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A Review on Exotic Carbon Nanomaterial: Graphene

Neelesh Kumar Mishra[1], **Kalpana Awasthi**[2]

[1]Research Scholar, Department of Physics, K.N. Govt. P. G. College, Gyanpur, Bhadohi-221304, India
[2]Associate Professor, Department of Physics, K.N. Govt. P. G. College, Gyanpur, Bhadohi-221304, India
*Corresponding Author: awasthi.k@gmail.com

Abstract

Nanostructured materials have been fascinating the area of science and technology in the last twenty years because of their tremendous possibilities in forming novel shapes, structures and the unusual phenomena associated with these materials. Because of versatile and unique chemical and physical properties, there is a very crucial use of carbon-based nanomaterials in the scientific era of nanotechnology. The theoretical and experimental study of the two-dimensional (2D) structure of single sheet of graphite, known as graphene, is an extremely growing area in materials science and condensed matter physics. Due to the extraordinary properties of graphene, it has revealed an outstanding material and potential applications. This paper highlights the structure, properties, formation of graphene and its applications in different fields.

Keywords: Carbon based nanomaterials, Graphene, Structure, Properties, Synthesis, Applications

1. Introduction

Nanotechnology has established a connection between various branches of science and technology and has had an impact on various sectors of society. In the scientific age of nanotechnology, the use of carbon-based nanomaterials is of great importance because of their versatile and unique chemical and physical properties. The word carbon comes from the Latin "carbo", which means charcoal. In the modern age of science, it is not limited to charcoal, it is much more than that. The element carbon is present in abundance in the universe, and after hydrogen, helium, and oxygen, it is the most available element in the solar system, which is generally found in the form of hydrocarbons and other compounds (Pierson 2012). Carbon exists in the form of many important and useful allotropes, one of which is graphene. Graphene, with unbelievable mechanical, optical, chemical, and electrical properties, is considered the most conductive material in the world. It exhibits very high flexibility and transparency in nature. It is about 97% transparent, but it is so dense that it can’t allow a helium atom to pass through it. The conductivity of graphene is greater than copper, and it is stronger than diamond, although it is millions of times thinner than printing paper. In 1947, Wallace tried to develop a basis for explaining some of the physical properties of graphite through the band theory of solids with a single atomic layer. In 2004, Novoselov and Geim of Russia exfoliated graphene by the mechanical exfoliation technique from graphite using Scotch tape. They received the Nobel Prize in Physics (2010) from the Royal Swedish Academy of Sciences for their commendable work on graphene. In the present work, we briefly summarize the structure, properties, synthesis, and applications of graphene.

2. Carbon Allotropes

Allotropes of carbon are different structural forms in which pure carbon can exist, each with distinct physical properties due to varying atomic arrangements. These forms, while composed of the same element, exhibit different crystal structures and, consequently, different physical properties. Well-known forms of allotropes of crystalline carbon include diamond and graphite.

2.1 Diamond

It is the purest crystalline allotrope of carbon, in which carbon atom is covalently bonded to four other carbons in a tetrahedral geometry (Fig 1 (a)). These tetrahedrons together form a three-dimensional network of six-membered carbon rings in the chair conformation, allowing for zero bond angle strain. The bonding occurs through sp^3 hybridized orbitals to give a C-C bond length of 1.54 Å. It has the highest thermal conductivity and an unusually high index of refraction, is the hardest-known material, has excellent natural lubricity in air, and has extremely high strength and rigidity.

2.2 Graphite

Graphite is an engineering material with bulk, activated, fibrous, and exfoliated forms. It is created when the ground state electron of the 2s orbital is combined with two 2p orbitals, resulting in three sp^2 hybridized orbitals and one free unhybridized p orbital electron. These three sp^2 orbitals are arranged in a hexagonal structure, forming the graphene layer. Graphite has anisotropic properties and layers can easily slide over each other due to the weak van der Waals forces between the adjacent layers (Chung 2002) (Fig. 1 (b)).

