Graphene and Its Composites

Marlinda Ab Rahman, Suresh Sagadevan, and Mohd Rafie Johan

Abstract Recent years have seen many innovations of graphene in various fields such as physics, chemistry, biology, and materials science. Graphene-based materials and their composites have a wide range of promising applications including electronics, biomedical devices, membranes, wearable sensors, and actuators. Graphene is a 2-dimensional (2D) array of carbon atoms in planar and hexagonal forms. Each carbon is sp^2-hybridized and connects three stable bonds C–C–μ at 120° apart. The unhybridized p-orbital, together with the same p-orbitals across the entire 2-D plane, is perpendicular to the sp^2-hybridization plane on other carbon atoms by Ś interaction. Graphene-based nanocomposites have drawn a great deal of attention in scientific communities, due to its extraordinary magnetic, mechanical, thermal and optical properties, and a large surface region. This chapter, therefore, presents different techniques in the graphene synthesis method and its excellent physical and chemical properties. Various manufacturing processes of graphene-based composites are introduced. In conclusion, the remaining challenges and perspectives in functional science and engineering for graphene nanocomposites are discussed.

Keywords Graphene · Composites · Synthesis methods · Physics · Chemical properties

1 Introduction

The graphite was discovered in a 16th-century mine close to Borrowdale in Cumbria, England. At the beginning of its finding, the graphite used for marking and descriptive purposes for example, nearby farmers used mine graphite blocks to mark their sheep. Due to its softness and dark colour, graphite has long been used as a particular type of lead. Graphene is a single carbon layer with a graphical structure in the chemical literature. Graphene is also classified as single carbon layers that appeared

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in the inter-compound of graphite. For example, it exists in a sequence of naphthalene, anthracenes, coronene, etc. For standard carbon jargon, the term "graphene layer" is also considered [1]. Since its discovery [2], the atomic structure with one layer has revolutionized the nanotechnology platform. After 2004, with its exponentially growing uses, graphene has invaded the nanotechnology market. Graphene is a "magic bullet" for the composite world due to its extraordinary properties. Graphene research achieved an unusually high elevation and was established in 2009 by Geim as a champion of the applied sciences.

Graphene is a single flat sheet from graphite and has the ideal two-dimensional (2D) structure with a monolayer of carbon atoms packed into a honeycomb crystal plane. Figure 1 shows the two-dimensional honeycomb structure of carbon atoms in graphene along with the high-resolution transmission electron microscopic (TEM) image. The TEM image clearly shows the hexagonal lattice with carbon–carbon distance 0.14 nm. Graphene is considered as the fundamental building block for graphitic materials of all other dimensions. It can be wrapped up into zero-dimensional (0D) fullerenes, rolled into one-dimensional (1D) nanotubes and stacked into three-dimensional (3D) graphite. Therefore, graphene is called the mother of all graphitic carbon-based nanomaterials. Figure 2 illustrates the schematic diagram demonstrating that graphene is the mother of all graphitic forms. Graphene is a crucial structural element in carbon allotropic, including graphite, carbon nanotubes, and fullerenes.

Therefore, graphene is a unique example of a layer of atomic membranes. Like any other membrane, two-dimensional structure distortions cost very little energy in a three-dimensional environment, so graphene has excellent potential for folding, distorting, forming scrolls, wrinkles, blisters, creases, etc. Nevertheless, graphene

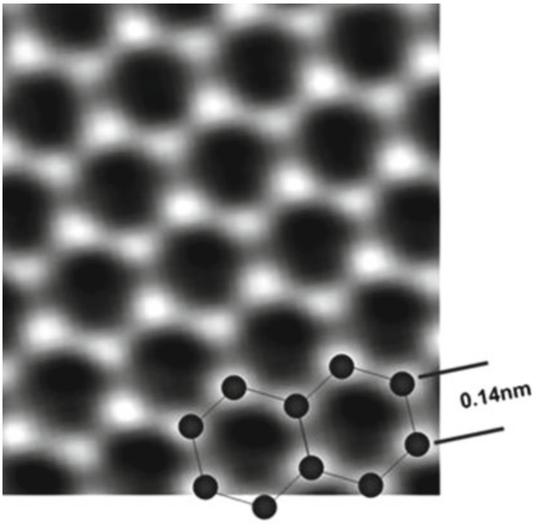


Fig. 1 TEM image of a graphene sheet illustrating the crystalline lattice (bond length ~0.14 nm) (Dato et al. 2009) [3]. It can be wrapped in 0D fullerenes, rolled in 1D nanotubes, or stacked in 3D graphite (see Fig. 2) [4]

