Chapter

# Graphene Based Functional Hybrids: Design and Technological Applications

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## Abstract

Because of the versatile chemical, physical, and electrical properties, graphene as well as its nanocomposites are regarded as the backbone of engineering and scientific innovation. Different physical and chemical methods are used to create sustainable carbon materials. Furthermore, fabrication methods are employed in order to produce the composites, which are of constituents with desirable properties. Because of their biocompatibility, graphene nanomaterials have enormous potential for improving biology and drug delivery. The proposed chapter provides a variety of fabrication methods for sustainable graphene composites and highlights various applications of graphene. Furthermore, graphene nanocomposites are promising multifunctional materials with improved tensile strength and elastic modulus. Despite some challenges and the fact that carbon nanotube/polymer composites are sometimes better in some specific performance, graphene nanocomposites may have a wide range of potential applications due to their outstanding properties and the low cost of graphene. Because these graphene composites have a controllable porous structure, a large surface area, high conductivity, high temperature stability, excellent anticorrosion properties, and composite compatibility, they can be used in energy storage as electrocatalysts, electro-conductive additives, intercalation hosts, and an ideal substrate for active materials. Meanwhile, the chapter summaries the graphene nanocomposites requirements for technological innovation and scientific applied research.

**Keywords:** graphene, physical properties, polymers, ceramics, technological applications

## 1. Introduction to graphene/polymer nanocomposites

Graphene, as well as its, distinguish variants, possess excellent characteristics. A remarkable achievement for them, because of these traits is their utility as fillers in the fabrication of composites. There are various factors that impart critically in the properties of graphene-polymer composites. They include dispersity of fillers, bonding between matrix and filler, the ratio of graphene filler to the polymer matrix, and quality. However, there are several fabrication approaches for undergoing modification in these factors in order to acquire desirable properties. In general, graphene comprises a very high aspect ratio, therefore it is capable of doing several modifications in properties of composites even at a very low percolation limit.

## 1.1 Fabrication of graphene/polymer composites

The most widely used methods for the incorporation of graphene into polymer matrices are, in situ polymerization, melt intercalation, and solution mixing. These three schemes are having schematic format as shown in **Figure 1**.

### *1.1.1 In-situ polymerization*

Here the scheme, constitutes the mixing of a monomer and graphene fillers, particularly in the presence of a catalyst (s). The stage is then generally followed by polymerization initiation through heat or radiation [1]. The utility of epoxy in in-situ polymerization is an excellent example [2]. However, fabrication done by this scheme yields some polymer/graphene composites like graphene/polystyrene (PS) [1], graphene/polyaniline (PANI) composite [3], and graphene/silicone composites [4]. Significance of this method is the strong interaction between the filler and the polymer matrix. This leads to a rapid stress transfer, enabling a quick formation of the homogeneous dispersion. In addition, fillers constitute homogeneity in their distribution. High filler loading in polymer matrices is also achieved through In-situ polymerization. Meanwhile, the mixture becomes more viscous during polymerization, and the ongoing process suffers difficulties, causing ultimate hindrance for the loading fraction [5]. Usually solvent is the prime option in the current method most of time in some cases, but it extensively raises the need for the removal of solvent [6].