2.3 Fullerene

In 1985, Kroto et al. at Rice University, discovered a C_{60} molecule consisting of 60 carbon atoms named fullerenes, also known as buckyballs. They suggested a truncated icosahedron, a polygon with 60 vertices, and 32 faces, 12 of which are pentagonal and 20 are hexagonal, as shown in Fig. 2. All fullerenes have an even number of carbon atoms and form a convex cage structure (Choudhary 2012). Fullerene has a zero dimension with discrete energy levels. C_{70} is another common fullerene, which contains 25 hexagons and 12 pentagons. In 1996, Harold W. Kroto, Robert F. Curl, and Richard E. Smalley were awarded the Nobel Prize in Chemistry. There are some important applications of fullerene and its derivatives, like superconductivity, ferromagnetism, chemical functionalization, and high electron affinity. Fullerenes are also used in solar cells, fuel cells, sensors, etc.

2.4 Carbon Nanotubes

The carbon nanotubes (CNTs) are long, thin, and hollow cylinders of carbon atoms arranged in a hexagonal array, and they have a one-dimensional structure on the nanoscale due to their high aspect ratio, exceeding 10000. In 1976, a new kind of fullerene, like cylindrical fullerene, known as buck tubes CNTs

(Oberlin et al. 1976). It was first discovered in 1991 by Sumio Iijima. There are three kinds of CNTs according to their structures, e.g., single-walled CNTs, double-walled CNTs, and multi-walled CNTs (Fig. 2). The CNTs have the property of superconductivity at low temperatures and are thermally stable up to 750^0C in air and 2800^0C in a vacuum, and higher intrinsic thermal conductivity is also observed in CNTs. Young's modulus of CNT is of the order of terapascal (TPa), which is five times that of steel, and its tensile strength is also fifty times that of steel (Yadav and Awasthi 2022). The measured value of current density is of the order of 10^9 A/cm², which is the highest value of known materials (Wei et al. 2001).

2.5 Carbon Nanohorns

Single-walled carbon nanohorns (SWNHs) also known as carbon nanohorns (CNHs), were first discovered by Iijima in 1999. SWNHs are tube-like or cone-like structures formed from a single layer of graphene, usually exist in a large spherical arrangement of about 80 to 100 nm in diameter, and look like dahlia flowers (Fig. 3) (Lanceta et al. 2020). The diameter and length of individual SWNH are 2-5nm and 40-50nm, respectively. They are bound through Vander Waals forces and the aggregated structure gives the advantages to CNHs due to the presence internal and external micropores. The SWNHs have intrinsic properties such as high dispersibility, high conductivity, and large specific surface area. Therefore, SWNHs have potential applications in supercapacitor, sensors, gas adsorption and drug carrier (Zhu and Xu 2010; Lanceta et al. 2020; Liu et al. 2020).

2.6 Carbon Dots

The carbon dots (CDs) are combinations of graphitic and turbostratic carbon in various volumetric ratios. The structure of CDs is quasi-spherical with diameters of 1-10 nm and has high oxygen content (Fig. 3) (Liu et al. 2020). It contains mostly sp^3 hybridized carbon and is generally amorphous with unique optical properties. The surface functional groups, such as hydroxyl, carboxyl, and amine groups, which provide the thermodynamic stabilities in different solvents. In 2004, CDs were discovered during the purification of SWCNTs (Xu et al. 2004). After two years, Sun et al. (2006) synthesized fluorescent carbon nanoparticles with a diameter less than 10 nm and named them carbon dots. The CDs show the properties of fluorescence, phosphorescence, electrochemical luminescence, up-conversion, photoinduced electron transfer, and optical absorption in the UV region (Oyohwose et al. 2023). Also, CDs have a huge impact on both health and environmental applications like sensing, water purification, solar cells, and bioimaging applications.

