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Review Article

A review on synthesis of graphene-based materials for energy storage devices

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Abstract

The extraordinary properties of graphene have made application of this wonder material in multidisciplinary domains. Promising applications of graphene and its composites have been made a reality through enormous scientific efforts to date. In the present era, the attention has to focus on the right choice of methodology analogous to each application. The prospective product range of graphene is broad, and the attentiveness of each application would be deviated accordingly. We believe priority should be given to the most essential needs among the vast variety of opportunities. Here we focus on methods followed in the production of graphene and composites and their applications in energy storage devices including batteries, capacitors, and fuel cells. Physical, chemical, and biological methodologies followed in minute scale synthesis as well as mass level are discussed. The information would be supportive of the development of more effective energy storage devices based on graphene in soon.

Keywords: Graphene, Energy storage devices, Graphene applications

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1.   INTRODUCTION

Though graphene is known to be a single layer of graphite, its synthesis did not become a reality until it was isolated from graphite by Geim and Novoselov in 2004 (1-2). Afterwards, significant scientific attention was drawn up to now. Graphene consists of carbon atoms in an atomic-thick hexagonal lattice as shown in Figure 1. The σ-bond between carbon atoms is responsible for the strength of the lattice structure. The most outstanding properties of graphene include superior electronic properties and crystal stability, high surface area to volume ratio, flexibility, high electrical and thermal conductivity, lightness, transparency, etc. (1-5). Different properties resulted in comprehensive research publications from diverse applications including electronics, sensors, energy, safety, sports, medical appliances, automobiles, spacecraft, and many more (6-9). Valuable commercial products based on graphene have reached the market by now. The potential product range is huge (10-12). Here we review methodologies followed in the production of graphene and its composites for various energy storage applications including Li-ion and Li-polymer batteries, supercapacitors, and fuel cells.
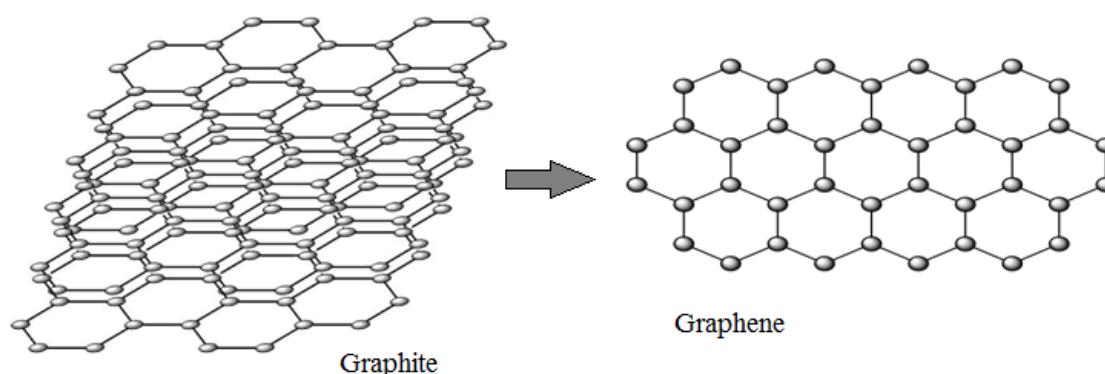

Figure 1: Structure of graphite and graphene

2.   GRAPHENE SYNTHESIS

Commonly used graphene synthesis methods for various applications have been extensively reviewed recently (13-15). In there, numerous methods are reported to synthesize graphene which can be categorized under top-down and bottom-up approaches (16). Stacked layers of graphite exfoliated into single graphene sheets are called the top-down approach (17) while Graphene produced via molecular growth from small molecular carbon precursors is known as the bottom-up approach (1).
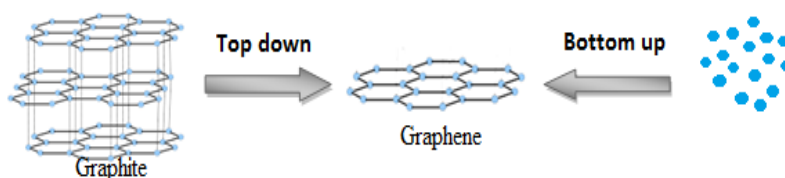
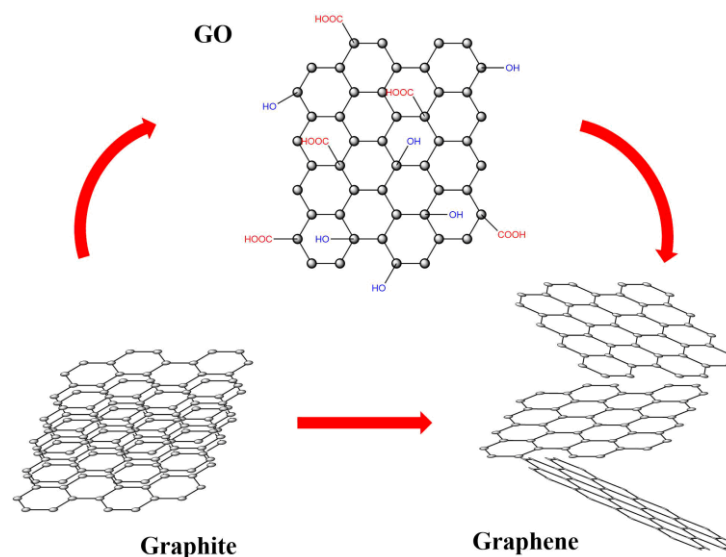Figure 2: Bottom-up and top-down synthesis approaches