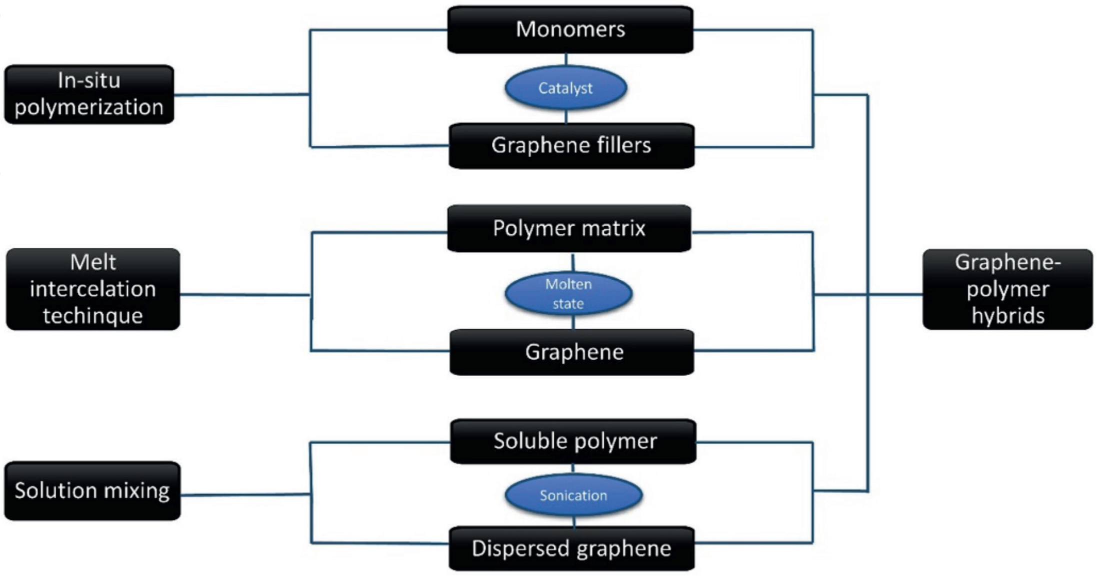

**Figure 1.**
*General fabrication routes for graphene polymer hybrids (GCHs).*

### *1.1.2 Melt intercalation technique*

Melt intercalation method incorporates designing of graphene in the molten state with the polymer matrix, thereby eliminating the need of solvent. High shear mixing with accelerated temperature results in the homogeneity of polymers and graphitic materials mixing. Thermoplastic composites are the most manufactured yield of this method. This method carries some of the graphene/polymer composites including exfoliated graphite/polypropylene nanocomposites [7], polystyrene/graphite nanosheet composites [8], polyethylene terephthalate (PET)/graphene nanocomposites [9], etc. On the contrary, this scheme has some discrepancies too, like poor dispersity and fillers distribution, than rest of methods. In addition, possibilities for breakage of graphene sheets and defects are comparatively much higher than rest of methods [10].

### *1.1.3 Solution mixing*

Fabrication of Polymer composites through solution mixing is the most frequent method. Potentially applicable on a bigger scale, simplicity and the demand of equipment with not much specifications are the main reasons for the common utility of this method. In this method graphene undergoes swelling as if s suited solvent refers solubilization of a polymer. This results in ultimate rise for the curvature of a surface. One of the most demanding parameter is graphene's dispersity and solubility as well. So, the need is functionalizing graphene sheets in a way ensuring ease for dispersion in various solvents. Some composites were having much higher solubility, that results in convenient fabrication like Poly(vinyl alcohol) (PVOH)/graphene oxide (GO) [11]. Sonication or shear mixing generally leads to a homogeneous mixture in many cases, as if the solvent is organic. There must be some definite schemes that alleviates aggregation while graphene sheets are functionalized [12].

## **1.2 Physical properties of graphene/polymer composites**

Exfoliated carbon sheets are usually obtained through rapid pyrolysis or chemical reduction of graphene oxide. However, there are some ways that creates variation in the properties of these carbon sheets. In the following section, we would reveal some of the physical properties of graphene/polymer nanocomposites.

### *1.2.1 Thermal properties of graphene/polymer composites*

Vibrating lattice is the key upon which material's thermal conductivity depends on. Graphene ensures a material to be thermally conductive and stable, because it exhibits excellent thermal conductivity at room temperature ( $\sim 3000 Wm^{-1}K^{-1}$ ). Therefore it imparts substantially in the improvement of thermal properties. Thermally less interface resistance, stiffness and the planar geometry of graphene are the major factors yielding remarkable thermal properties (1266 Epoxy/Graphite).Graphene's utility as transparent conductors attributes to its peculiar characteristics like higher conductivity, and incorporation towards ceramics and polymers [13]. On the contrary, thermal properties are suffered by graphene's orientation and aspect ratio.

### *1.2.2 Mechanical properties of graphene/polymer composites*

Recent discoveries introduced fascinating insights of material science as graphene proved experimentally as nanomaterial. Graphene constitutes remarkable elastic modulus (125 GPa) and intrinsic strength ( $\sim 1.0 TPa$ ). Scientists after having a comprehensive overview of all intrinsic properties of graphene, are now able to manufacture graphene-filled led polymer composites with desired properties. However, other milestones for the betterment include reinforcement phase aspect ratio, distribution in the host matrix, interface bonding, and the reinforcement phase concentration. Noticeable thing is that the increment in properties of polymer matrix attains at low filler loading [14].