2.7 Graphene

In graphene, carbon atoms are bonded by sp^2 hybridization and arranged in a honeycomb-like structure, as shown in Fig. 3. Being very thin, about 0.33 nm, graphene acts as a basis of all graphite allotropes of carbon, such as graphite, carbon nanotubes, and fullerenes (Geim and Novoselov 2010). Graphene is a single layer of graphite, while bilayer and tri-layer graphene are made up of two or three layers of graphite. Graphene having more than five to ten layers is referred to as few layer graphene (Das and Choi 2011). Due to the wide range of applications, graphene has gained substantial research interest nowadays (Singh et al. 2022; Tiwari et al. 2016; Bellier et al. 2022). Graphene derivatives are materials based on or derived from graphene, often with modified properties due to structural changes or the addition of other elements or molecules. These derivatives, including graphene oxide (GO), reduced graphene oxide (RGO), and graphene quantum dots (GQDs), are created through modifications to the basic graphene structure shown in Fig. 4. The GO is derived from graphite using chemical oxidation and the oxygen-containing functional groups such as hydroxyl, epoxy, and carbonyl attached to both sides of the layer as well as the edges of the plane. These oxygen groups make GO more hydrophilic and dispersible in water and other polar solvents compared to pristine graphene. The RGO is derived from GO through a process that reduces oxygen-containing groups. It provides a cost-effective and scalable alternative to pristine graphene, spanning the gap between the oxygen-rich GO and the nearly perfect structure of graphene. The reduction process helps restores the sp^2 carbon network by eliminating oxygen-containing functional groups. Another derivative of graphene is GQDs, which are tiny fragments of graphene particles, with a lateral dimension of less than 20 nm (Li et al. 2010). Due to their zero dimensionality, quantum confinement and edge effects, GQDs have distinct optical and electrical properties (Li et al. 2015; Ghazali et al. 2023).

3. Properties Of Graphene

Graphene is a single layer of carbon atoms arranged in a hexagonal honeycomb lattice, with two sublattices of carbon atoms bonded to each other. It is the basic unit of graphite, with each layer interconnected by Van der Waals forces. Carbon atoms in graphene have four valence electrons, with three responsible for sigma bonding in a honeycomb lattice crystal, which makes it stronger and flexible, and the remaining p_z electrons form π-bonds with neighbouring orbitals. This creates a continuous pi orbital of an electron cloud responsible for the rich collective behaviour of graphene. Graphene has high electrical conductivity due to the free electrons available for the π bond. Carbon in graphene forms bonds with only three carbons present around it and releases an electron for the formation of a π bond, which is responsible for the conductivity of electrons in graphene (Tiwari et al. 2016).

Graphene has a high chemical reactivity due to its 2D structure and the availability of atoms on both sides. Doping with heteroatoms can further improve its reactivity, which is enhanced by structural defects (Denis et al. 2013). Graphene has an extremely short C-C bond (1.42 Å), which makes it the strongest material in the universe. It has been reported to have a tensile strength in gigapascals (GPa), i.e., 130 GPa, which is stronger than diamond and 200 times stronger than steel. The Young’s modulus of the graphene sheet was 1.0 TPa and the spring constant was found to be in the range of 1 to 5 N/m (Lee et al. 2008). The study of graphene thermal conductivity becomes more important with the use of graphene-based electronic devices. As a result of continuous progress in the electronics industry, the size of the devices is being minimized; as a result, the density of the circuit is also increasing, so heat removal is becoming an important issue that is essential for the proper operation of the devices. In such a situation, the possibility of applications of graphene with ultra-high thermal conductivity increases (Bae et al. 2011).

4. Overview of Graphene Synthesis

Monolayer (single-layer) is the purest form and ideal for electronic devices. The properties of bi-layer and tri-layer graphene alter as the number of layers increases. Graphene-based materials have seen significant growth in manufacture and utilization over the last two decades. The synthesis of graphene with a monolayer and high purity is still a challenge, which limits the scalable production of graphene and its potential commercial applications (Lee et al. 2019). This section describes the major and most popular graphene synthesis process, including mechanical exfoliation, chemical exfoliation, chemical synthesis, and chemical vapor deposition (CVD) (Mbayachi et al. 2021).