2.1 Top-down approach

Figure 3: Graphite exfoliation to graphene through chemical and physical approach

2.1.1 *Mechanical exfoliation*

Mechanical exfoliation enables separating graphene layers into individual atomic layers of graphene as shown in Figure 3. For the first time, Novoselov and Geim exfoliated graphite to single graphene layers by using a simple adhesive tape also known as the scotch tape method in 2004 for which they obtained Nobel Prize in physics in 2010 (1). A piece of tape removes the graphene sheet after the exfoliation of graphite by dissolving it in a solvent. It gives high-quality, defect-free graphene but produces lower yields and high production costs which cannot be used for large-scale production (18).

2.1.2 *Sonication method*

Favourable for scalable production, exfoliate graphite is subjected to ultra-sonication. Hernandez et al. reported graphene production via sonication-assisted liquid-phase graphite exfoliation in 2008 (19). Since the graphene surface energy was estimated as 46.7 mN/m,

solvents with surface tension matching ~40 mN/m, were identified as the best solvents (20). Later, many researchers optimized sonication time, altered ambient conditions, increased initial graphite concentrations, introduced surfactants, and polymers as well as adopted solvent exchange methods, etc.(21-24). The resulting graphene was incorporated in applications, including thin-film transistors, light-emitting diodes (LED), and photovoltaic cells (25-28). However, the sonication method has more defects and disorders with advantages in cost, scalability, and easy production (29).

2.1.3 *Ball milling*

In the powder production process, thick graphite flakes are turned into graphene by using milling impact enabling exfoliation and fragmentation (30). Both wet and dry ball milling are used to synthesize graphene under this method (31). It involves dispersing graphite in suitable solvents to match the surface tension to overcome the Vander Waals force of adjacent graphene flakes (32). A mixture of graphite and chemically inert water-soluble inorganic salts are used for the dry method (33). Advantageous on large-scale production but fragmentation, defects, long processing time, and high energy consumption seems to be the disadvantages. The ball milling is suitable for the production of functionalized graphene for functional coating, energy storage, and electrochemical sensors (34-36).
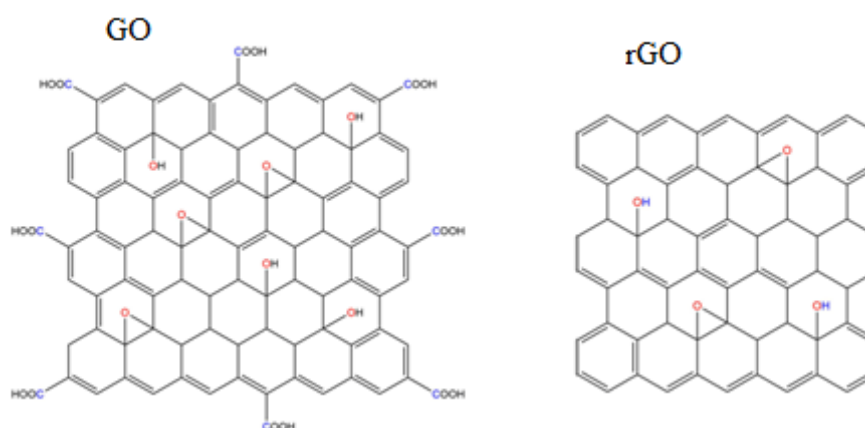
2.1.4. *Electrochemical exfoliation*

Figure 4: Difference between graphite oxide and reduced graphite oxide

The inter-planer distance is increased by the ionic species in an electrolyte intercalating into the graphite electrode as a result of an applied voltage (37). Although, graphite exfoliation conducts in two-electrode and three-electrode (38), commonly used two-electrode

exfoliation involves the intercalation, oxidation, and exfoliation steps of graphite (39). First reported by F. Beck et al. in 1995(40) later in 2011, You et al. prepared GO by using a two-electrode exfoliation method with KCl as the electrolyte and expanded graphite anode and platinum wire cathode at a voltage of 4 V (41). In 2018, S. Pei et al. reported a scalable, safe, ultrafast, and green method to synthesize clean GO sheets by water electrolytic oxidation of graphite (42). High electrical conductivity environmental friendliness, high efficiency, and relatively low cost are main advantages (43). Graphene prepared through electrochemical exfoliation is used in supercapacitors, batteries, sensors, catalysts, hybrid membranes, anticorrosion coatings, fire hazard suppression materials, etc. (44-46).