### *1.2.3 Electrical properties of graphene/polymer composites*

Electrical conductivity for graphene is its notable attribute, among all physical properties. Conductive graphene itself raises the possibility for elevation in electrical conductivity, when it comes to be utilized as fillers along with insulating polymer matrix. When loading fraction touches certain limit, particularly at a stage of percolation threshold, electrical conductivity increases rapidly right after the formation of network by filler. Electrical conductivity influences by the concentration of fillers as well as by percolation threshold. Conduction takes place through tunneling in polymer layers, so there is no such need for the filler to be in contact with the current flow directly. The limiting factor in thermal conductivity for polymer composites is ultimately the tunneling resistance [15]. Graphene in its other variant generally named as pristine graphene have much higher electrical conductivity, yet mechanical exfoliation scheme for its production on a bigger scale reduces its utility [16].

## 2. Introduction to graphene/semiconductor hybrids

Graphene semiconductor hybrids as well as composites prove to be more fascinated substitute for manufacturing comparatively much lighter, compact as well as effective batteries. If for instance, the concern is to alleviate various issues directing from synthesis routes to the recreatability of a homogeneous component, then the ultimate consequence would be the commercialization of products. Agglomeration rate of graphene matters too. Electrochemical sensors along with biosensors have upgraded their work by these hybrids. These sensors consider to be the best regarding detection of bacteria, viruses etc. [17].

### 2.1 Fabrication of graphene/semiconductor hybrids

ZnO combination with graphene reveals its speciality for better as well as increased performances. Fabrication of graphene semiconductor hybrids can be done by number of ways, like solvothermal method, ultrasonic spray pyrolysis, microwave-assisted zinc ions reduction with graphene as shown in **Figure 2**.

All these techniques possess limited utility because of some issues being unaddressed. These includes cumbersome procedure, equipment's sophistication and economically low feasibility. However, need is to adopt a method having no such issues. Therefore, In situ thermally decomposed zinc dihydrazinate complex on graphene's surface at comparatively moderate temperature is the best choice. Prime leverages for

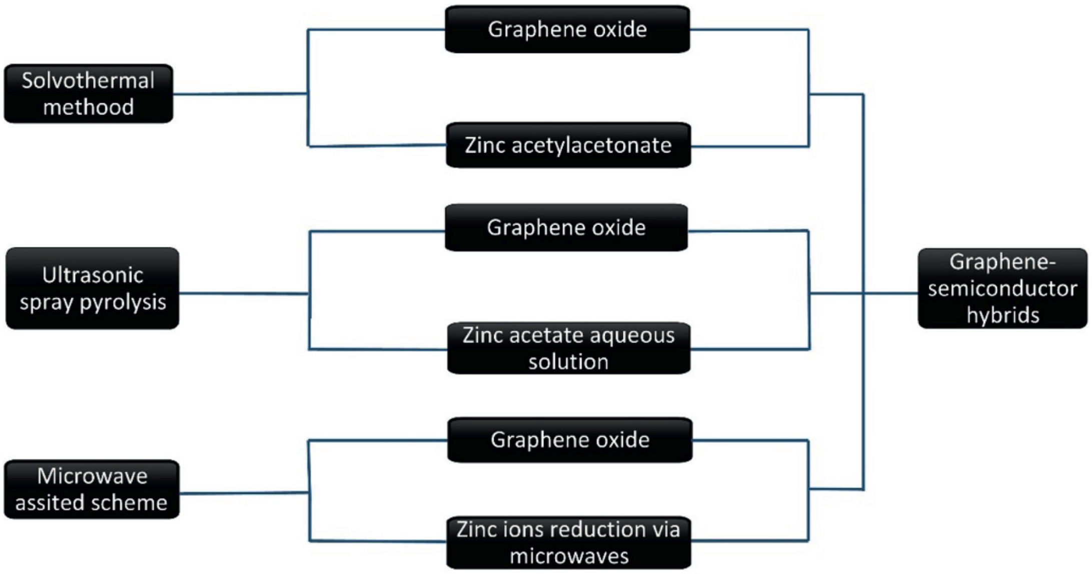

**Figure 2.**
*General fabrication routes for graphene semiconductor hybrids (GSH).*

this new method are its simplistic behavior, uniformity in size of particles, high end product with no much residual impurities [18].