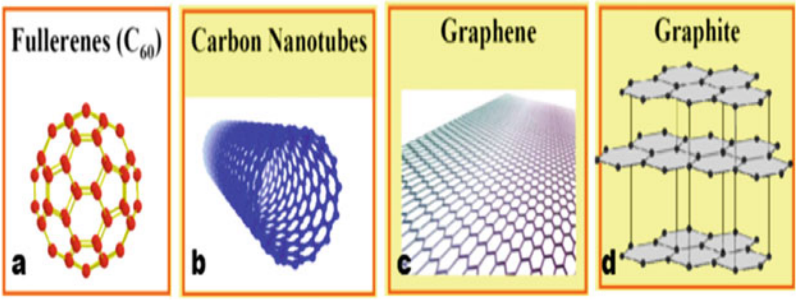


Fig. 2 Graphitic allotrope structure of **a** 0 D-fullerenes, **b** 1D-carbon, **c** 2D-graphene and **d** 3D-graphite (stacking of graphene layer)

can be stabilized to measure its properties through a substratum or scaffold experimentally. The high exfoliation potential of graphite-stacked strata is the most challenging aspect of graphite preparation [5]. Graphite is defined as a stack of graphene layers that rely on the stacking arrangement. This stacking relatively depends on the graphene orientation plane. Due to the weak interaction of each layer of graphite, the graphite constituent can easily be cleavaged. Graphite is a heap of graphene sheets that are tied together by van der Waals (see Fig. 2d), which is less fragile in-plane bonds than the covalent. As mentioned, the weak van der Waals interaction between the graphene sheets may make it easy to exfoliate a thin stack of graphene sheets from a bulk graphite, i.e., when it is scratched over a rough surface thin layers of graphite sticks to that surface.

There have been several attempts to synthesize graphene on a large scale in response to the demands of different industries, in particular, the composite sector, where graphene use has changed the state-of-the-art global market radically for the manufacturers. Graphene has drawn enormous interest in the scientific community because of its extraordinary physical, electrical, and mechanical properties [6, 7]. Some of the unique properties of graphene including high electron mobility at 2.5×10^5 cm^2 V^{-1} s^{-1} [8], high mechanical rigidity with Young's modulus of 1 TPa and intrinsic strength of 130 GPa [9], extremely high thermal efficiency over 3,000 W m K^{-1} [10], α-optical absorption is about 2.3% (in the network constant α [11], entirely permeable for any form of gas [12] and capable of extremely high densities of electrical current (a million times higher than copper) [13]. Graphene is applied in many sectors to a host matrix with a range of improved properties, including aeronautical technologies [14], electronics [15], long-lasting electricity [16], environmental [17], medicines [18] and food.

2 Synthesis Methods

Since Geim and Novoselov's first discovery of stable graphene monolayer in 2004, tremendous efforts have been made to prepare the graphene. Two approaches, classified as top-down and bottom-up, can be used to prepare graphene, as shown in Fig. 3. There are six significant techniques, including mechanical exfoliation [2, 19], epitaxial growth [20], CVD [21, 22], solvothermal [23, 24], graphite thermal expansion and chemical reduction of graphene oxide (GO) [25, 26] and microwave-assisted exfoliation [27] have been employed to prepare these two approaches to produce graphene. However, thermal [28, 29], hydrothermal [30], chemical [31, 26] and solvothermal reduction can be obtained in bulk quantities [23, 32]. These approaches are also versatile, scalable, and suitable for wide-ranging applications.