2.1.5 *Chemical exfoliation*

Chemical oxidation was used to produce graphite oxide by B.C. Brodie in 1859 (47), Hummer and Offeman in 1958 (48) has become a popular method by now. First, graphite is oxidized to graphite oxide using a powdered form of graphite, sulfuric acid, sodium nitrate, and potassium permanganate. Altering drawbacks of the hummers method in 2010 Tour's method was introduced to gain oxidation reaction without sodium nitrate and increased potassium permanganate with phosphoric acid. This method produces multi-layers or a few layers of graphene in flake types having a lateral length in the range of several hundred nanometers to several micrometers (49). The method in mass production of graphene flakes with low purity for various applications including fillers, coating, conductive ink paste, batteries, etc. Various other physical and chemical methods are followed to prepare graphene from graphite oxide (Figure 3). A simple and fast low-temperature thermal method has been followed to produce graphene from graphite oxide in a mass scale (47). Jayaweera et al. in 2014, 2021 demonstrated a novel one-step in-situ sonochemical method to synthesize graphene from graphite oxide. $SnCl_2.2H_2O$ has been used to reduce graphite oxide while graphene/SnO2 composite is formed in a scalable and straightforward one-pot process (49-50). These graphene composites have shown good gas sensing properties as well as high current density, and outstanding charge-discharge characteristic supercapacitors. GO has also been reduced to rGO using coffeaarabica leave for bio-medical application for the first time which is referred to a green synthesis method (51).

Figure 5: Reduction of graphite oxide using Coffea arabica.

However chemical exfoliation is not suitable for applications where high purity is mandatory such as in electronics. High scalability and low cost are the advantages of the chemical exfoliation method. Disadvantages of this method are low purity, hazardous chemical use, product agglomeration, etc. (52-53).

2.2 Bottom–up approach

2.2.1 *Chemical Vapor Deposition (CVD)*

A large-scale and high-quality mono-layer graphene could be produced by Chemical Vapour Deposition (CVD). Thermal CVD Plasma-enhanced CVD, and hot/cold wall CVD are popular methods that employs thin film deposition substrates from vapor types over chemical reactions (54-56). Molecules being heated into a gaseous state are deposited on the surface of a substrate (51,57). Pressure, temperature, depositing time, precursor nature, gas flow state, and wall/substrate temperature, seems to affect the product quality (58). A.E Karuin et al. in 1966, obtained highly crystalline graphite films on Ni substrates (59). Land at el. in 1992, deposited mono-layer graphene on Pt substrate which was a hydrocarbon decomposition at 530 °C (60). Later Ni, Cu, Pd, Ru, Ir, and Pt substrates became the studies of many research groups (61-62). Ethylene ($CH_2=CH_2$), methane (CH_4), and acetylene (C_2H_2) are used as typical gaseous carbon precursors [63]. High quality and large-scale production processes proved to be the advantages while toxic gas, complexity, and high costs are the disadvantages (64). Applications developing touch screens, smart windows, flexible LCDs, and solar cells could be highlighted (65-66).

2.2.2 *Epitaxial growth*

Thermal decomposition of SiC in a high vacuum is the way of this method. Could be classified into two types homo-epitaxial and hetero-epitaxial (67). If the same substrate and deposited materials are used it is categorized under the homo-epitaxial process, while different materials are classified under hetero-epitaxial process. In 1893 Acheson et al. introduced the concept of producing silicon carbide by heating different carbonaceous sources (68). A. Charrier et al. revealed the thermal decomposition of an electronic-grade wafer of 6H-SiC after annealing under increasing temperature which leads to a layer-by-layer growth of non-distractive single-crystalline graphite (69). Kim. et al. developed ferroelectric non-volatile memory devices which used graphene electrodes as the epitaxial growth substrate for crystalline polymer (70). Hoang, et al. generated a high-quality MoS_2/graphene hetero-structure. They have been found to exhibit inverse photo response with highly uniform photo responsivity in the photodetector pixels fabricated across a full wafer (71). High-quality graphene is the main advantage while the high production cost, low yield, and inhomogeneity are the major limitations.