### *2.1.1 In-Situ thermal decomposition*

At first, aqueous solution of zinc (II) nitrate hexahydrate and hydrazinium benzoate undergoes through in situ mixing, thereby yielding a precursor complex. However, molar concentration of these two chemicals is 0.01 mol and 0.02 mol respectively. Both of them should be stirred consistently. However, the final mixture exhibits concentrated product comprising an alleviation in its volume. Then, all constituents should be subjected to drying after cooling, and cleaning process. Now take 1 mg of graphene and disperse it into 5 ml of ethanol, allowing it for stirring of 1 h, similarly 10 mg complex precursor of zinc benzoate dihydrazinate is to add up and repeat sonication process. The decomposition of complex into nanoparticles of ZnO results in their coating on graphene sheets as the transfer of contents to a fine silica pot, undergoes an environment of 200°C temperature for 2 h.

## **2.2 Physical properties of graphene/semiconductor hybrids**

Graphene as well as Zinc oxide exhibits discrepancies up to some extent when considered separately. Therefore, the prime role of this combination mainly leads to the compensation of these flaws. This synergistic combination has been undergone comprehensive investigation, as it proves to be an excellent candidate of future devices, because of the enhancement in most of the properties like chemical stability, electrical etc. [19].

### *2.2.1 Photocatalytic properties*

The interaction of graphene along with ZnO semiconductor, is ultimately vigorous. Therefore, it results in quickest movement of photo-generate negatively charged particles from Zinc oxide to graphene. Yet, another notable activity is the alleviation, for recombination process of these electrons. In general, activity of the photocatalytic process rises, thereby imparting a constructive role in these properties [19].

#### *2.2.2 Electrical properties*

Another big advantage of graphene semiconductor hybrids is particularly its improvement of the electrode materials for supercapacitors. This renovation arises because of the various factors like specific surface area enhancement, outstanding cyclical stability, elevation in energy as well as power density, better ionic and electrical conduction performance. ZnO particles as well as graphene enclosed porous carbon have much improved conductivity of electrode entirely due to their combination. In addition, electron's movement from ZnO to the layer of graphene is the manifestation to the alleviation of defect emission. On the contrary, movement of charge from graphene to defect state of the given semiconductor causes an enhancement of defect levels contribution [19].

## 3. Introduction to graphene ceramic composites

Graphene is an excellent alternative ensuring the availability of composites having remarkable traits. Primarily its exceptional mechanical strength, results in potentially favorable supporting constituents for ceramic composites. Graphene is fascinatedly electrically conductive and this proves its versatility for various applications. Prime challenge for graphene's utility lies in its homogenous implantation into the ceramic matrices. Therefore, to address such challenges major milestone is its processing scheme. Herein the following sections will reveal number of processing schemes as shown in **Figure 3** for graphene based ceramic composites [20].

### 3.1 Fabrication of graphene/polymer composites

There are various segments upon the peculiar characteristic of these composites rely on. These includes fine particulates, equiaxed shape ensuring better packing, and homogeneous graphene's distribution into the ceramic matrix. However, another big challenge during investigation of toughening process, has been the fabrication of composites having well supervised micro/nano-structures [21]. Nowadays, appreciations for the sophisticated fabrication process are far more than conventionally adopted powder processing methods. These complex processing techniques comprise, colloidal processing scheme, sol–gel method, and polymer derived ceramic method as shown in **Figure 3**.

