Review:

Surface Properties of Graphene and Graphene Oxide Aerogels for Energy Storage Applications

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Received: October 10, 2023
Accepted: January 7, 2024

DOI: 10.22146/ijc.89639

Abstract: *This review is mainly on the relevance of graphene aerogels for energy storage systems highlighting their distinct properties and applications. Today, electronic devices such as smartphones, laptops, and other electrical appliances have become the axe of our daily lives. As a result, electrical energy is required for these devices. Despite the discovery of renewable energy sources as an alternative to fossil fuels, the construction of energy storage systems is still necessary to store energy. Lithium-ion batteries and supercapacitors are considered essential systems for this purpose and have witnessed tremendous development in recent years. The efficiency of these systems depends on the structure of the materials used in their formation. Graphene oxide and graphene aerogel materials improve the properties of energy storage systems in terms of stability of charging and discharging cycles, longevity, and reduction of combustion incidents resulting from ordinary compounds. However, the development of graphene aerogels faces challenges in improving their mechanical properties, the cost of their preparation, and their high agglomeration ability in solvents. Therefore, intensive efforts are needed to develop these materials for a new revolution in energy storage.*

Keywords: *energy storage; graphene aerogel; graphene oxide; lithium-ion batteries; supercapacitors*

■ INTRODUCTION

Rechargeable energy storage units are the heart of handled electronic devices, smartphones, and laptops in particular [1]. Commercial use of lithium-ion batteries was introduced by Sony in 1991 when the company was looking for ways to increase the lifespan of the batteries in its surveillance cameras, which constituted a breakthrough in this field. Lithium-ion batteries offered promising solutions for energy storage and also paved the way for a revolution in the production of the electronic devices we use today [2]. Many contemporary electronic products, including cellphones, laptop computers, and electric toothbrushes, are now powered by lithium-ion batteries. We are expected to rely mainly on this type of battery in our daily lives [3-4]. Lithium-ion batteries are used in electric vehicles as a substitute for fossil fuels, reducing the use of raw materials and thus reducing environmental pollution from fossil fuels [5]. However, they become an essential renewable energy source that provides electric energy around the world. It is anticipated that enormous amounts of batteries are needed [6]. Lithium-ion battery sales have reached more than seven billion units worldwide, which is expected to increase to more than 15 billion units by 2027 [7].

Despite the fantastic advantages of lithium-ion batteries such as energy density, high quality, longevity, and low warranty, they also have some defects [8]. Its ability to retain and store energy for long periods will decrease with repeated charging, so its performance will decrease [9]. The heat of the battery is due to the phenomenon of thermal escape, where the excess heat promotes the chemical reaction that makes the battery work and thus generates more heat and enters a spiral of disasters [10]. These defects prompted scientists and researchers worldwide to develop new types of batteries to avoid these defects and thus increase the battery's efficiency [11]. Perhaps one of the most prominent of these solutions is the use of a new group of materials in the manufacture of these batteries. Graphene is expected to achieve unparalleled success in the manufacture of batteries due to its peculiar and unique qualities [12-13].

The Nobel Prize in Physics was given to André Geim and Konstantin Novoselov in 2004 for their work demonstrating that carbon in such a flat shape has exceptional properties in quantum physics, which served as the foundation for graphene's initial candidacy [14]. Graphene can be defined as a group of carbon atoms that are tightly bound together in a structure hexagonal or similar to a honeycomb, as shown in Fig. 1 [15]. This is the essential thing that distinguishes it, as it's thickness of one carbon atom. Its two-dimensional structure makes it possesses many distinctive features [16]. It can conduct thermal and electrical conductivity even higher than copper. Since it has a high ability to conduct thermal conductivity, it will reduce the battery's temperature by dissipating the resulting heat, thus increasing the battery's efficiency and allowing it to be used at high temperatures [17]. It also has a high degree of strength and flexibility in addition to its low weight, which is about 5 times less than the weight of aluminum. These properties make graphene a suitable material for many various electronic industries [18]. When graphene is used in the manufacture of batteries it will store current as in regular batteries, but what distinguishes it is that it can charge and discharge at a very high speed. Therefore, graphene is the promising hope for obtaining a battery that is more sustainable, more efficient, longer in life, faster in charging, and thinner in shape [19].

However, despite all these advantages, graphene-based materials are still limited in the spread because they possess some disadvantages such as their high production cost and very high electrical conductivity, which makes them difficult to turn them off in electrical appliances. Additionally, the fact that graphene is easily agglomerated in the solvent leads to reducing the accessible surface area and impeding ion diffusion [20-21]. Many changes to the graphene surface and the construction of dimension-tailored functional graphene structures were undertaken to prevent the abovementioned flaws. Among these are graphene quantum dots (0D), graphene fibers (1D), graphene sheets/films (2D), and graphene gel (3D) [22]. Perhaps the most important of these materials are 3D graphene-based materials that possess many advantages, including ease of preparation and high efficiency, as well as chemically modified 2D graphene sheets stacked with large pore 3D structures through various techniques can be referred to as materials; graphene-based 3D GBMs which can be classified into hydrogels and aerogels (sponges or foams) [23-24]. This review focuses on using graphene aerogel in electrochemical energy storage devices. It illustrates the material's properties, production process, and applications, but before we delve deeper into graphene aerogels, it is essential to understand the concepts of hydrogel and aerogel.