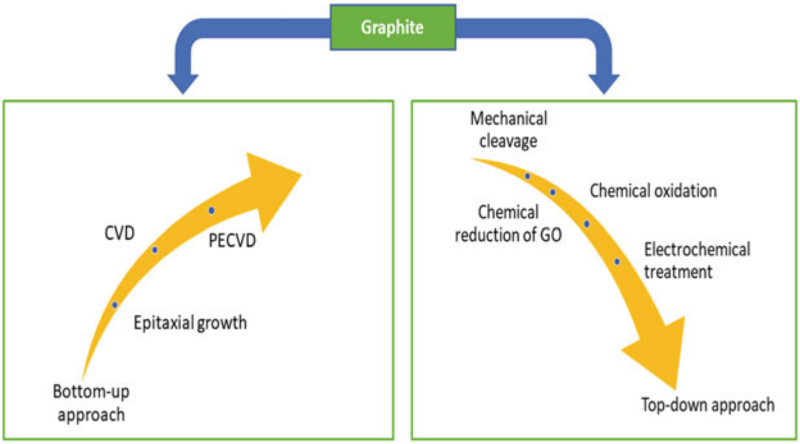
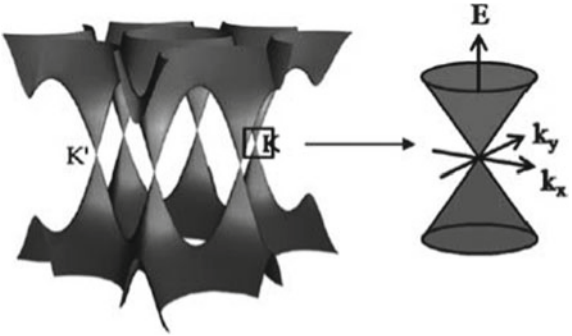


Fig. 3 Synthesis method of graphene from graphite

Fig. 4 shows the electronic band structure of single-layer graphene [33]



2.1 Top-Down Approach

In the top-down approach, graphene is made from graphite or graphite derivatives physical exfoliation, including mechanical cleavage, sonication, chemical oxidation, and graphite electrochemical exfoliation. The micromechanical technique of cleavage exfoliated graphene, which highly oriented via a scotch tape technique within graphite flakes. In 2004, Geim and Novaselov pioneered this technique. The consistency of single sheets of graphs [4] has benefited from this method. Stankovich et al. later introduced the ionizing process for the preparation of graphite oxide. This method involves the dispersion of graphite oxide n by acoustic hydrazine in a fluid medium [26]. Centrifugation eventually is an isolation process of non-exfoliated graphite from graphene. Unfortunately, incomplete removal of different functional groups during reduction processes causes the low quality of graphene as compared to graphene that is produced by the scotch-tape method. The two primary forms of liquid exfoliation methods, i.e., surfactant-free and surfactant-assisted liquid phase exfoliation have been successfully identified in Ciesielski and Samorì [34]. At present, chemical oxidation is the most promising technique with a high yield of graphene products. Graphene oxide sheets are obtained from graphite oxidation flakes subjected to three main methods formulated by Brodie [35], Hummers (1958), and Staudenmaier [36]. These methods produced GO layer structures with a brighter colour than graphite due to the loss of electronic conjugation brought during the oxidation of GO.

The GO layers are constructed of oxidized graphene sheets adorned with epoxy and hydroxy groups and with edge carboxy (Lerf—Klinowski model). These layers are composed of the basal planes. Such oxygen functionalities make hydrophone oxide galleries simple to interpose with GO hydrophilic graphene and water molecules. This oxygen versatility can also be called a graphite-type of intercalation complex between covalently binding oxygen-bonding, non-covalent, water-binding layers of carbon graphite oxide, and water molecules. In comparison, electrochemical graphite preparation was used to create the colloidal suspension of chemical sheets of Graphene [37].