4.1.1. Mechanical exfoliation

This method is known as the Scotch tape or peel-off method, and it is the most famous and rarest process for extracting single-layer graphene flakes on preferred substrates. Natural graphite or graphitic material, like highly ordered pyrolytic graphite (HOPG) or single-crystal graphite, is the stacking of a single graphene layer, and one can obtain the graphene layers of different thicknesses by peeling off layers from these graphitic materials. After exfoliation, these layers were deposited on a silicon wafer using dry deposition. This process can be done using ultrasonication (Ci et al. 2009), electric fields (Liang et al. 2009), and even the transfer printing technique (Liang et al. 2007), etc.

4.1.2 Chemical Exfoliation

The chemical method is one of the most suitable methods for the synthesis of graphene, this method forms colloidal suspensions that modify graphene from graphite and graphite intercalation compounds. In this process, alkali metals are intercalated with the graphite structure to separate the few-layer graphene dispersed in the solution. Alkali metals have the characteristic that they can readily form graphite-intercalated structures with various stoichiometric ratios of graphite to alkali metals. Due to their ionic radii, alkali metals insert easily in the graphite interlayer spacing because they are smaller than the interlayer spacing (Das S and Choi 2011).

4.1.3 Chemical Synthesis

The graphite oxide was first prepared by chemist Benjamin C. Brodie (1859) of Oxford using a mixture of potassium chlorate ($KClO_3$) and fuming nitric acid (HNO_3) to oxidize graphite. Hummers and Offeman (1958) replaced the fuming HNO_3 acid with sodium nitrate ($NaNO_3$) and also $KClO_3$ with potassium permanganate ($KMnO_4$) as the oxidant. Graphite oxidation was achieved by a treatment where the $NaNO_3$ was added to the concentrated (conc.) H_2SO_4 and graphite powder. Another method was reported by Staudenmaier (1898) GO was prepared using conc. H_2SO_4, conc. HNO_3, and $KClO_3$ oxidizing agents. This process is accomplished in two steps, in which the first step is to increase the interlayer spacing by reducing the van der Waals forces so that the graphene-intercalated compounds are formed. Then, by rapid heating or sonication, single- or few-layered graphene is exfoliated from graphene-intercalated compounds. The microstructures of GO are shown in Fig. 5 (Talat et al. 2016). Fig. 5(a) represents the scanning electron microscopy (SEM) image of agglomerated sheets of GO. Fig.5 (b) shows the transmission electron microscopic (TEM) image of GO.

Ultrasonication is used for single-layer GO (Hummers and Offeman 1958), whereas density gradient ultracentrifugation is used for different layer thicknesses. Graphite is oxidized by the Hummer's method with the strong oxidizing agents $KMnO_4$ and $NaNO_3$ in the presence of H_2SO_4/ phosphoric acid (H_3PO_4). A single layer of graphene is produced from the mixture of DMF (dimethyl formamide)/water (9:1) by ultrasonication and in consequence, interlayer spacing increases from 3.7 to 9.5 Å. For oxidization high density of functional groups and reduction are required (Park et al. 2009; Tung et al. 2009).

4.1.4. Chemical Vapor Deposition (CVD)

In the chemical vapor deposition synthesis method, there is a chemical reaction in which process molecules are heated and changed to a gaseous state, and then it is called a precursor, and at high temperatures, a substrate is diffused on thermally disintegrated precursors. In CVD process, crystalline, solid, liquid, or gaseous precursors' thin films are deposited on the surface of the substrate. Various transition-metal substrates like Ni , Pd , Ru , Ir , and Cu are generally used in the CVD process for the deposition of high-quality graphene (Reina et al. 2009; Patel et al. 2022). When a reacting gas involved in the chemical reaction of the CVD process, it generates plasma inside a vacuum chamber, which leads to the deposition of thin film on the substrate surface, this process is known as plasma-enhanced chemical vapor deposition (PECVD). The CVD process is more convenient for industrial-scale applications.