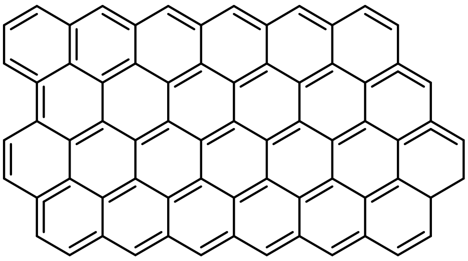

Fig 1. Illustrates the graphene structure

■ HYDROGEL

Hydrogel is a three-dimensional crosslink structure with a low level of crosslinking, usually composed of either natural materials or synthetic materials such as polyvinyl alcohol, sodium alginate, polypyrrole, and chitosan [25]. Hydrogel is characterized by the ability to absorb a large amount of water because its structure contains many hydrophilic groups, and this crosslinked structure includes many physical and chemical bonds and thus leads to a hydrogel structure [26].

Hydrogel has many distinctive properties, which make it suitable for many applications. For example, it has good stretching properties and it consists of thousands of strands, which makes it very stable and does not need to spend a lot of energy on stretching [27]. Its flexibility and high viscosity also characterize hydrogel, and it contains a lot of hydrogen bonds through which can deposit many different substrates, which makes

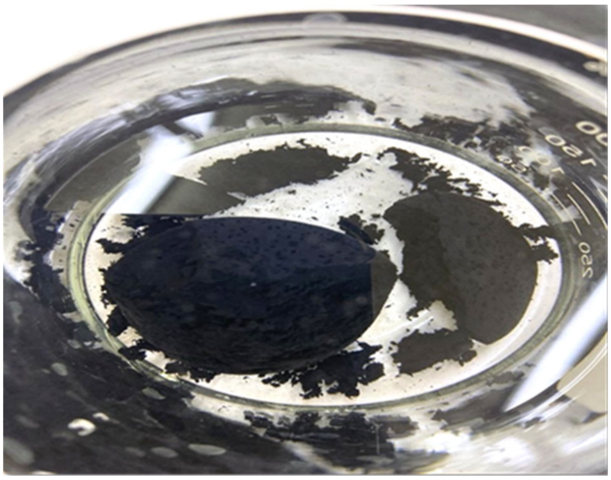

Fig 2. Graphene hydrogel

it suitable for many applications [28]. Hydrogel can also work in harsh conditions and at different temperatures by combining it with anti-freeze materials. All these properties make it suitable for manufacturing electronic devices based on hydrogels [29]. Fig. 2 shows graphene oxide (GO) hydrogel.

AEROGEL

The aerogel can be defined as a hydrogel, but instead of the porous network retaining water, it will contain the gas instead, making it a solid frame filled with air and very light in weight [30]. Therefore, the aerogel was derived from hydrogel by replacing water molecules with air or other gas [31]. Some materials can be used to make aerogels, for example, polymer-based aerogels result from drying polymer hydrogels to form polymeric aerogels, and the properties of aerogels vary depending on the base material from which they are prepared [32]. Common preparation methods are freeze-drying and supercritical drying [33]. These aerogels are characterized by having a vast surface area estimated at 250–3,000 m^2/g. These aerogels usually lack a crystalline structure, so they are very fragile and are characterized by their high transparency [34]. The aerogel formation process contains three basic stages, i.e., gel formation, solvent exchange, and drying process to obtain the aerogel [35]. The first stage is considered essential in determining the exact composition of the aerogel. In contrast, the second and third stages are essential to maintain the final shape of the aerogel [36]. Aerogels can be synthesized using some materials, such as graphene, semiconductors, transition metal oxides, carbon nanotubes, and silica. The aerogel's properties depend mainly on the raw materials and the properties from which it was prepared [37-38].

Aerogel shows some distinctive physical and chemical properties, making it essential in energy storage and transfer device applications. Aerogel has a high surface area and a sizeable porous structure, which contributes to the freedom of movement and transfer of ions and thus makes it well-suited in manufacturing energy storage devices. These advantages also make aerogel well-suited for all solar cell applications and in the manufacture of batteries [38-39].

Aerogels can be classified based on several methods, depending on the preparation method into aerogel, xerogel, cryogel, and hydrogels [40]. It can also be classified based on its physical state into powder, film, and monoliths [41]. Also, the aerogels can be classified into organic, inorganic, and superimposed aerogels based on their composition [42]. At the same time, organic aerogel is derived from carbon, carbon nanotubes, graphene, and polymers, while inorganic aerogel is derived from transition elements, metals, and metal oxides [43]. The composite aerogel is derived from carbon compounds, metals, and metal oxides [44]. Fig. 3 shows GO aerogel.