#### *3.1.1 Colloidal processing method*

Colloidal processing method exhibits intimate spread of ceramic as well as graphene matrix forming composites. These composites comprise microstructure that are homogenized and having properties ultimately controlled by colloidal chemistry. In colloidal processing technique, graphene is normally covered with ceramic fragments by the colloidal suspensions. This alters its surface chemistry, thereby alleviates repulsion between graphene, causing homogenous spread of graphene into ceramic grains. However, variation in surface chemistry consequences homogenous dispersion, that preserves itself even after sintering. Generally, for the demand of

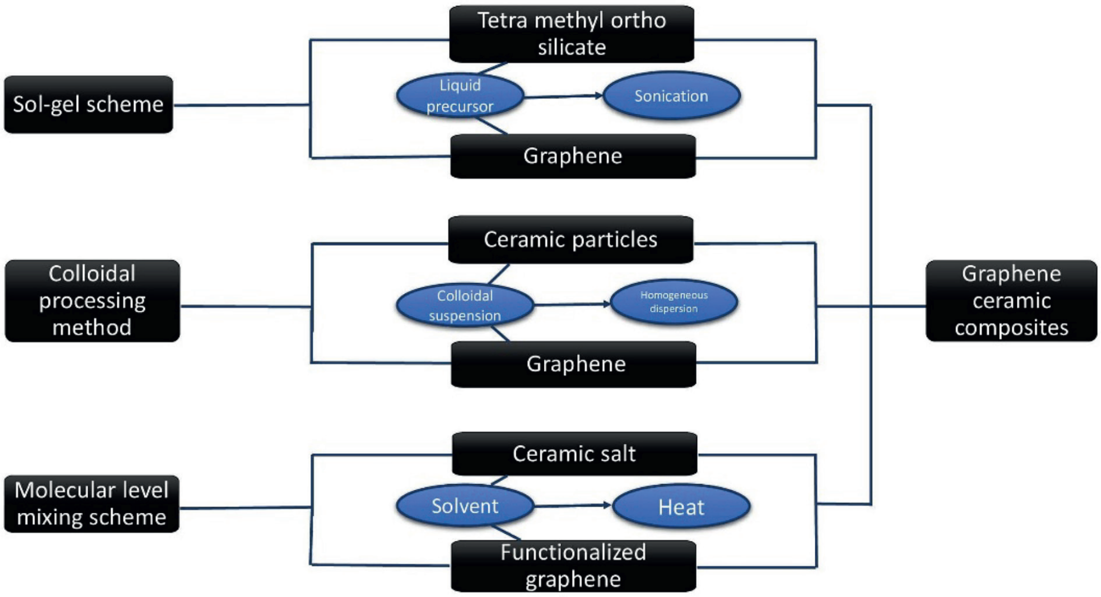

**Figure 3.**
*General fabrication routes for graphene ceramic composites (GCCs).*

homogenous spread, both these materials should be dissolved into the same kind of solvent. Although magnetic stirring is another key factor that causes slow mixing and imparts positively for the uniformity in spread of constituents [20].

### *3.1.2 Sol: gel method*

Sol–gel method comprises a substitute for the supply of dispersion of ceramic as well as graphene composites. Herein, graphene undergoes dispersion through a suitable molecular precursor ((TMOS) tetra methyl ortho-silicate), thereby leading to a condensation process for reinforcement. Homogeneous sol is readily available as the suspension of both the molecular precursor as well as graphene encounters sonication. In general, the method mainly ensures availability of nanocomposites of silica. Although, the technique suits well for fine spread, aggregation for precursor limits its utility [22]. However, sol–gel method need liquid precursors only, thereby providing ease for the processing of well-dispersed composites [23].

### *3.1.3 Molecular level mixing method*

Molecular level mixing scheme encapsulates another way for the production of ceramic and graphene composites. In this method, ceramic salt as well as functionalized graphene is combined in a suitable solvent. This then subjects its conversion to ceramic constituents via proper heat, thereby results in molecular level coating [24]. However, peculiar characteristic of this method includes excellent interface bonding between graphene and ceramic at molecular state. As a result of this strong bonding, a substantial enhance in the properties of the resulting composites becomes easier. Traditional methods of processing graphene-ceramic composites result in fall of mechanical properties. This occurs specifically owing to the effect of aggregation, caused by intermolecular forces [25]. Although, sol–gel method proves to be an excellent candidate for the homogenous dispersion of graphene into the ceramic matrix, however interfacial bonding between graphene and ceramic lacks stability [26].

## 3.2 Physical properties of graphene ceramic composite

Combination of graphene along with ceramic to form composites yields excellent physical properties, like mechanical, electrical and thermal.