3.   GRAPHENE IN LITHIUM-ION BATTERIES

Li-ion battery (LIB) is an advanced rechargeable battery introduced by Sony in 1991 using graphite as the anode replacing Lithium metal. Since then the Li-ion battery is being used in almost all electronics from smartphones and computers to electric vehicles. The LIB is consisting of $LiCoO_2$ cathode, graphite anode and LiPF6 in propylene carbonate, and diethyl carbonate (PC:DEC, 1:1) as the electrolyte which is still the core battery today (72). During the discharge, lithium ions pass from the anode to the cathode through a separator in the electrolyte and recombine with electrons from the external circuit where they neutralize electrically (Figure 06). The electrodes used were lithium cobalt oxide cathode and graphite anode.
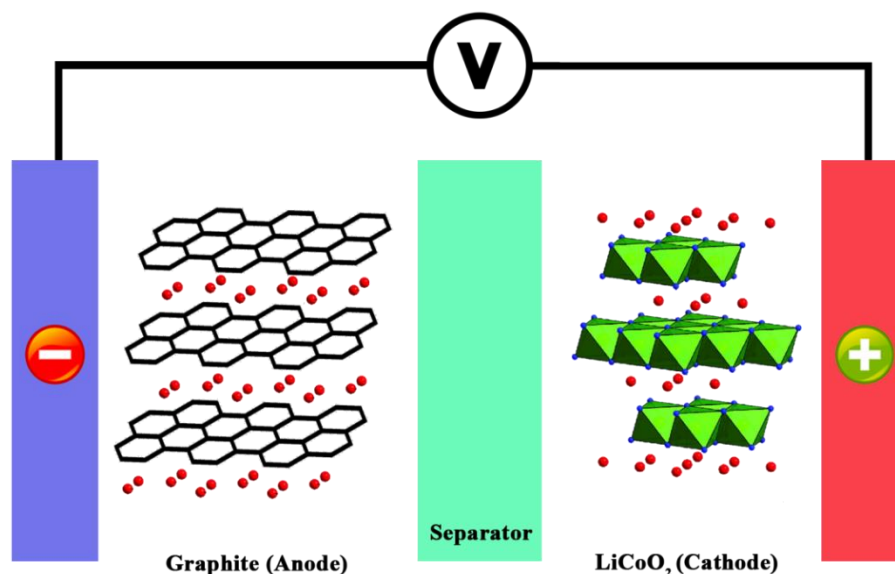
Figure 6: Schematic diagram of Li-ion battery

Mogalahalli et al. recently reported on the history of the development of graphite based LIBs through 50 years (72). The concept of using two intercalation materials for cathodes and anodes in LIBs had been proposed by Michel Armand in the 1970s in which lithium ions are transferred from one side to the other during the charge and discharge (73). Armand's investigations on electrodes and electrolytes have played a major role in today's Li-ion and all solid-state batteries. In addition, LIBs have been named as the rocking-chair battery due to the transferring of ions from one electrode to another on charge-discharge cycling (74). The LIB gained major success in the battery industry with efforts by Sony in 1985 (75) as well as Sanyo in 1988 (76). After proposing $LiCoO_2$ as a positive electrode by Good Enough, Lazzari, and Scrosati have shown their promises by 1980 (77). Subsequently, commercialization of the $LiCoO_2$-based LIB had been done by the Sony and Asahi Kasei teams led by Nishi in 1991(72).

More recently, after the invention of graphene, worldwide research attention has been attracted to use graphene as an anode to replace graphite anode. The theoretical specific capacity of graphite is 372 mAh/g while that of graphene can be almost double since there is the possibility of intercalating Li-on on both sides of graphene layers. Graphene has been used to improve the rate performance and stability of lithium-ion batteries. Recently, Ali et.al. (2022) reviewed on the function of graphene on Li-ion batteries (78) in which critical issues, benefits, challenges, research directions, and perspectives of graphene were discussed. In addition, Chen et. al (2021) reviewed the application of graphene in cathode

(79), and synthesis and functions were discussed. Liu et al (2022) have reviewed graphene-based composites in the anode of Li-ion batteries (80). The challenges and guidance for improvements were discussed.

