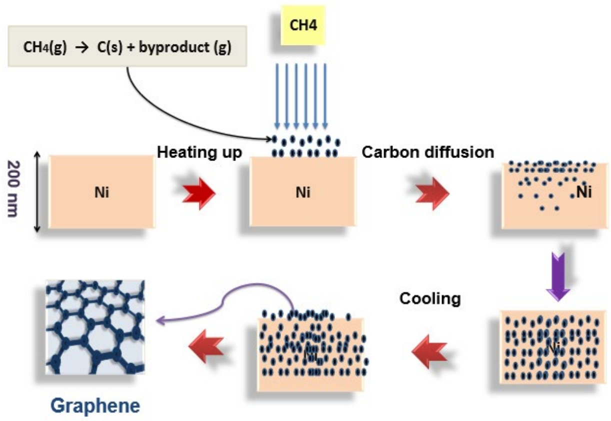

Fig. 2 Mechanism of CVD method on Ni substrate [23]

In the super saturation process, carbon atoms become segregated on the surface of the Ni thin film. During the cooling process of the film whose temperature from 900°C to 725°C, 1.6 layers of graphene obtained on the surface of film. This number of graphene layers which are generated through results proved acceptable in relative to experimental data.

First decomposition of hydrocarbon at around 1000ºC and then diffusion and saturation on different substrates such as nickel. Substrate selection is based upon the low solubility of carbon on the surface of substrate at reduced temperature. On these kind of substrates which has property of low carbon solubility carbon atoms decomposed from bulk material and nucleated on the substrates which further grow into larger domains. Once 100% of the surface covered with graphene further growth terminates automatically and thus it is a self-limiting technique/process. CH4, C_2H_2, ethanol, etc. can be used as carbon source and substrates Cu, Ni, Fe, Ge etc. can be used. Though there are many advantages of this process, however, this method is complex/complicated as it needs gas supply line, high temperature involved, and also high cost is involved and mass production is also still a challenge with traditional CVD. Further research is required to for industrial production like improve gas supply line, operating power and types of substrates.

Growth on Copper: Li *et al.,* used a same method to generate the monolayer graphene at large scale on copper foils. Copper foil of 25 μm thickness firstly heated at 1000◦C then added in a flow of 2 standard cubic centimetres per minute hydrogen at very low pressure and then explosion of methane flow of 35 sccm and pressure of 500mTorr. Dual gated FETs are fabricated by using graphene with a carrier mobility of 4050 $cm^2V^{-1}s^{-1}$ [40]. But now, roll-to-roll process was introduced to produce graphene layers with 30 inches of diagonal then transfer the graphene layer to transparent flexible substrates. CVD method is used to grown the graphene on copper and to support a graphene copper polymer support is used. After that chemical etching is used to remove the copper and then graphene film is transferred on a polyethylene terephthalate (PET) substrate.

Doped 4-layer graphene sheets were generated with a graphene sheet of resistance 30Ω/W and optical transmittance is higher than 90%. This 4 layer of graphene sheets are better than to indium tin oxide (ITO) which is currently used in touch screens and flat panel displays, with each sheet of resistance (100Ω/W for ITO) and very high optical transmittance.

E. Transfer Process

Next step to CVD technique in which first substrate on which graphene is grown is removed and then a layer of polymer such as poly-dimethylsiloxane (PDMS) or polymethyl methacrylate (PMMA) is coated (Fig. 3). The coating can be easily done using spin coating technique on graphene, which further gives it strength/support. After this substrate such as copper is etched from other side using chemical such as ferric chloride ($FeCl_3$). Now the substrate is removed and with polymer on one side it is easy to transfer graphene for any required application such as on solar cell. Graphene is leaving on any required substrate; a solvent can smoothly dissolve the polymer. To explore the uniformity of growth procedure, silicon dioxide (SiO_2) is an advantageous substrate for obtaining transparent graphene.

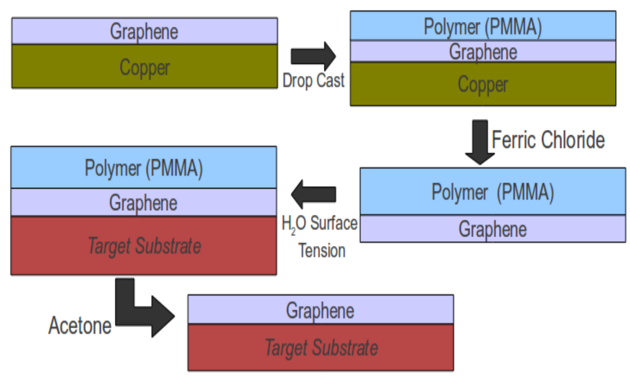

Fig. 3 Schematic diagram of the transfer process [25]

For various measurements like to measure the purity, electrical measurement and to check the conductivity, silicon dioxide is a good substrate. When the graphene is generated and verified through characterization on silicon it is easy to transfer the graphene to substrates so as to generate a solar cell such as organic films or glass slides.

TABLE I COMPARISON OF SYNTHESIS METHODS [26-32]

Method	Layer size	Mobility ($cm^2V^{-1}s^{-1}$)
Exfoliation	1 to 10+ 1mm	15000
Thermal SiC	1 to 4 50μm	2000
Ni-CVD	1 to 4 1 cm	3700
Cu-CVD	1 65 cm	16000

V. CONCLUSION

A 2D miracle material Graphene is known for its astonishing thermal, electrical/electronic, optical and excellent mechanical properties. Because of these surprisingly properties especially electrical/electronic conductivity this material can have potential application in various types of sensors. Large surface area of this material also contribute in the sensing properties with accuracy and high sensitivity. They are also stable in long range of temperature, pressure and environment conditions. Though there are various graphene synthesis methods available, but there is still need to work a lot in the direction of finding a low cost method to produce graphene films accurately and at large scale. Hummer method is good method in this direction. But scientist need to find right chemical which can act as reducing agent to reduce graphene oxide. In our research project we have found N_2H_4 (Hydrazine) better reducing agent as it speeds up the process in comparison to $NaBH_4$, nitric acid (HNO_3) and $C_6H_6O_2$ as a reducing agent. Our method also increases the conductivity of graphene and is also economical.

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