The graphite sheets also can be prepared by using commercial graphite rods. This technique can be achieved by submerged graphite rods in an imidazolium-based phase-separated mixture of water and ionic liquids with an applied potential of 10–20 V. This technique was claimed to create ionic-liquid graphite functional sheets deriving from the precipitated anode of graphite (Liu et al.) [37]. In this DMF suspension, AFM calculated the average CMG box thickness of ~1,1 nm. Diverse forms of ionic fluids, as well as various ratios of ionic liquid to water, can affect the properties of graphene nanosheets.

2.2 Bottom-Up Approach

Walt de Heer and Claire Berger from Atlanta group [38], has developed another alternative method to obtain graphene consists of exposing to high temperature (>1100 °C) known as an epitaxial growth on SiC crystal (4H or 6H–) [39]. This method produces graphene epitaxial growth that the dimensions are depending on the size of the selected SiC substratum. The SiC surface shapes graphene, silicone, or finished carbon and has excellent effects on the thickness of the graphene, its stability, and its carrier density. This approach helped to define the critical characteristics of graphene and attracted more researchers, especially in the semiconductor industry.

The synthesis of graphene in a single layer (0001) ruthenium crystal plane using the chemical vapour deposition (CVD) process was reported in 2008 by Vazquez de Parga et al. [40]. The CVD process for the production of few-layer graphic image films on nickel layers documented by Kim et al. [21]. The template films were also successfully transferred without extreme mechanical or chemical processing onto random substrates to maintain the high crystalline consistency of the generated graph. Graphene may also be synthesized by using plasma-enhanced chemical vapour (PECVD) deposition to reduce costs at lower reaction temperatures [41]. Wang et al. have reported the first development of mono and some layers of graphite by radiofrequency PECVD on multitudes by injecting a gas mix of CH_4 and H_2 at 900 W at a temperature of 680 °C [42]. and by introducing a gas mixture of CH_4 and H_2 at a temperature of 680 °C [42].

3 Physical and Chemical Properties

This section aims to show the exciting features of a single, two, or few layers of graphene sheets. Until implementing the properties of the single graphene, it is crucial to understand the graphene structure clearly. Graphene in one layer may be described as a single two-dimensional material that is densely packed into a honeycomb crystal grid with carbon atom sheets. Bilayer and few graphene layers have two-dimensional carbon atom sheets of 2 and 3–10 layers, respectively. The graphene structures composed of over 10 two dimensional layers are used as a dense graphene board. The best way to determine the thickness of the graph sheet is to use microscopic and spectroscopic techniques.

Information on morphology and graph layer numbers can be investigated by using transmission electron microscopy (TEM). Single-layer Graphene displays a dark line and two or a few layers of graphene showing a folded edge with two faint lines. Raman spectroscopy, on the other hand, is another way to investigate graphene layer thickness. The first graphene spectra order of the Raman spectra of D, G, and 2D bands that match plane vibrations [43]. The number of graphene layers of G and 2D bands, their form, and the section strength ratio can be verified. G-band sensitivity decreases as the number of layers increases. The G-band downshift also demonstrates the increasing proportion of graphene layers. The change in layers shown by changing the shape, width, and position of the 2D band. The 2D bandwidth will distinguish between the single, bi, and few layers of graphene. The graphene of a single layer shows a peculiar electrical structure, showing an alternating band in the Brillouin region (Fig. 4), in two sharp points (K and K′). SLG is, therefore, expected to possess specific unique characteristics concerning metals, semiconductors, and semi-metals. The optimistic features of the space are portrayed in single-layer graphene, while there is no energy difference in the few layers graphene. The more layers the graphene structure contains, the more polished the stuff. Graphene has an excellent gas adsorption structure and hydrogen absorption.