G. Wang et.al (2009), P. Lian et.al (2010), and E. Yoo et al (2008) have synthesized graphene nano-sheets in large quantities using the Hummer method followed by a thermal method which has exhibited an enhanced lithium storage capacity above the theoretical capacity of graphite as anodes in lithium-ion batteries (81-83). However, continuous decay in capacity has been shown throughout 100 cycles which is impractical in commercial batteries. Kottegoda et.al. have compared two graphene samples prepared from two graphite sources using the Hummer method followed by a novel low-temperature thermal method, in which high theoretical specific capacity as well as comparable cycling stability has been exhibited (52). High irreversible capacity during the first few charge-discharge processes and continuous decay in capacity limits the application of graphene as an anode despite the high specific capacity and rate capability. While their Graphene/NiO composite, which has been synthesized using a hydrothermal method from graphite oxide and nickel acetate, has shown better cycling stability and capacity as an anode (84). In the study graphene synthesized using the Hummer's method followed by a hydrothermal method has been shown higher specific capacity at the beginning and high irreversibility up to 20 initial cycles. Graphene/SnO$_2$ composite synthesized using combined methods through in-situ metal tin reduction and thermal treatment by Q. Tan et.al in 2019 has exhibited not only a higher reversible capacity but also a long and stable cycling life under a high current density (85) while C/CoO/Graphene composite that W. Zhang et.al studied with core–shelled micro balls and electrochemically exfoliated graphene has shown high reversible capacity, good rate capability and long-life retaining high capacity (86). Moreover, Si/Graphene composite as anode material had been prepared by M. Sui et.al using a spray drying approach and low-temperature reduction method has delivered an initial charge capacity of 1298.1 mAh g^{-1} and good rate performance (87). Gao et.al (2015) have used multilayer Si/rGO composite electrodes synthesized from bulk Si particles through electroless etching and graphene self-encapsulating method has shown a reversible capacity of 2787 mAh g^{-1} at 100 mA g^{-1}, and a stable capacity over 1000 mAh g^{-1} (88). Z. Ma et.al. have fabricated Si/graphene layer structure fabricated by a high pressure method so that Si particle surfaces are covered by graphene layers. They have shown a significant capacity of 2096.9 mA h g^{-1} at 300 mA g^{-1} and capacity retention even at a high rate (89).

B. Shen et.al have prepared porous silicon in the network of graphene/ carbon nanotube by a single step reduction and self-assembly approach showing a high charge capacity of 2319 mAh g^{-1} at 0.1 A g^{-1} and stable cycle ability (90). Furthermore, there are a significant number of publications on Si/graphene metal oxide composites though referred a few in the present review (91-92). The volume expansion of Si-graphene electrodes makes high irreversibility although the capacity is considerably higher than the other composite. The significant reality in all the recent and previous work of graphene as well as graphene-based composite for LIB involves high initial capacity together with high irreversible capacity loss and capacity degradation throughout subsequent cycles. This major limitation needs to be addressed before the commercialization of graphene as well as its composites as an anode in LIBs. As a result, the synthesis method of graphene or its composite remains substantial work to be done before commercialization.
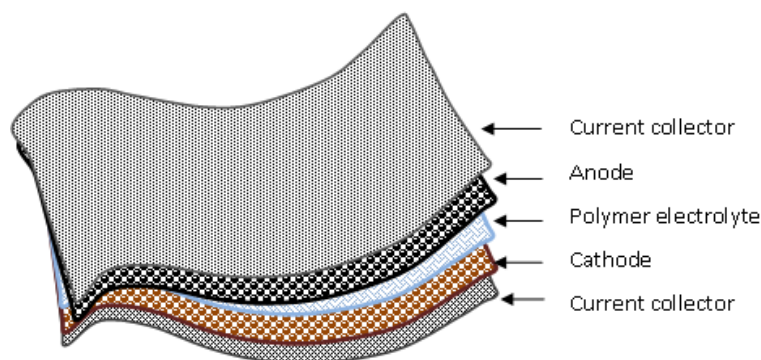
4. GRAPHENE IN LITHIOM POLYMER BATTERIES

Figure 7: Schematic diagram of lithium polymer battery

It is well understood that the Lithium battery based on a solid electrolyte which is known as 'solid state battery' can greatly overcome energy density, reliability, safety, and the performance of the common lithium-ion battery based on the liquid counterpart. However, the lack of a polymer electrolyte with good thermal stability, efficient Li-ion transport, and acceptable ionic conductivity hinders the early development of the device. In 1978-1981, polyethylene oxide ~PEO-based solid polymer electrolytes were proposed for battery application by Armand et al. (93-95). Having used Li metal as the anode, higher theoretical energy density is possible than with the liquid electrolytes. However, the main limitation is

poor ionic conductivity of the PEO and polymers at room temperature and perform mostly at elevated temperatures. The approaches such as the introduction of inorganic filler to the PEO-LiX matrix (96-97) and plasticized or gel polymer electrolytes with liquid additives (98-100) and PEG oligomers (101-102), derivatives of polymeric borate esters (103-105) and PC with borate ester plasticizers (106-107) have also been used as polymer electrolytes which shows significant electrochemical performance. Later on a new class of single-ion solid polymer, solvent-in-salt electrolytes innovative polymeric materials have attracted much attention in practical aspects (108-111). In this regard, polymer electrolytes have been extensively investigated in Li-metal batteries in which considerable progress in electrochemical performances has been shown (112). It is important to review compatibility of polymer electrolytes with graphite and graphene since graphene-based materials have been highly attractive as an anode in Li-ion batteries. Graphene-based polymer Li batteries could be lighter, slimmer and have greater capacities than today's lithium-ion cells. If Lithium-ion stores certain energy per kilogram graphene can store much higher energy per kilogram. Moreover, graphene is safer, much more stable, flexible, and stronger. However, graphene-based all solid-state batteries find exciting potential in commercialization. More recently D. Jeonget al. in 2022 have prepared ion-conducting gel polymer electrolytes by an in-situ cross-linking reaction of methacrylate graphene oxide and lithium cation conducting monomer using poly(vinylidene fluoride-co-hexafluoropropylene) as a matrix polymer with a view of lithium dendrite suppression and much improved cycle performance has been obtained as a result (113). Li-ion conducting poly (vinylidene fluoride) /sulfonated graphene oxide nanocomposite polymer electrolytes have been prepared by M. Fouladv and et. al in 2021 and have shown high discharge capacity and capacity retention in lithium-ion batteries (114). In addition, companies all over including Samsung and Huawei are working on graphene-enhanced batteries. And all other extensive R&D works going on at present would yield results shortly.

