Review of Electrical Properties of Graphene Conductive Composites

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Abstract
Conductive composites include any composite having significant electrical conductivity. When an electrically conductive filler such as carbon-based materials like graphene, is added to insufficient quantities of a polymeric resin, a conductive composite is formed. when the filler content in the composite is increased, the conductive fillers particles start to contact each other, and form a continuous path that make the free electrons travel easily and conduct the electricity. These conductive composite materials have lightweight, resistance to corrosion, and can be easily adapted to meet the needs of a specific application. Therefore, these conductive composites may replace the metals in some applications. The applications of conductive composites include, for example, electromagnetic interference shielding materials, coatings, sensors, batteries, electrodes. This review paper focuses on carbon-based materials such as graphene, as a conductive nanofiller, and it provides a review of the properties and synthesis of graphene. Then, the paper provides a comprehensive review of electrical properties of conductive graphene composites, and how the graphene is utilized to improve and enhance the electrical conductivity of some polymers such as epoxy resin to be used in more applications.

Keywords: Conductive composite, carbon fiber, graphene, polymer, electrical conductivity, conductive ink.

1. Introduction:

In recent years, the interest in composite materials has increased as a new engineering material due to its unique properties. Where it was used prominently in structural applications that requires lightweight and high resistance to corrosion. These materials have proved success in many applications (space, transportation, civil and military industry, agricultural industries, medical industries, etc.) [1, 2]. Composite materials can be defined as a material composed of two or more different substances that share their properties, to form new material with new properties, where there is no chemical reaction between these substances and each substance retains its basic properties. Composite materials consist of reinforcing material that have high strength and stiffness, such as glass fibers and carbon fiber, immersed in a matrix material such as polymer or resin. The matrix material bonds the fibers together, and also transfers the shear stresses to the reinforcing material. Composite materials have outperformed traditional structural materials (which made from metals) in many ways. For example: have more durability than traditional structural materials, easy to form complex shapes with different sizes, light weight without affecting the strength, exposed to low fatigue rates, high thermal resistance (for ceramic mixtures), larger resistance to corrosion and weather conditions, a huge resistance to the spread of cracks that may occur as a result of vibration, and therefore, they are excellent to be used in the rotational axes. All these reasons have pushed composite materials to the top of the list and made them the preferred materials in aircraft construction [3-5].

In the aviation, metals were used to shield the electronic systems in the aircraft, to protect them from the electromagnetic interference (EMI). In addition to that, metal coils are currently used in the installation of aircraft structures to protect the aircraft from lightning strikes. However, these metals are heavy which increase the weight of the aircraft and are also costly. Therefore, aerospace companies tried to find conductive composite materials instead of metals. The advantages of conductive composites are: lightweight, high strength and stiffness, and high electrical conductivity capable of protecting the aircraft and its systems (sensors, integrated circuits, etc...) from lightning strikes and electromagnetic interference (EMI). In addition to that, they can also be used as a heating element capable of melting the ice that forms on the surface of the aircrafts [6-8]. The major focus of this paper is to provide a comprehensive review of conductive composites. A brief introduction to the chemistry and properties of the well-known graphene material, followed by review of the recent literature on graphene conductive composites is provided.

2. Conductive Composite Materials

Recently, extensive research has been performed to produce lightweight conductive composite materials with high electrical conductivity, environmental stability, and good engineering properties. Conductive composite materials would be useful in many applications such as battery components, electrodes, electromagnetic interference shields, circuitry components, lightning strike protection in the aircraft. Conductive composites include any composite having significant electrical conductivity, adding conductive filler to a nonconductive (or less conductive) matrix lead to the so-called electrically conductive composites (ECCs), which exhibit a large scale of electrical conductivities needed for different applications. These composites are characterized by a percolation threshold or a critical value at which the conductivity starts to increase as a function of filler contents, where the electrical conductivity can change by several orders of

magnitude for small variations of the filler content. By controlling the volume fraction of the conductive filler, composites with a nonconductive matrix can exhibit a wide range of electrical conductivities as needed for various applications [9-11]. One example on these composites are conductive polymer composites which have high electrical conductivity, also they have excellent mechanical properties which are possible to replace metals in some applications. The polymer conductive composites are made by adding conductive fillers to the polymers using certain method, producing high electrical conductivity. The electrical conductivity can be modified with a greater range, and these conductive composites can be easily processed. Due to the mentioned advantages, polymer conductive composites can be used in different applications (see **Figure1**), such as electronic devices, energy fields, and chemical engineering [12, 13].
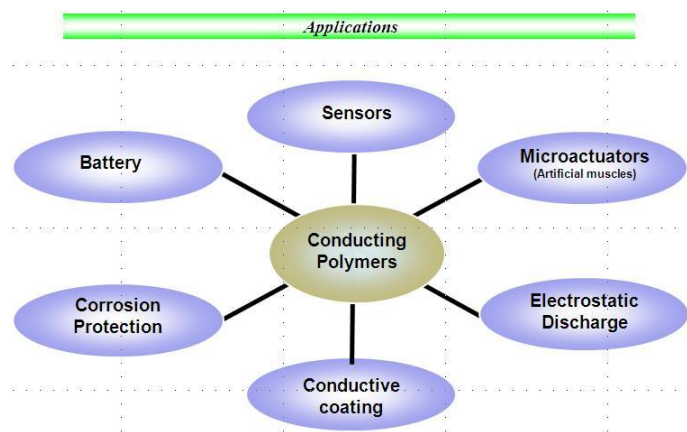

Figure 1: The application of polymer conductive composites [14].