GRAPHENE OXIDE

In 1859, GO which is considered the oxidized form of graphene, was discovered. It was found before the

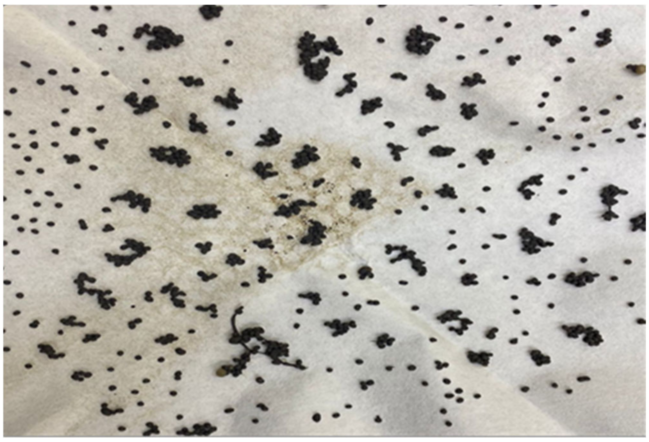

Fig 3. Graphene aerogel

discovery of graphene, but GO remained on the sidelines until it was found [45]. Then, GO gained great importance as one of the sources of graphene synthesis. From this point, studies on GO expanded, which led to the widening of its applications [46].

GO is usually synthesized through two basic approaches. The first is a top-down approach, which treats graphite using strong oxidizing agents such as sulfuric acid and potassium permanganate, followed by an exfoliation step through sonication [47]. The second approach includes a bottom-up approach, which uses the chemical vapor deposition (CVD) method [48]. Fig. 4 shows GO-based materials. Through the treatment processes of the graphite surface and structure, which consists of *sp*^{2} hybridization, will be disrupted, which will lead to the graphite acquiring different oxygen-containing functional groups such as epoxy, hydroxyl, and carboxyl groups [49], as shown in Fig. 5. The oxidation process of graphite leads to the separation of graphite layers within the structure. In contrast, the exfoliation process separates the graphite layers to obtain a solution of homogeneous GO layers [50].

■ GRAPHENE AEROGEL

Two-dimensional graphene is essential for many applications due to its large surface area of about 2,630 m^2 g^{-1} and outstanding chemical stability [51]. Despite these distinctive properties of graphene, the accumulation between the graphene layers due to strong π-π bonds and van der Waals forces reduces this surface area. Therefore, researchers found that converting the 2D graphene into a 3D form will prevent this aggregation and increase the surface area. Therefore, we can make the most possible use of these useful properties represented by high thermal resistance, electrical conductivity, specific capacity, excellent mechanical strength, and high flexibility [52-53]. Due to these unique and distinctive properties, the applications of graphene aerogel have expanded, as it has become used in sensors and capacitors, in the manufacture of lithium-ion batteries, in electric cars, vacuum cleaners, mobile phones, and portable laptops, and in the field of cleaning the environment from pollutants. Researchers have found that graphene aerogel compounds are materials with a high ability to adsorb contaminants from water and air due to their exceptional features, including a large surface area and high porosity, which facilitate the process of combining with other groups and thus enhance the diffusion of ions and molecules [54-55]. In addition, the presence of many functional groups containing oxygen

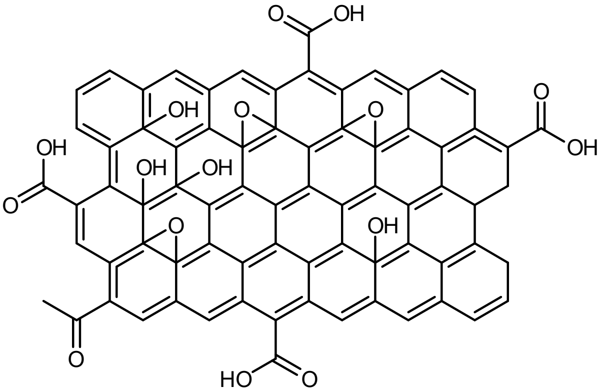

Fig 5. Graphene oxide structure

Fig 4. Graphene oxide (GO)

in graphene aerogel, such as epoxy groups, hydroxyl groups, and carboxyl groups, facilitates the process of its association with metal ions and thus facilitates the process of adsorption of contaminated metal ions from the environment [56]. As mentioned earlier, graphene aerogel has voids that can be filled with air or other gaseous substances instead of a solvent. This property makes it very effective in adhering to and capturing pollution particles within its pores, facilitating the process of purifying the environment [55-56]. Therefore, graphene aerogel is a solid material with high mechanical strength that can bear a weight 6,000 times greater than its natural weight and is extremely lightweight, even lighter than air [54]. As a result of these distinctive properties, graphene aerogel has become one of the most promising materials in the manufacture of capacitors, lithium-ion batteries, and many electrical devices. We mentioned above that graphene aerogel is characterized by high electrical conductivity. Still, it is worth noting that this electrical conductivity quickly decreases due to the contact resistance between the graphene layers. Therefore, to avoid this problem, conductive polymeric materials are combined with graphene aerogel, which will improve the structure of the graphene and its electrical conductivity. It is also possible that using active metal oxides improves the structural structure of the graphene aerogel and increases its electrical conductivity, thus making it ideal for manufacturing batteries such as lithium-ion batteries and supercapacitors [58].