5. GRAPHENE IN CAPACITORS AND SUPER CAPACITORS

A capacitor is a passive electronic component that is used to store and discharge electrical energy. It consists of two dielectric plates separating two conducting plates (Figure 8). An electric field is formed in the dielectric material when a voltage is applied across the plates, and this field stores energy. Capacitors are frequently utilized in electronic circuits to filter, store, and regulate electrical signals. They are also used in timing circuits, power supply, and other systems where the storage and release of electrical energy are crucial (11).

Supercapacitors also are a promising alternative to batteries for energy storage applications because they can store and release energy more quickly than batteries, have a longer lifespan, and can operate over a wider range of temperatures (115).
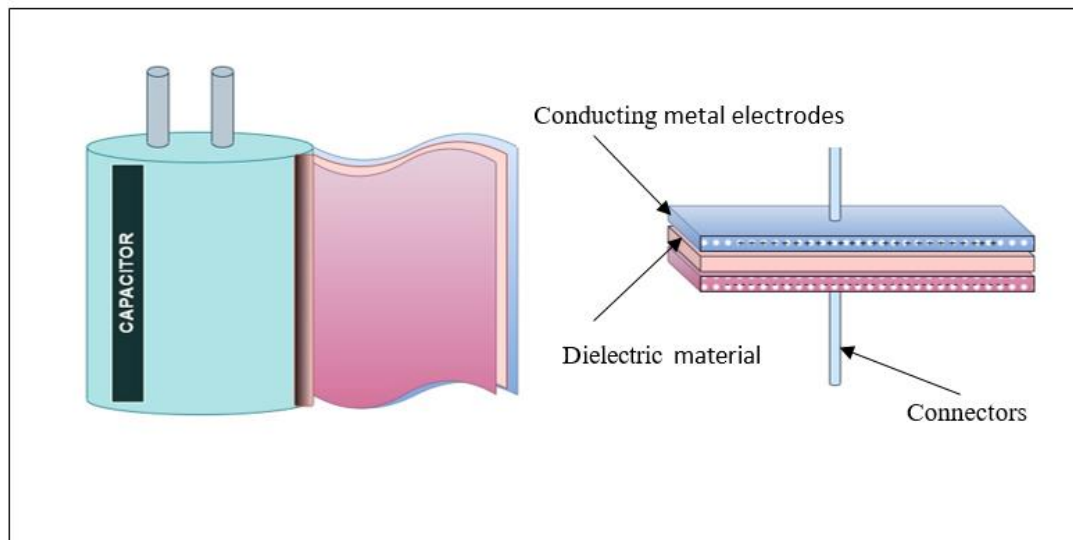

Figure 8: Schematic diagram of supercapacitors

With a higher theoretical specific surface area, greater chemical stability, and superior mechanical properties, graphene and reduced graphene oxide (rGO) have also been used in energy storage systems like capacitors and supercapacitors. Properties such as high surface area, exceptional electrical conductivity, and extreme mechanical strength make it an ideal candidate for use in capacitors and supercapacitors. Graphene-based supercapacitors have been shown to have higher energy and power densities compared to traditional supercapacitors while making them attractive for use in a variety of applications. At present extensive research on graphene-based supercapacitors is ongoing, and scientists are exploring ways to further improve the energy density while reducing their production costs to make them commercially viable (116).