4.1.5 Epitaxial Growth of Graphene on SiC Surface

Epitaxial thermal growth graphene on a single crystalline silicon carbide (SiC) surface is one of the most acclaimed methods of graphene synthesis. The epitaxy is a method in which the deposition of a single crystalline film on a single crystalline substrate, where this single crystalline film is known as epitaxial film or the epitaxial layer, and the process is known as epitaxial growth. Epitaxial growth of graphene is a process by which, high-crystalline graphene is fabricated on single crystalline SiC substrates. Layers are defined by the nature of the substrate and deposited films. When the deposited film and substrate are different in material then the deposited layer is known as a homo-epitaxial layer and if the film and substrate are different materials it is called a hetero-epitaxial layer (Das and Choi 2011).

5. Applications of Graphene

Graphene replaces silicon from transistors and supercapacitors. Graphene is also used in sensors for the detection of hazardous chemicals, photovoltaic applications, and optoelectronics (De Arco et al. 2010; Bonaccorso et al. 2010; Zhao et al. 2011). Due to good conductivity and the high surface area of reduced GO nanocomposites used in supercapacitors and lithium-ion batteries. The GO-based supercapacitors and lithium-ion batteries possess high-energy storage capacity, a long-life span, and good cycle stability. Graphene and its derivatives, such as GO are widely used in the biomedical field as lead agents in drug delivery systems. To kill cancer cells, magnetite adsorbed on the system with GO and doxorubicin hydrochloride (DXR) drug at a specific site is used. At present days, GO and RGO are included in many gadgets, and these gadgets can identify biologically significant molecules accurately because GO is used in specific characteristics to work effectively as a biosensor, known as fluorescence resonance energy transfer (FRET). It was found that the GO membrane allows to pass water but prevents some larger ions. Graphene nanoporous membranes can be ideally used for water desalination and filtration with an efficiency ranging from 33% to 100%, depending on the pore size and the applied pressure. In 2012, Tanugi and Grossman studied the classical molecular dynamics and reported that water could flow through graphene membranes at a range of up to 100L cm^{-2} day^{-1}, which is higher in magnitude than RO diffusive membranes. The factors that affect the desalination capacity of salt water are the pore size of the membrane, the quantity of chemical composition, and the applied pressure.

The performance of graphene lubricants is better than that of regularly used graphite lubricants. Graphene lubricants can last up to 6500 cycles if applied to ball and roller bearings or steel balls and discs, while graphite lubricants can only last up to 1000 cycles. A layer of graphene deposited on a glass substrate can absorb the radio wave of the bandwidth 125-165 GHz by 90%. In modern houses, graphene is used as a safeguard from radio wave interference by coating it on the roof, door, and window (Wu et al. 2014). Graphene and GO were used in solar cells as a transparent conductive cathode electrode (Yin et al. 2014). They utilized the RGO for solar cell application as a cathode electrode as well as a working electrode for the electrochemical deposition of material.

A graphene-based plasmonic nanoantenna (GPN) operates within the radio wavelength range. It is better than conventional antennas because its operational surface plasmon polaritons wavelength is much smaller compared to the wavelength of electromagnetic waves propagating at the same frequency. Due to the lightweight of graphene, it is a good material for the manufacturing of electrostatic audio microphones and speakers, which provides generally good frequency response. In 2015, an ultrasonic microphone and speaker were fabricated and operated in the frequency range of 20-500 kHz (Tiwari et al. 2016).

Conclusion

Two-dimensional carbon nanomaterial, i.e., graphene, has brought a revolution in science and engineering due to its structure and properties, and its discovery is proving to be a milestone in the field of materials science. Graphene is an attractive nanomaterial due to its superior properties, such as high tensile strength, high electrical conductivity, high carrier mobility, high elasticity, high thermal conductivity, and optical transparency. Due to these properties, graphene has applications in different sectors like transistors, solar cells, water purification sensors, batteries, and supercapacitors, etc. Several different methods were reported to synthesize the graphene/GO with varying degrees of success. The further work needs to be carried out to monolayer layer synthesis of graphene/GO in high quality and controlled manner. The current study summarizes the structure, properties, synthesis, and application of graphene.