### *3.2.1 Mechanical properties*

Ceramic graphene composites believe to be mechanically strong, when subjects to certain appropriate conditions. Yao et al. [27] analyzed that size of grains and fracture toughness are interconnected by each other. Their idea was that increasing the size more than 0.5 $\mu m$ predominately effects fracture toughness. Therefore, alleviation in fracture toughness of composites like AWTG2 as well as AWTG3 can be elaborated on the basis of the elevated growth in ceramic grains. Low concentration of ceramic composites results in number of pores, that results in substantial reduction of the interfacial bonding between graphene and ceramic grains [28]. However, graphene tends to pull-out because of this weak interfacial bonding, thereby enhances fracture toughness.This proves to be the ultimate cause for better fracture toughness of AWTG4 clears in (**Table 1**) [29].

### *3.2.2 Thermal properties*

Thermal conductivity suffers by various factors, and sintering at different pressure is one of them. At low temperature, thermal conductivity is proportional to the square of the temperature, reflecting amorphous like character. This happens because of various scattering phenomenon, especially phonons scattering by pores [30]. On the contrary, at high temperature conductivity rises at different rates, lightly for samples pressed by pressure of 5 GPa, while strongly for pressed at 4 GPa. For a high temperature range with increasing pressure, there is an unusual decrease in the value of thermal conductivity. Porosity as well as quality of graphene and ceramic composites are the of the main concern [31].

### *3.2.3 Electrical properties*

Electrical properties of graphene ceramic composites can be revealed on the basis of percolation theory as shown in **Figure 4**. Generally, in this theory threshold in percolation magnitude attributes to critical filler constituents, where increase in electrical conductivity results because of the presence of several conducting paths for electrons. These conducting paths of electrons do not stand if percolation magnitude lacks certain transition limit. Therefore, in order to ensure the availability of conducting chains in ceramics, percolation threshold must be much higher than threshold

| Sintered samples of (graphene-ceramic composites) | Fracture toughness |
|---|---|
| AWTG2 | 5.24 ± 0.17 |
| AWTG3 | 5.64 ± 0.21 |
| AWTG4 | 5.95 ± 0.23 |

**Table 1.**
*Sintered composites vs. fracture toughness.*

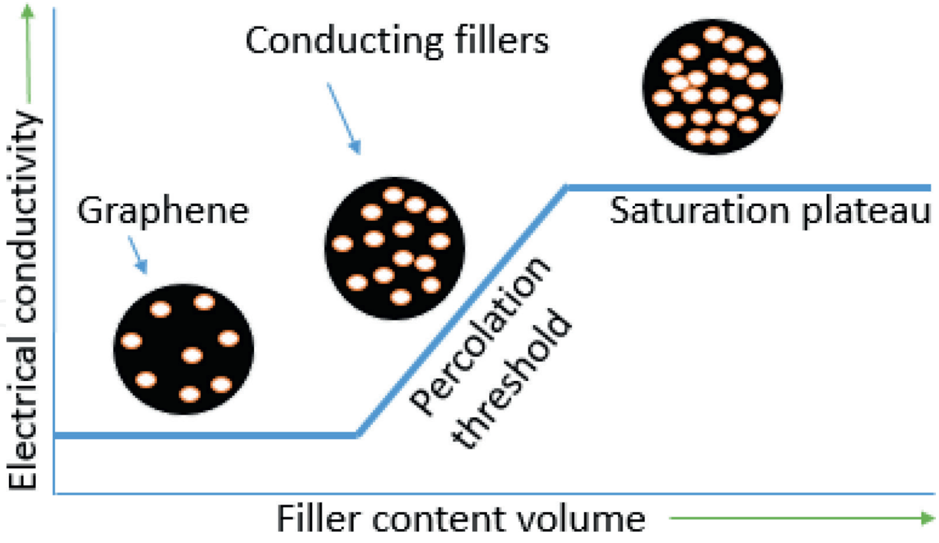

**Figure 4.**
*Electrical conductivity and percolation phenomenon as a function of filler volume fraction in graphene based ceramic composites.*

value. Hence, number of these electron paths causes an elevation in electrical conductivity of graphene ceramic composites [20].

## 4. Application of graphene

Comprehensive research in graphene's physical as well as chemical properties, has created a revolutionary in fields of electronics, energy storage devices, corrosion coating, etc. [32].