B. Wang et.al (2016) demonstrated the development of a high-performance graphene-based supercapacitor using a new method that involves a blade-coating technique to make GO films, for constructing graphene-based supercapacitors in a scalable way (117). This method resulted in achieving high capacitance, owing to the 3D structure of graphene sheets in the electrode, allowing efficient energy storage. J. Yin et al. (2018) reported on highly conductive and flexible thin film electrodes for supercapacitor applications based on silver nanowires-wrapped carbon fiber networks (118). The researchers induced the rGO layer on the Ag Nano-wires wrapped carbon fiber networks to increase the active surface area,

resulting in fast accessibility of electrolyte ions into the microporous structures, for use in flexible supercapacitor applications. Gao et.al 2012, published the article high-performance asymmetric supercapacitor based on graphene hydrogel and nanostructured MnO_2 (119). In this study, an asymmetric supercapacitor with high energy and power densities has been developed by using graphene hydrogel (GH) with 3D interconnected pores as the negative electrode and vertically aligned MnO_2 nano-plates on nickel foam as the positive electrode in a neutral aqueous Na_2SO_4 electrolyte. The resulting device showed excellent electrochemical performance, with high energy density and power density. Arkhipova et.al. 2021, Nitrogen hetero substitution in graphene nano-flakes has also been discovered as an efficient method to enhance the performance of supercapacitors with ionic liquid electrolytes (120). They have developed a graphene-based supercapacitor using Nitrogen-doped graphene nano-flakes with a high nitrogen content with a non-aqueous electrolyte. The device demonstrated that introducing nitrogen into the graphene layers roughly increased the electrode's specific capacitance when compared to nano-flakes that weren't doped. M.S. Arsha et al. in 2023, reported binder-free supercapacitor electrodes by hydrothermal deposition of thermally reduced graphite oxide on Nickel foam (121). The device exhibited 81.5 % capacitive retention of over 1000 charge-discharge cycles.

Jun Mei et al. in 2015, synthesized nanostructured MnO2/nitrogen doped graphene (MnO_2/N-RGO) hybrids by a facile and economic method at low temperatures with ammonia pre-treatment as electrode materials for supercapacitors (122). They have compared MnO_2/graphene (MnO_2/RGO) hybrids. The novel hybrid exhibited higher capacitance value and enhanced cyclic stability illustrating that ammonia pre-treatment is effective for nitrogen atom doping and the hybrids are potential as electrode materials for supercapacitors. Overall, all studies demonstrate the ongoing R&D in the field of graphene-based supercapacitors, and the potential of these devices to play an important role in future energy storage.

6. GRAPHENE IN FUEL CELLS

The fuel cell converts chemical energy into electricity through electrochemical reactions (123). It is considered a promising technology to solve global energy as well as environmental problems (124). A typical fuel cell contains two electrodes, an electrolyte, and a photon exchange membrane shown in figure 9 (125). It has been classified according to the electrolyte type in the cell recognized as phosphoric acid fuel cell (PAFC), polymer

electrolyte membrane fuel cell (PEMFC), alkaline fuel cell (AFC), molten carbonate fuel cell (MCFC), and solid-oxide fuel cell (SOFC) (126). High energy conversion efficiency, low operation temperature, low emission, high energy, and power density are the significant advantages of using fuel cells (127).
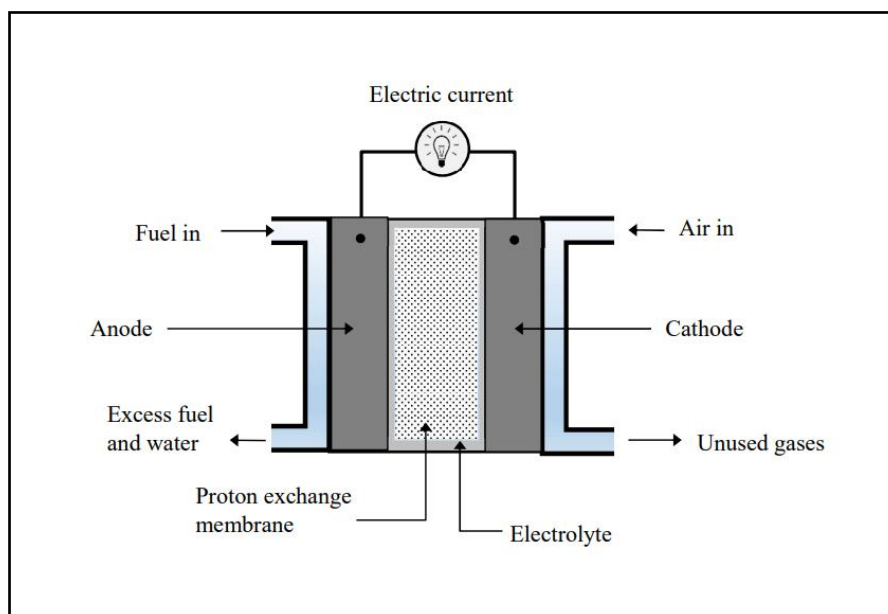

Figure 9: Schematic diagram of a fuel cell

The discovery of graphene and the related materials have been widely used for fuel cells, due to their extraordinary properties such as high electrical conductivity, larger electrochemical active surface area, corrosion resistance, and high thermal stability during the electrolysis process (128). Graphene and its derivatives have been investigated for photon exchange membranes, electrodes, and electrolyte in fuel cells.