4 Graphene with Composite Materials

Graphene can increase the conductivity and strength of bulk materials. This may also help to establish superior composite consistency. To create composites that are conductive and highly resistant to heat and pressure, graphene can be added to metals, polymers and ceramics. Graphene composites have many potential applications, creating unique and innovative materials with a lot of research going on. Recently, there are many applications based on coupling materials like graphene-polymer, which is widely used in medical implants, automotive and recycling, etc. The composite materials are structures formed to create unique finished content, incorporating two or more elements with different properties. They are also known as combination materials or direct composites. They’re not combined or merged, but in the final organic form, they remain separate. Thanks to their properties, composite materials can be made stronger, lighter, or softer than traditional materials by incorporating their different components. The bulk of the composites contain two materials-matrices (or binder) composed of a bundle of more durable fibres or fragments. Fibre glass, produced as the first plastic composite in the 1940s, is a typical example of this structure and is still commonly used. Fine glass fibres, tissued- into a fabric, are used as reinforcing fibre glass in a plastic or resin matrix. Although composites are not a new concept (for example, for thousands of years, dry mud bricks embedded in stroke have existed), many new and exciting composites have been produced using the latest technology. Through carefully choosing and updating the matrix, as well as the best manufacturing process to implement it, substantially superior products can be produced with tailored features for specific needs. Composite building materials, for example, concrete or asphalt, various metal composites, plastic blends, and ceramic composites, are usually constructed.

4.1 Composites with Polymers, Metals, Metal Oxides

It has attracted excellent graphene properties to be used as potential composite materials to achieve more exciting performance in electronics, optics, and photonic applications. In various matrix substances such as polymers, epoxy metals, and metal oxides, graphene and its derivatives have been incorporated. In 2006, Ruoff et al. synthesized the early introduction of graphene into the polymer matrix by solution-phase mixing with polystyrene. The polystyrene—graphene composite that formed exhibits a low percolation threshold of nearly 0.1 vol. percent at room temperature. Most of the previous composite graphene/polymer studies used graphene derivatives such as graphene oxide (GO) and reduced GO (RGO) as polymer matrix fillers combined with polyvinyl alcohol (PVA) [44], poly(methyl methacrylate) (PMMA) [45], poly(arylene sulfide) [46] and epoxy [47, 48].

However, the technique of ex-situ hybridization is likely much more straightforward than the method of in-situ hybridization. The graphene and pre-synthesized or commercially available nanocrystals are generally mixed in the medium of the solution. In this process, stacking through some linking molecule is one of the mechanisms involved in absorbing nanoparticles such as several examples, gold nanoparticles [49] and CdS nanoparticles [50] with surface modification on initial nanocrystal preparation and dispersion of graphene. Also reported was the manufacture of other metals such as silver, palladium, and platinum nanoparticles/graphene composites synthesized through covalent interaction [51]. Whereas graphene hybridization using semiconductor oxides was typically applied to TiO_2 or ZnO as well as to nanocomposite materials based on graphene synthesis. This method can be used to produce a high-quality graph-based nanocomposite without the use of any stabilizing agent. Many approaches have been used for the acquisition and analysis of these composites: electrochemical path, covalent mixture, thermal breakdown, and a sol–gel phase.

With support from UV and ethanol solution-photocatalytic reduction, William et al. [52], as well as Akhavan et al. [53], developed graphene/ZnO or graphene/TiO_2 composites. In all the above synthesis, carbon material/ZnO systems have outstanding characteristics. Graphene ZnO is therefore vital for environmental applications employing a quick, reliable, and cost-effective method.

5 Civil Engineering Applications

The unique mechanical, thermal, optical, and electrical properties of graphene will soon benefit civil engineering, says the National Physical Laboratory's Andrew Pollard. Graphene has many potential applications in civil engineering, but consistency raw material issues need to be overcome. Scientists at Manchester University first isolated graphene flakes in 2004 and winning a Nobel Prize in Physics 6 years later. Every year in this new and exciting area of research, the many excellent properties of the material have led to thousands of scientific publications and patents. It has already been used for flexible electronics, energy storage, nanocomposites, sensors, thermal management, and liquid filtration. Carbon is one of the universe's most common chemical elements and Life's essential elements. Furthermore, graphene is one of the universe's most straightforward molecular structures. For this 2D carbon material, though, while the future is bright, it is not without challenges. Lack of trust in the supply chain is one of the crucial barriers to real-world applications.