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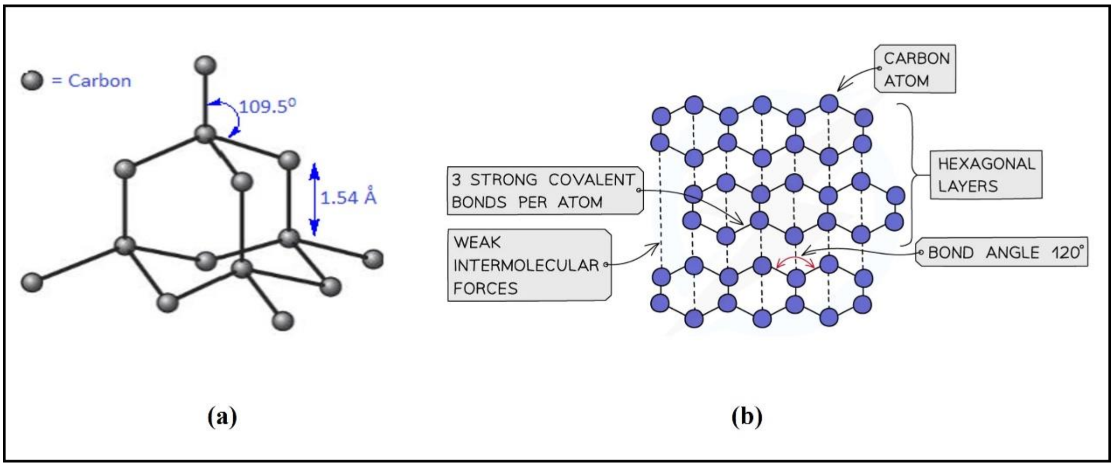

Fig.1: Crystal structure of diamond (a) and graphite (b) (source: google).

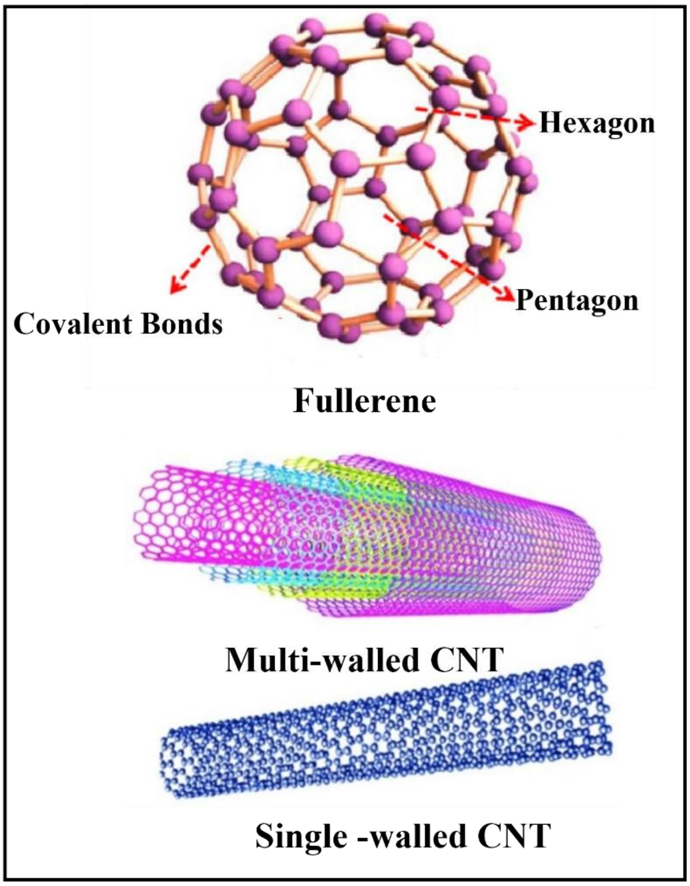

Fig. 2: The structure of fullerene and CNTs (Yadav and Awasthi 2022).