Commercially available fuel cells use Nafion as an effective proton exchange membrane because of its excellent thermal and mechanical stability (129). To enhance the efficiency and reduce the production cost in fuel cells, much research has been conducted using graphene and its derivatives combined with Nafion. In 2012, Ravi Kumar et al. fabricated Graphite oxide (GO) / Nafion composite membranes and found high power density which was much better than Nafion (8). Peng, K et al. have reported a method for chemically bonding graphene oxide with Nafion for proton exchange membrane in fuel cells (130). They have investigated chemically bonded Nafion on graphene oxide through atom transfer radical addition reaction and the Nafion nanocomposite photon exchange membrane has exhibited a 35–40% increase in the fuel cell performance (131). Chien et al. have reported a

photon exchange membrane for fuel cells with Nafion engineered with sulfonated Graphene oxide. They have used the modified Hummers method to prepare Graphite oxide from flake graphite. The membrane showed higher selectivity via the steric hindrance effect reducing the ionic clusters (132). In 2017, a modified cathode with titanium dioxide and graphene has been fabricated by M. Mashkour et al. They synthesized graphene using a modified Hummers' method to investigate the catalytic activity of the cathode and reported significant improvements in microbial fuel cell performance (133). Furthermore, an innovative, chemical strategy for the synthesis of GO–polymer composite membrane has been reported by Bong Gill Choi et al. They claimed that GO–polymer composite membranes achieve a high proton conductivity and effective methanol rejection, leading to high-performance fuel cells (134).

Platinum is the most widely used electrode for fuel cells (135). High production cost limits the commercialization of hydrogen fuel cells with the platinum electrode. Researchers have made several attempts to reduce the cost of electrodes using alternatives to expensive platinum. In one of the studies, Tsang, C et al. demonstrated a process to prepare Pd-Pt loaded graphene aerogel deposited on a nickel foam plate as an electrode for an alkaline direct ethanol fuel cell under room temperature. It has been observed that the possibility of producing a size customized metal loaded graphene aerogel electrode for fuel cells with high performance (136). Yan, Z. H et al. have investigated the performance of the catalytic activity of Pt-Co alloy particles on reduced graphene oxide in microbial fuel cells. They claimed that Pt-Co/graphene can be an excellent high-performance catalyst in microbial fuel cells with the advantages of easy fabrication; lower cost and high power production (137). Gyen Ming et al. have reported a scalable technique used to develop catalysts for hydrogen fuel cells. They have shown that graphene-based catalyst was more durable than commercially available catalyst comparing their performance (138). They have prepared homogeneous nanoparticles consisting of high catalytic activity and remarkable stability. In most of the research, GO has been synthesized by using the modified Hummers method. In contrast, charged graphene dispersion has been synthesized via the "metal-ammonia" method and achieved a scalable synthesis for Pt nanoparticle-coated graphene where $PtCl_2$ is reduced directly by negatively charged single layer-graphene sheets in solution.

7.   CONCLUSION

In the present review, detailed production methods of graphene and its composites for potential application in energy storage devices are discussed since it has become a major

need on a personal, domestic, and industrial basis. There are top-down and bottom-up methods used to prepare graphene and graphene composite for various applications. Physical and chemical exfoliation are common methods followed in the synthesis of graphene. Physical exfoliation through the scotch tape method produces pure graphene, however mass scale production of graphene is infeasible by this method. Alternatively, chemical oxidation of graphite to GO followed by reduction of GO mostly generates reduced graphene oxide (rGO) instead of graphene, in which complete reduction of GO to graphene is not possible. Still, the method is popular in the synthesis of graphene and composite in a mass scale for various applications. The use of graphene as an anode in Li-ion batteries has resulted in higher specific capacity, and rate capability despite the high irreversible capacity loss during the first few cycles which limits the practical application of graphene in Li-ion batteries. A similar limitation exists when graphene polymer batteries are concerned in terms of performance. A great deal of further work is necessary to use graphene in Li-ion and Li-polymer batteries. Supercapacitors have emerged as a promising class of energy storage devices with their unique electrode materials and electrolytes. Supercapacitors bridge the gap between traditional capacitors and batteries, offering higher energy density than capacitors and faster charge/discharge rates than batteries. The advancements in electrode materials with graphene based materials, have significantly contributed to the improvement of supercapacitor performance, including energy density, power density, and cycling stability. Furthermore, Graphene and graphene-based materials have been investigated for photon exchange membranes, electrodes and electrolyte in fuel cells. Most researchers have been followed the modified hummer's method to synthesis of GO and graphene. The devices have been showed well performance than without graphene. Up-to-date R&D work reveals that a great deal of further work is still necessary to commercialize graphene-based energy storage devices.

Acknowledgments

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