There are many ways in which graphene aerogel can be prepared, such as self-assembly of GO, hydrothermal reduction method, chemical reduction method, crosslinking method, CVD method, sol-gel method, ice template, or sol-cryo [56]. GO is considered one of the most important basic materials in the construction of graphene aerogel because it contains many functional groups containing oxygen that can covalently bond with many different compounds, thus producing a new class of materials with unique properties for many different applications. Fig. 6(a,b) shows the shape of graphene aerogel before and after grinding, respectively.

Flexible Graphene Aerogel Manufacturing Techniques

Direct self-assembly of graphene oxide

This technique is considered one of the most important for preparing aerated graphene. GO is a precursor for preparation because it contains carboxyl and hydroxyl groups [59]. The GO segments in the stable suspension of GO will repel each other due to the electrostatic forces, and van der Waals forces of the functional groups present in the GO, and the loss of balance between these forces will make the GO segments interfere with each other [60]. This will lead to the formation of hydrogel and then drying it using freeze-drying. We will have a three-dimensional porous structure, so the process of forming graphene hydrogel depends on the interference between different forces such as electrostatic forces, van der Waals forces, hydrogen bonds, dipole forces, and others [61].

Hydrothermal-reduction-induced self-assembly

Also, this method is considered one of the effective

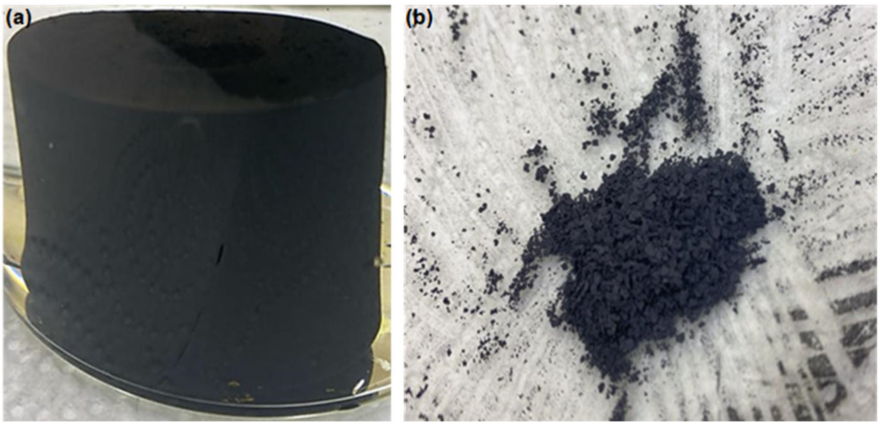

Fig 6. Shape of graphene aerogel (a) before and (b) after grinding

techniques in preparing graphene hydrogel by removing all the hydrophilic functional groups present in the GO sheets, forming reduced GO (RGO) with an increase in the π-conjugated that enhances the bonding of the RGO to create a three-dimensional structure [62]. The scientist used this technique to prepare graphene hydrogel at high pressure, where he was able to obtain graphene hydrogel containing 2.6% RGO by weight and had tremendous mechanical strength [58,63]. Many efforts were also devoted to finding out the effect of the pH of the initial dispersion of GO on the physical composition and mechanical properties of graphene aerogel during the technique of hydrothermal reduction, where the other scientist prepared the graphene aerogel through the dispersion of GO at different pH values, where they noticed that there were differences in the porous structure [64]. The total size of the pores and the surface chemistry in addition to the morphology of the nanosheets, where it was noted at pH 11, the graphene nanosheets would become twisted and bent with less compaction and larger pore size. In comparison, the graphene nanosheets will become flat at pH 3 with more compaction due to the removal of oxygen from the structure [65]. Although the graphene aerogel resulting from this technology is characterized by using a high degree of purity and is prepared without additives, high temperatures and high pressure are among the most critical challenges that face this technology and make it of limited use [66].

Chemical reduction method

This technique is considered one of the most distinctive methods for preparing graphene aerogel compared to hydrothermal, as the experimental conditions used in the chemical reduction method are moderate. The temperature is less than 100 °C atmospheric pressure, in addition to its low cost compared to the hydrothermal method. Perhaps some of the most essential reduction factors used in this method are ethylenediamine, sodium bisulfite, hydrogen iodide, and hydrazine [52-53]. All oxygen-containing functional groups in the GO sheets will be eliminated during the chemical reduction process, boosting the -interaction between the neighboring GO sheets and forming a three-dimensional network [67]. Controlling the reducing agent will also impact the self-assembly behavior of RGO, ultimately changing graphene aerogel's structural and mechanical characteristics [68].