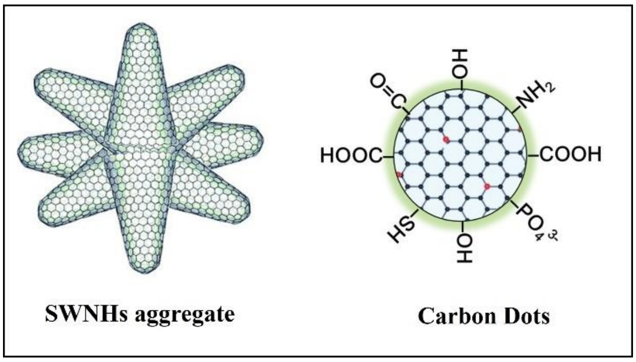

Fig.3: The structure of SWNHs (Source: google) and carbon dot (Liu et al. 2020)

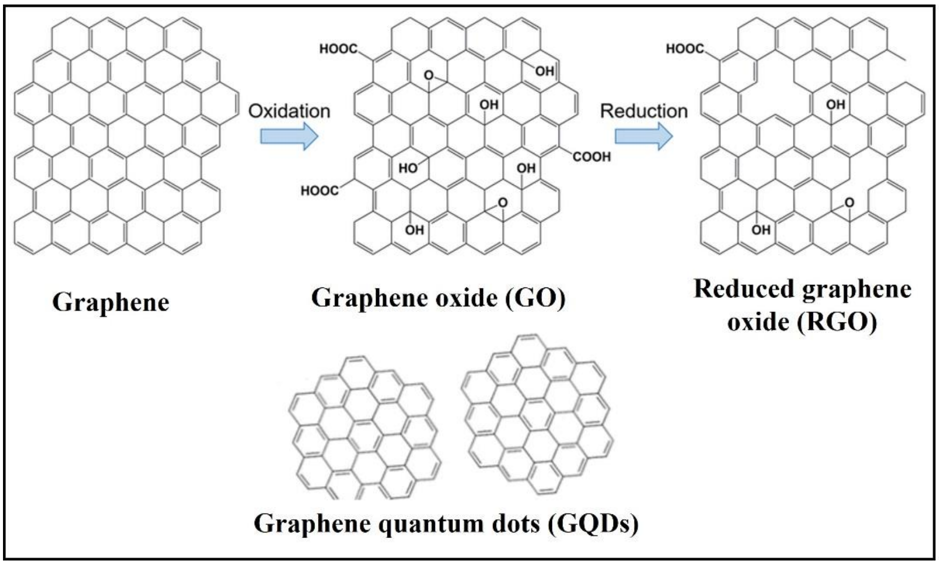

Fig.4: Derivatives of graphene (Bellier et al. 2022; and google).

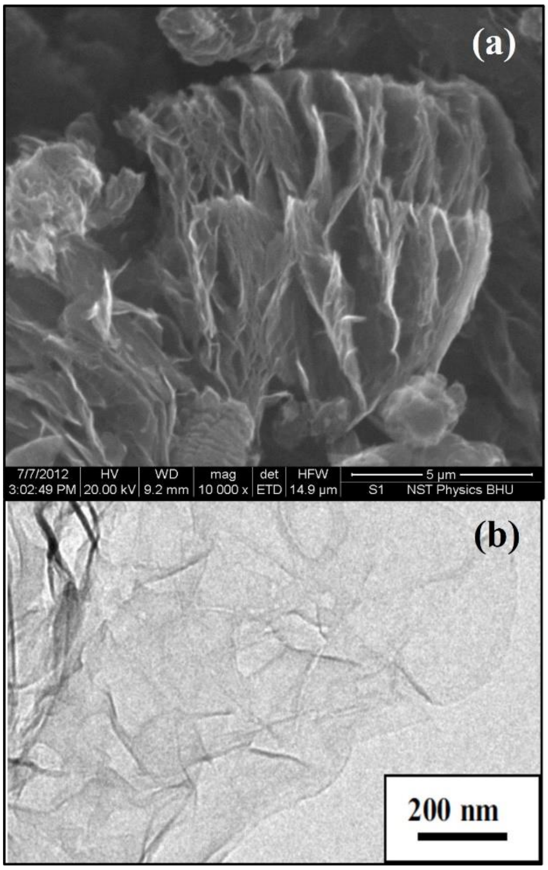

Fig. 5: (**a**) and (**b**) the SEM and TEM microstructures of GO, respectively (Talat et al. 2016).