Most companies that want to develop graphene-enabled products are commonly struggling to find a reliable source of graphene suppliers' material. It is due to batch-to-batch variability. This problem can be worsened by the fact that the supplier nor the end-user uncertain about the physical or chemical properties of the graphene. The issue of commercialized graphene is the black powder or liquid containing flakes of one or several layers of graphene, as well as graphite, from the hundreds of suppliers worldwide. It is challenging to measure a statistically representative amount of nanoscale flakes for each piece of material that can be produced on the ton-scale. Similarly, there are no standardized measurement procedures to do this, and therefore there are no well-defined quality control techniques.

Graphene, another type of carbon nanoparticle, has recently revealed its excellence in enhancing various advanced properties as well as the desired mechanical properties, showing a path to the formation of a super concrete structure. Graphene and derivatives such as graphic nanoplatelets, graphene oxide, and lower graphene oxide provide increased stress sensing, temperature sensing and EMI shielding, wave absorption. Graphene has some remarkable and excellent properties such as large unique surfaces (2630 m^2 g^{-1}), high intrinsic strength (130 GPa). Young's firm package (~1.0 TPa). There is no improvement only in the rheology characteristics, but in the electrical properties of the material by integrating graphene into the cement composite. The electrical properties of the GNP-cement composite are critical. They can be used to track damage in a concrete structure so that the civil infrastructure is protected, secure, and reliable. Non-destructive testing has been shown to provide quick and effective inspection of these systems [54, 55].

Nevertheless, it is the concrete self-sensor that can track its stress that is important for this age. Through measuring shifts in the electrical resistivity factor [54], the pressure of cement-based composite reinforced conduction fillers can be detected. Automotive sensing is associated with the breakdown of conduction fibres when cracks are initiated in the cement composite, which enhances the sample's resistivity. As cracks open due to tensile loading or cracking, the resistivity values will be positive while the compressive load will be negative. A multifunction, high-performance composite based on types of cement, capable of perceiving damages in real-time automatically, will be generated in newly developed Nano-carbon engineering [56–59]. CNT consists of sp^2 derivatives, graphene sheets rolled up into a cylindrical tube, of a single layer of carbon [60]. The two-dimensional structure will be formed if these rolled CNT sheets are opened in one direction. Such two-dimensional sheets have an even higher surface area and aspect ratio (GNP). The effect of the improved GNP mortar crack on the adjustment of electrical resistance [61]. Samples (cube and prism forms) have been found to have responded to an increase in electrical strength with an increase in crack size. For the same relative crack size, the adjustment of the electrical resistance of the cube was more critical than that of the prism. According to the authors' knowledge, however, the response of the GNP-cement composite to different damage levels was not investigated. Therefore, in this analysis, Bingham, Revised Bingham, Herschel—Bulkley, and Casson models were used to test the fluvial properties of the GNP cement paste. In the flow curves of cement paste, a discrepancy was measured for different percentages of graphene nanoplatelets. The rheological properties of graphene cement paste with varying periods of rest (time between sample casting preparations) and shear rate cycles also were evaluated. The self-sensing properties of cemented concrete based on GNP were later determined. For calculating the electrical resistance, the four-sample procedure was used. Fractional improvements to strain sensitivity and intensity have been observed and used to determine self-sensing characteristics. Ultimately, the use of the GNP-cement composite sample was tested with the reinforced plate.

6 Mechanical Engineering Applications

The global automotive industry is facing major challenges, including CO_2 emission reductions, health risk, and energy efficiency. A new direction for greener and healthier cars is thus more than necessary. This situation opens up such a chance that graphene-based innovation is the right solution. With our commercial secret manufacturing process, the final cost of innovating the car industry by graphene is no longer the slow down factor. Graphene has significant uses in composites and coatings for producing light, sturdy and healthy vehicles. Graphene is more than 200-fold stronger than steel and can absorb carbon fibre, concrete, and aluminium components which able to increase the physical stability of the car. Heated car seats graphene-enhanced can be realized now without high-energy waste. In the years to come, graphene will strengthen or replace existing lithium batteries.