Crosslinking method

Crosslinking prepares graphene aerogel through the self-assembly of the dispersed GO through the bonding between the strong covalent bonds and the weak non-covalent bonds of the GO sheets as in the bonding of electrostatic forces and the interaction of hydrogen bonds and the π-π interaction that leads to the formation of 3D graphene aerogel. The crosslinking of the chemical covalent bond provides a stronger interaction than those mentioned above weak ionic bonds which improves the structural stability of the graphene-based aerogels [69-70].

Chemical vapor deposition (CVD)

This method is considered one of the most widely used methods in preparing graphene aerogel because it preserves the basic properties of graphene. For instance, Chen et al. [71] prepared graphene aerogel by using nickel foam directly in the CVD process, where initially, a thin layer of graphene was deposited on the surface of nickel foam by the decomposition of methane at a temperature of 1,000 °C in the presence of hydrogen and argon gases. Before starting to prepare a nickel foam mold, a support material is deposited on top of the graphene film, usually poly (methyl methacrylate) is used, which can be removed later by using acetone, and all sheets of graphene in the prepared graphene foam are well-bound in together [72-73]. In the chemical evaporation technique, it is possible to increase the number of sheets of graphene in the aerated graphene by increasing the concentration of methane, and this leads to a change in the electrical conductivity and surface area, as the graphene generated in this way is characterized by its lightweight and high electromagnetic performance in addition to its high electrical conductivity with a tiny percentage of disadvantages [74]. Despite the advantages of graphene aerogel prepared in this way, this method faced some difficulties related to high temperatures and the use of some dangerous reagents to remove the template, which made it limited in application [75].

Sol-gel

The sol-gel technology is considered one of the simplest and easiest techniques used in the preparation of graphene aerogel. This technique is sometimes called wet chemistry because it initially consists of sol and then ends in the formation of a 3D crosslinked structure that resembles a gel [76]. This technique usually consists of three basic steps, as shown in Fig. 7. The first step involves hydrolysis for precursors, as water and sometimes a small percentage of alcohol are used to accelerate the hydrolysis process. The second step is the step of digestion and condensation, where in this step, the viscosity of the solution will increase and, as a result, a colloidal solution will be formed. The final step is drying and heat treatment, and sometimes purification of the solution is needed before the drying step [77].

Usually, the formation mechanism of the gel begins with the aggregation of particles in the form of clusters inside the solution, which includes using some organic solvents [78]. Therefore, graphene aerogel is formed by removing these solvents. This technique takes several factors into account, such as the components of the solution, the temperature of the solution, the pH of the solution, the nature of the precursors, the solvents used, stabilizing agents, surface tension-reducing agents, as well as the additives used for drying, since these elements have a significant impact on the final gel's characteristics [79-80]. This technique was used for the first time by Worsley et al. [81] to make RGO aerogels, where the work team suggested that the aqueous dispersion of GO by using resorcinol and formaldehyde are used with sodium carbonate, where the RGO aerogels were obtained with high conductivity of about 10^{-2} S m^{-1} compared to the conductivity of structures that contain only physical properties and whose conductivity is within the limits of 0.5 S m^{-1}. They also noticed a significant increase in the area surface of the graphene aerogel prepared in this way. Newly, Huang et al. [82] prepared graphene aerogel/TiO_2 materials by using the sol-gel method, as shown in Fig. 8. The results showed that due to the effect of porous separation, the TiO_2 particles would be loaded evenly in the graphene aerogel with an increase in the