### 4.1 Drug delivery

Exploring various domains of graphene like biological one for enhancing its utility in biomedical field results as scientist's consistent effort in recent years. Free drugs leave defects intrinsically that comprises burst discharge, low blood circulatory span, irrelevant targeting. Therefore, it causes low therapeutic efficiency, lesser feasibility and variety of complexities [33]. Drug delivery server (DDS) is the ultimate need for alleviating these drawbacks. These DDS's have been used in various versions for last several decades as shown in **Figure 5**.

It simply encapsulates drug and commutes it to the desired destination, results as a controllable therapeutist [32].

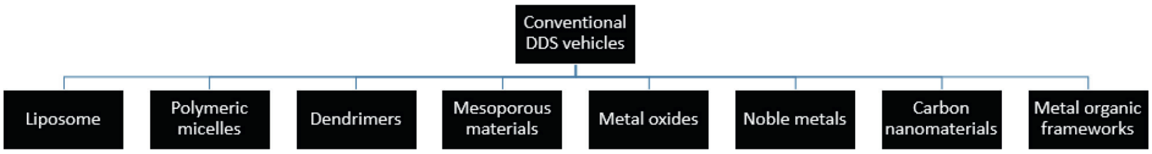

**Figure 5.**
*Traditionally used DDS vehicles from several decades.*

### 4.2 Electronics

Graphene's utmost demand for electronics results owing to its hike in the physical properties. High current capacity of transportation and various experimentally analyzed properties having numerical values are shown in **Figure 6** [34].

Radio-frequency transistors and analog electronics are ultimately the very finest tasks driven by graphene. It is because graphene comprises exceptional rise in its trans-conductance, material's stability and thinness. In addition, there is no need of switching off the devices entirely irrespective of its capability to do so, in radio-frequency transistors. For instance, in the process of signal's amplification in a single amplifier,the transistor is usually remains in the on condition [35]. Electro-absorption modulator is another utility as gate field intrinsically adjust graphene's Fermi level [36]. On-chip optical interconnections demand ultimately a very high bandwidth modulator, large speed and tiny footprints. Graphene having single layer causes slight light's absorption as the interaction of graphene with light is substantially rigorous. This problem is generally resolved by the coupling of graphene with a silicon waveguide. It creates an elevation by 0.1 dB $\mu m^{-1}$ of 1.35–1.60 $\mu m$ at frequencies greater than 1 GHz [36]. The notable benefit of graphene-based modulators is its sustainability in integrated form along with Si-CMOS electronics. One of the most dominant edge of graphene is its saturated absorption, that reveals particularly decrease of absorption light as a function of increase in light intensity. Saturable absorbers therefore, helps to turn continuous wave output into ultrashort light pulses. Picosecond laser pulses can be generated by graphene due to its peculiar traits like higher stability, quick decay and a broad absorption range [37].

### 4.3 Flexible and stretchable display

Flexible and stretchable display is the utmost need of the future electronics, and graphene is doing its job elegantly in this domain [38]. Beside remarkable properties, mineral resources consider so far unsuccessful for rigid components, and this limits their utility. On the contrary graphene and its variants are emerging candidates in flexible as well as in stretchable electronics [39]. Various distinguished traits ensures its availability in modern devices like LEDs and for conversion and in energy storage devices as well as shown in **Figure 7**.