About seven or eight years, lightweight vehicles with graphene will be on the market and could lead to a much lighter and more energy-efficient production of automobiles. That is the expectation of Sunderland University, leading a consortium promoting revolutionary material. There is currently a pilot plant operating in Japan. University scientists worked on bumper made of graphene that can absorb 40% more energy than the standard item when added to carbon-reinforced plastic in some way. The result could be much lighter cars and more compact vehicles. Graphene is developed to produce a single-atomic substance or 1 million times thinner than a human hair, by breaking down graphite as the same material used in pencils. When dissolved powder in fluid, small amounts can be combined with the solvent used for impregnating carbon fibre. In the same process, a thermoplastic material can be moulded when it is heated and cooled actively. In this case, when molten, the graphene is combined with the rubber.

7 Electronics Engineering Applications

7.1 *Energy Storage*

Graphene has been looked at as an alternative to the current materials used in storing ions on the electrodes of supercapacitors. The greater the surface area, the more ions can be stored on it. Graphene has a theoretical surface area of around 2600 square meters per gram. Unfortunately, that theoretical surface area can be translated into a real device because that surface area is only achievable with a standalone sheet of graphene. In order to get it to work in a practical device that will provide a decent volumetric capacitance, you need to stack several sheets on top of each other. At this point, the theoretical surface area is lost, and you get about the same surface area you can get with the activated carbon used today.

Graphene thus makes it a rare commodity for batteries and supercapacitors to use. Graphene can enable faster charging and more energy-saving devices. It is also possible to use graphene to improve fuel cells. Graphene has a range of other practical applications: anti-corrosion coatings and lacquers, reliable, precise sensors, faster and better electronics, versatile screens, powerful solar panels, quicker DNA sequencing, and more. Applications of graphene are so broad and received attention in every company to gain benefits from this new material. The impact of these new materials will be more fitting in the soon future.

7.2 *Electronics*

Graphene is an important and broad building block that seems to gain every industry's attention for this new material. Moreover, this entire carbon atom structure has unique quantum properties. These graphene properties will help us study Majorana Fermions' properties and achieve superconductors at a high critical temperature. Based on such research, new types of flexible solar cells and new types of quantum computers can be created. In new energy and quantum computing, graphene technology has excellent potential. Graphene edges have become the best material for making supercapacitors in the field of new energy because they can carry a large amount of charge. In addition, a large number of studies have been conducted using graphene or derivative structures for thin-film solar cell electrodes. It is possible to make the most efficient all-carbon-based solar cells with 3D structures and organic chemical modification based on the newly discovered graphene quantum properties. Carbon nanotubes are excellent microelectrode materials as graphene derivatives and have great potential in brain-computer interfaces. It is biocompatible compared to the metal electrode material while having excellent electrical conductivity, and it is also easier to perform molecular-level modifications to induce synapse growth. Also, these electrodes have been tested by some studies. Smaller and more efficient computers can be manufactured using graphene-based materials. This computer can improve the efficiency of the EEG signal analysis, thus achieving a more individualized two - way human and computer adaptation. Thus, it can help users to learn more quickly on how to operate the brain-computer interface.

8 Conclusions and Future Work

Graphene has excellent mechanical, thermal, electrical properties and is a better substitute for traditional nanofillers for polymer matrices. We addressed recent advances in the materials, processing and future applications of graphene nanocomposites. It also defined various techniques and the excellent physical and chemical properties of the graphene synthesis process. Therefore, it is necessary to make further contributions to the identification of biocompatibility, to improve the stability of nanocarriers, to their size and also to provide more studies on the toxicity of this new material. The physical and chemical features of graphene have appeared with graphene electronics, including biosensors, organic solar cells, actuators, and touch screens. Graphene is used in future electronics via lightweight, stretchable structures as a possible part of these properties. Although many technically feasible solutions are currently being established, there are still several practical difficulties. The advantages of graphics-based materials are primarily mentioned in this analysis, as described and listed.

Nonetheless, the world of graphene is feasible with less costly pilot-scale production technologies. Rising GO can achieve this to graphene, one of the major challenges shortly. After the reduction process successfully reaches the pure graphene, its outstanding electronic and mechanical properties will be recovered. In short, it has unique features for various engineering uses. Graphene and its composites have unique properties.

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