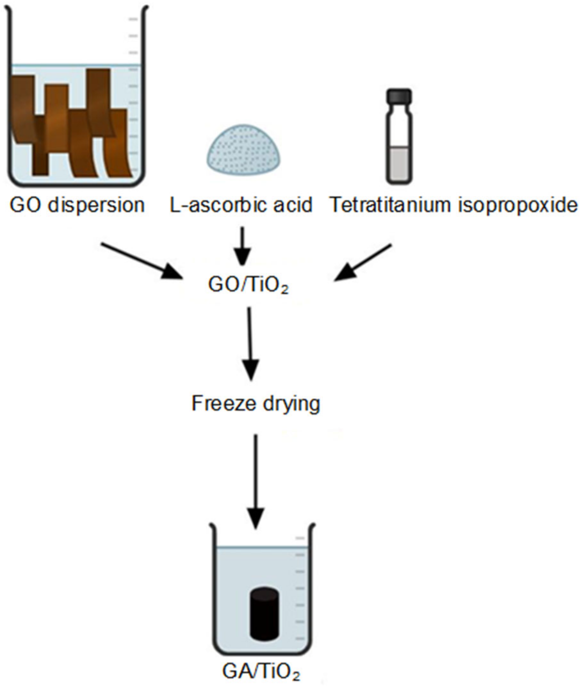

Fig 8. Sol-gel preparation of graphene aerogel/TiO_2 materials

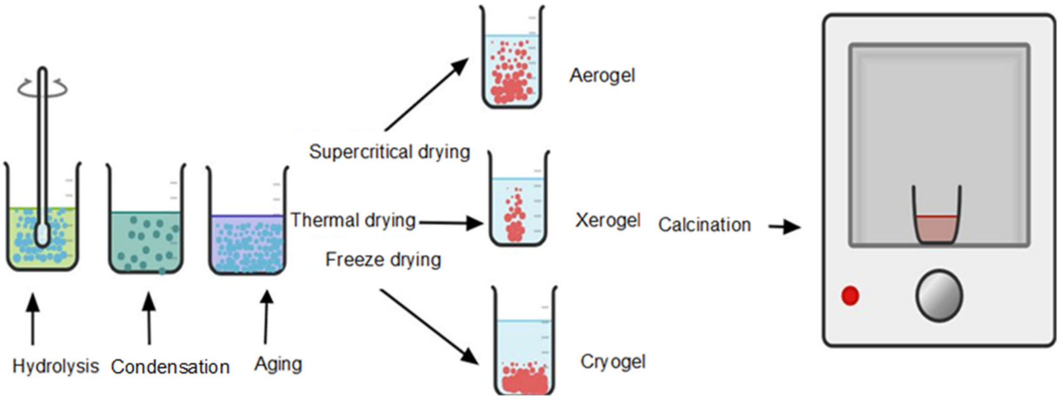

Fig 7. Sol-gel method

concentration of graphene. This equal arrangement of TiO_2 nanoparticles with graphene aerogel resulted in obtaining a material with excellent mechanical and frictional properties, excellent corrosion resistance and high thermal conductivity.

Ice template or sol-cryo method

This method is considered one of the common and innovative methods for the preparation of graphene aerogels because this method is free of chemicals. This method designs the morphology of graphene aerogels through the growth of ice particles in a freeze template [83-84]. The use of different freezing methods will lead to control of the pore size and the shapes of the ternary 3D compounds resulting, and perhaps one of the essential freezing methods used in the preparation of 3D structures is air freeze-drying, directed freeze-drying, non-directed freeze-drying, and others [85]. The shapes and sizes of 3D structures are also affected by the type of template used in the freezing process, Wang et al. employed a directed freeze-casting method at a low temperature to obtain highly porous aerogels. GO is poured into fast-moving ice crystals to be collected inside these ice crystals. It was noted that the graphene aerogel prepared in this way has a significantly low density that reaches 0.23 mg/cm^3 and has high porosity, excellent electrical conductivity, and a vast surface area of about 432 m^2/g [86]. The random structure graphene aerogel can also be prepared by immersing the GO suspension directly in liquid nitrogen and then freeze-drying [87]. Air drying is a high-speed, easy, and inexpensive technology at the same time due to the possibility of its application in ambient, experimental conditions and the fact that it does not require special equipment. Geng et al. [88] presented a simple, easy, and inexpensive green preparation method for the preparation of graphene aerogel by combining both one-step reduction techniques, freezing, thawing, and air drying.

3D printing method

It is considered one of the modern technologies used to prepare aerogel polymers and composites in 3D and lightweight structures by assembling the building units in layers one after the other to build the required model [89]. This technology was used to manufacture cellulose aerogels, nickel and cobalt nanocomposites, and composites with honeycomb and pyramidal structures [90]. An excellent example of this method was performed by Choudhury et al.in the preparation of a graphene/poly imidazolium-based composite aerogel by using a 3D freeze-printing technique where a group of polymers was used to prepare stable graphene dispersions in many different solvents [91]. In this work, the presence of the polymer did not lead to the stability and enhancement of the properties of the graphene nano-inks not only extruded but also served as a binder to construct graphene nanosheets in a 3D graphene/polymer composite aerogel. Thus, the 3D printing method is very appropriate for preparing high-resolution graphene-based materials on graphene [92].

Graphene-Based Materials/Polymer Composite Aerogels

Polymeric foams are considered one of the types of aerogels. They have great importance in our daily lives because of their distinctive characteristics, such as high porosity and lightweight, which have led to the possibility of using them in thermal insulation, packaging, and even sound insulation. However, their use is still minimal due to the large size of the pores and the low number of functional groups they have [93]. In recent studies, it was found that the addition of the polymer to the graphene aerogel significantly improves its properties [94]. As mentioned, graphene aerogel has many good characteristics, such as great mechanical strength and high thermal and electrical conductivity [96]. The latter depends on the size of the graphene sheets, where the more significant the length of the graphene aerogel sheets, the greater its mechanical strength and vice versa [95]. Also, the mechanical properties depend on the functional groups that contain oxygen and are present in the graphene oxide sheets used to prepare graphene aerogel [96]. They work to reduce the bond between the sheets of graphene aerogel, which gives it an unstable structure. Therefore, removing these groups containing oxygen through annealing at high temperatures will increase the efficiency of the graphene aerogel [97]. Fig. 9 shows graphene aerogel/TiO_2/sodium alginate nanocomposite.