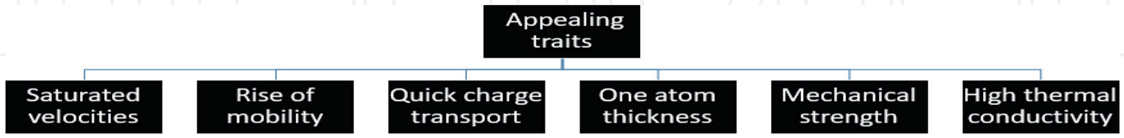

**Figure 6.**
*Fascinating characteristic of graphene in electronics.*

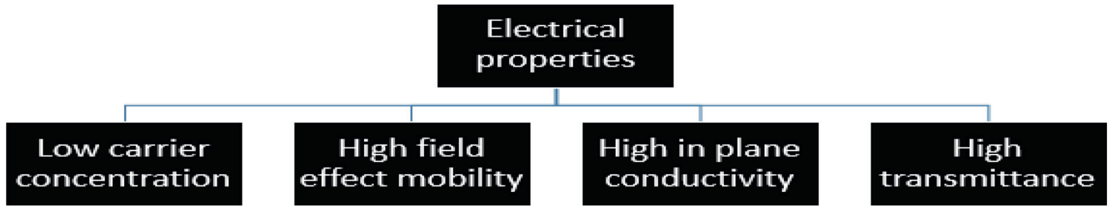

**Figure 7.**
*Excellent characteristic for flexible and stretchable displays.*

On the contrary, most utilized material like Indium tin oxide, exhibits wide range of discrepancies such as high corrosion rate, much higher refractive index, and less economic feasibility etc. [40].

### 4.4 Energy storage devices

Application for energy storage devices also attributes to graphene. This is due to the Ultimate reduction in restacking of graphene sheets specifically when interacting with metal oxides. The prime logic for graphene's utility in this arena of energy storage devices is its highly conductive nature, especially when it comes to interlinked network. Mechanically as well as electrochemically stable along with porosity in the structure all proves its significance in this current domain of application. Photovoltaic cells, batteries and fuel cells entirely rely on energy storage devices. For supercapacitors, ensuring material for electrode is graphene's job. The process of dysenetization can also be performed by these electrode material [41].

### 4.5 Corrosion coating

Graphene inherently behaves as a corrosion resistant, whenever coated. Penetration power constitutes a substantial increase in polymer coating. However, graphene enters along thickness of coating into the medium, and anti-corrosion process initiates right after the entrance of graphene to the interface. Carbon atoms in graphene exhibits sp2-hybridization, comprising high electron density, that renders all incoming corrosion molecules [22]. Hence, such kind of fascinating structure in graphene leads to its durability. In addition, graphene holds an excellent protection shield in the infiltration stage due to its capability of prolonging the path. Therefore, durability as well as corrosion resistance can be enhanced remarkably.

## 5. Applications of graphene based hybrids

Graphene based hybrids have a major role in food microbiology, where the microbes are applied on the Nano-scale level to prevent the packaged food from being rotten and it gives the food a long lifetime. Graphene based hybrids have been used in nano robotics. Nano-robotics is an emerging technology that creates machines or robots whose components are at or near the scale of a nanometer. Moreover, graphene based hybrids have been extensively used in drug delivery, food sciences, nano-optics, nano-energy, photo-voltaic, nano-chips, light-weight mechanical and electronics applications. Recently, Billions of dollars are being spent to research the use of nano-materials for the detection and treatment of diseases like chemotherapy and the manufacturing of minuscule sensors. Miniscule sensors can be injected into our body which can monitor our vitals even more closely than a doctor. Graphene based sensors are one of prominent addition in recent times for detections of diseases. Graphene based hybrid sensors can be manufactured in large quantities at a very low cost. These sensors are used to monitor the health of frameworks such as bridges and nuclear reactors as well as in the industries.

In addition, graphene-polymers based materials are trying to replace conventional materials in food packaging. And Nano-sensors are also developed to detect the presence of contaminants and dirt in the food particles respectively.

Nanotechnology provides a range of options to improve food quality and helps in improving food quality taste. Nano-encapsulation techniques have been used to improve the flavor of food.

## 6. Conclusion

Because of the unique mechanical, thermal, chemical, optical, and electrical properties, graphene and its nanocomposites have gotten a lot of attention in scientific research in recent years. Graphene with its nanomaterial, such as graphene polymer composites, graphene semiconductor composites and graphene ceramic composites are regarded as the foundation of material science and technological innovation. These nanomaterials are created using various physical and chemical methods to produce high-quality materials with excellent properties. Graphene nanomaterials are also used in electronics, organic photovoltaics, energy conservation technology, and drug delivery, among other things. In the future, these advanced materials could be used to create a variety of materials with various applications. A great deal of research is being conducted in order to produce these materials on an industrial scale. Because of their ability to store energy on a large scale, these materials are the future of sustainable energy production and storage devices. Fuel cells are also expected to replace battery-powered energy systems in the near future. Graphene sheets have the potential to revolutionize microelectronics. The demand for graphene nanocomposites will continue to rise as a result of technological advancement.

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