Graphene Aerogel and Energy Storage

Lithium-ion batteries are regarded as a breakthrough in energy storage and transportation technologies. They helped to launch a revolution in the electronic devices we use today [8]. Still, they are not without some defects represented in their low ability to retain energy and store it for long periods with repeated charging and the decrease in their performance when the weather is very hot or very cold [9]. In addition, concerns have been raised about the safety and security of using these batteries and their impact on the environment [10]. Lithium-ion batteries may burn or explode in certain circumstances, and the extraction of the metals used in their manufacture may cause severe damage to the environment [98]. To avoid these defects, a new class of materials, most notably graphene aerogel, has been introduced to manufacture these batteries because it has many unique properties that we mentioned [99]. Previously and below, we will review the mechanism of action of the graphene aerogel in energy storage.

The electrodes made of graphene aerogel have been of great significance in improving the performance of lithium-ion batteries because of their distinctive properties [54]. As it has high electrical conductivity, which allows electrons to be transported with high efficiency within the electrode, the movement of electrons in the charging and discharging operations will become fast, and since graphene aerogel has a relatively high surface area, allowing for a significant number of active sites for lithium-ion adsorption and desorption during battery cycling. As more lithium ions can be accommodated, this improves energy-storage capacity, leading to superior battery performance [58].

Furthermore, graphene aerogel is characterized by its high flexibility and excellent mechanical strength, which makes it more able to withstand expansion and compression in volume associated with the movement of lithium ions in each cycle, and this leads to maintaining the integrity of the electrode and prolongs the life of the battery [55], as shown in Fig. 10. Graphene-based aerogel compounds showed a high ability to improve the performance of lithium-ion batteries, as these compounds work by providing active

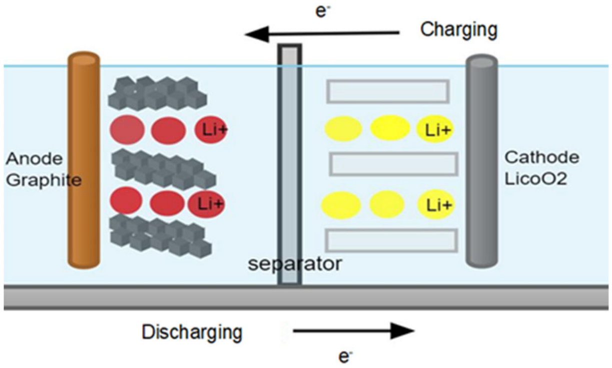

Fig 10. Principle work of graphene-based electrodes for LIBs

Fig 9. Graphene aerogel/TiO_2/sodium alginate nanocomposite

sites to receive lithium ions through their vast surface area [98]. In addition, these compounds offer a high ability to quickly transfer electrons during charging and discharging due to their high electrical conductivity [58]. Yoo and his partners [100] used graphite and graphene in the manufacture of lithium-ion batteries to improve the characteristics of the lithium-ion battery and increases its energy storage capacity. When using graphite, the storage capacity is about 372 mAh/g. In comparison, the capacity of graphene is about 540 mAh/g for a combination of carbon nanotubes, graphite, and graphene. The change in capacity of about 730 and 784 mAh/g, respectively. This demonstrates the significance of producing appropriate graphene composites that boost the distance between layers and prevent aggregation, thus increasing the efficiency of the lithium-ion battery [101].

Some graphene composites have also been developed to improve their structural properties by increasing the surface area and electrical conductivity, reducing the phenomenon of aggregation, and reducing its production cost, thus increasing the efficiency of lithium-ion batteries [102]. SiO_2/graphene composites showed a high reversible 1127 mAh/g capacity at 0.2 A/g with 87% capacity retention after 200 cycles. Another instance of GO/black-arsenic-phosphorus/CNT composite showed stable capacities of 1286 and 339 mAh/g at current densities of 0.1 and 1.0 A/g, respectively, with a capacity of 693 mAh/g after 50 cycles [103]. It is worth noting that the thickness of the electrodes has a significant impact on the properties of the electrode itself as the traditional electrodes are characterized by the fact that they consist of stacking many layers of active materials [104], and therefore, the thickness of the electrode will increase. The efficiency of transferring charge and ions will decrease. Still, the electrical conduction is ineffective and impedes the diffusion of ions, which requires a longer time for charging discharge and thus reduces the battery's efficiency [105].

Graphene-based composites are also used as anodes in LIBs. A MoO_2/graphene composite was made by Bhaskar et al. [106] using a one-pot solution method for application in Li-ion batteries as a high-performance anode material. Also, Zeng et al. [107] prepared the porous composite Co_3O_4 saturated with nitrogen/graphene derived from anthracite (Co_3O_4/AG) through a self-assembly technique and using heat treatment. He noticed that the uniform distribution of Co_3O_4 nanoparticles on the porous graphene assisted in obtaining a high surface area of about 120 $m^2\ g^{-1}$ and absorbing a large amount of nitrogen doping (5.4%). The Co_3O_4/AG electrode showed high reversible capacity (845 mA g^{-1}) and good cyclic stability (510 mA g^{-1} after 100 cycles). Tang et al. [108] used the hydrothermal method to prepare a composite of silicon and graphene aerogels with a 3D porous structure. The specific charge capacity of the composite was stated to be higher than 1330 mAh g^{-1} at 0.2 A g^{-1}, even after 100 cycles. Shen et al. [109] prepared $SnO_2/NiFe_2O_4$/graphene composites as anode materials for lithium-ion batteries. He achieved a high storage capacity by adding SnO_2, which facilitates the movement of Li^+ ions. In contrast, graphene added a very high surface area, and this nanocomposite showed a high reversible capacity of up to 731.5 mA/g, with a current density of 200 mA/g after 50 cycles. A graphene-coated $MnCO_3/Mn_3O_4$ nanocomposite was prepared by Chen et al. [110] by using the one-step method. This composite showed excellent electrochemical performance, with a capacity of about 2457.4 mA g^{-1}, high cycling stability (93.3% of the capacity of the second cycle after 200 cycles), and high-speed capability (605 mA g^{-1}, even at a high current density of 5 A g^{-1}). A unique sandwiched mesostructure was fabricated by Liu et al. [111], where graphene was incorporated between the layers of V_2O_5 to form a V_2O_5@graphene@V_2O_5 cathode. This demonstrated a full electrode basis capacity ≈203 mAh g^{-1} after 2,000 cycles. YongJian et al. [112] presented a simple and easy preparation method of graphene slurry at 1% concentration and lysed by using mechanical agitation, ultrasonic dispersion and the addition of different dispersing agents where the initial system charge/discharge capacity was measured as 1273.8/1723.7 mAh/g, and the efficiency was Coulomb 73.9%, at a current of 100 mA/g.

Through all the experiments mentioned above in the use of graphene in the manufacture of electrodes, the "graphene battery" proved that the energy storage system made of carbon materials exclusively was and still is a promising technology for the future. Lithium-ion batteries are not the only exciting area of energy storage. There's also interest in developing graphene aerogel-based electrodes for supercapacitors and fuel cells. Aside from large energy storage devices, these aerogels can also be used within many other devices, including solar photocatalytic devices, air purification devices, sound systems, and electrochemical sensors [113].

Applications of Graphene Aerogels

Graphene aerogel is an exceptional material with several unique properties that make it appropriate for many applications. The following is a review of the most critical applications of this fantastic material. Graphene aerogel has become the most widely used material by the Apollo and NASA organizations in the manufacture of astronaut suits and energy storage devices [114]. Graphene aerogel began to occupy the first positions in manufacturing batteries, capacitors, solar cells, mobile phones, and other electronic devices by giving them a significant edge, beating out traditional batteries in charging time and lifespan [63]. Graphene aerogels have a promising future in the LED industry. A research team at the University of Manchester managed to manufacture a dimmable graphene-coated filament LED, which reduced energy requirements by 10% [115]. Graphene aerogel particles are considered materials that have a remarkable ability to absorb, making them suitable for environmental cleaning, pollutant removal, and water and air purification [116]. Graphene aerogel has become used in the paper industry to improve the properties of ordinary paper, characterized by fragility and ease of damage. A research team at the University of Sydney found that paper based on graphene is 10 times stronger than steel and is conductive, durable, and thin enough to be used in numerous industries [117]. Graphene aerogel is used as an attractive material in flame retardancy, thermal insulation, and fire alarm applications, thereby eliminating all the drawbacks of commercial fire alarm equipment. Graphene aerogel can be used in the production of thermal sportswear and is amazing use of this material due to its high heat dissipation capacity [117].

Challenges on Graphene Aerogel-based Devices

It is known that all electronic devices based on graphene require that the structure of graphene be coherent with a high degree of arrangement of atoms and maintain a consistent structure to obtain unique properties suitable for applications for electronic devices [118]. So, the presence of impurities, the irregular arrangement of atoms, or some defects in the structure will hinder the achievement of the main objective of using graphene aerogel [35]. Among the problems that face graphene-based devices is the method of transferring the graphene product, which may sometimes lead to cracks, breakage, or impurities entering the structure of the graphene. Therefore, it became necessary to find an effective method of transfer to avoid these problems, for example, making the graphene grow on the surface of another substrate [119]. Perhaps one of the biggest challenges facing the use of graphene in electronic industries is the high production cost of producing large quantities of graphene, which depends on the type of substrates and energy sources used in preparing graphene [120].

■ CONCLUSION

In this review, we focused on graphene aerogel, which is the lightest material in the world and is known for its large surface area and honeycomb-like structure. This material was discovered in 1995 and is composed of air. It is characterized by its low density and high porosity, which makes it very suitable for many diverse applications such as supercapacitors, batteries, cleaning the environment, and manufacturing astronaut suits. Despite these unique features and enormous potential, it is necessary to conduct more research and experiments in this field and provide the necessary infrastructure for that.

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