Conducting Polymers are organic polymers that conduct electricity. They are used in many applications such as fuel cells, computer displays and microsurgical tools. Conductive Polymers can be synthesized alone, as hydrogels, combined into composites or electrospun into microfibers. The biggest advantage of conductive polymers is their processability, mainly by dispersion. Conductive polymers are generally not thermoplastics, i.e., they are not thermoformable. But, like insulating polymers, they are organic materials [14, 15]. They can offer high electrical conductivity. Therefore, they have promise in antistatic materials and they have been incorporated into commercial displays and batteries, but there have been limitations due to the manufacturing costs, material inconsistencies, toxicity, poor solubility in solvents, and inability to directly undergo the melting process. Also, they do not show similar mechanical properties to other commercially available polymers [16, 17]. Therefore, the conductive polymer composite that are synthesized by adding conductive fillers such as carbon based materials, are used in more applications nowadays, because they have higher electrical conductivity than conducting polymers, in addition to their excellent mechanical properties. When conducting polymers are combined with carbon materials like CNT, graphite, and graphene, they show good thermal and electrical properties [18,19]

There are three types of conductive fillers used to prepare the polymer conductive composites, carbon, metal and ceramic as shown in **Figure 2**. The carbon based materials include carbon black (CB), carbon fibers (CF), graphite, carbon nanotubes (CNTs), and graphene. Graphene is used frequently to form polymer conductive composites, because it has light weight, forms conductive networks easily, and resist the oxidation [19,20]. The polymer conductive composites commonly used thermosetting and thermoplastic resins as their matrices. The thermoplastic resins include liquid crystalline polymers like: polypropylene (PP), polyethylene (PE), and polystyrene (PS). Polypropylene is a highly available, because it has excellent mechanical properties, good heat resistance, low cost, ease of processing, and full recyclability, while thermosetting resins include epoxy resin, vinyl-ester, and polyester [9, 20, 21]. Thermoplastic and thermosetting resins are insulating to electricity, where their electrical conductivity values are very low as shown in **Table 1**.
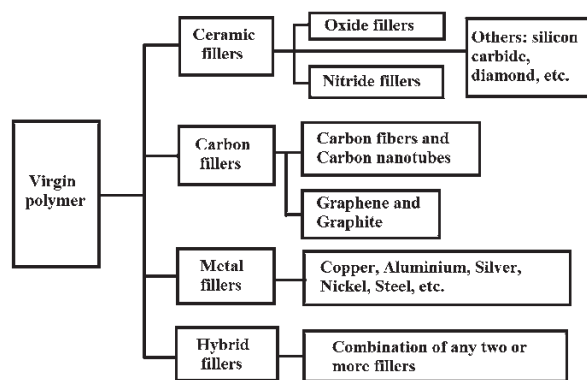

Figure 2: Outline of the different types of fillers used in the preparation of composites [21].

Table 1: Electrical conductivity of thermoplastics and thermosetting resins.

Thermoplastic Resiens	Electrical Conductivity (S/m)
PET (polyethylene terephthalate)	10^{-14}
Polypropylene	10^{-14}
PBT (Polybutylene terephthalate)	10^{-14}
Vinyl	10^{-14}
Polyethylene	10^{-14}
PVC (Polyvinyl chloride)	10^{-15}
Nylon	10^{-14}
Thermosetting Resiens	
Polyester	10^{-13}
Vinyl Ester	10^{-14}
Epoxy	10^{-12}
Phenolic	10^{-13}

The electrical conductivity of a composite is generally characterized by its dependence on the conductive filler content (see **Figure 3**), when the conductive filler is increased to high enough content inside the composite, the conductive path is formed which make the free electrons in the composite to travel easily, and eventually the electrical conductivity will be increased to high levels as shown in **Figure 4**. The formation of this conductive network is based on the principles of percolation theory [22] [23].
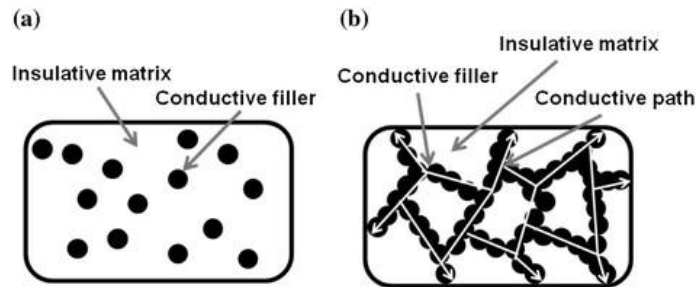

Figure 3: Filler distribution in polymer composite: (a) at low content, (b) conductive path at high enough content [24].
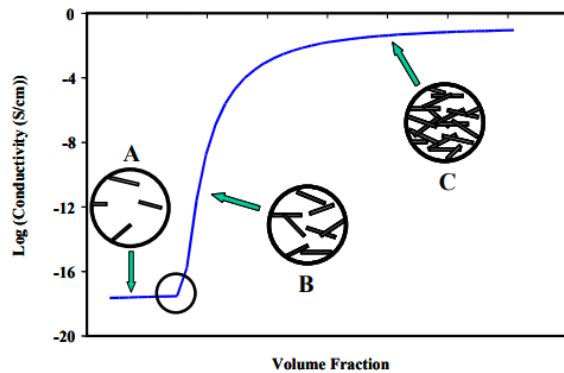

Figure 4: The dependence of electrical Conductivity on the filler content [25].

When the content of the conductive filler increases gradually, the composite material reaches the percolation threshold, where any further increase in the content of conductive filler in the material after this threshold, the electrical conductivity value of the composite material will increase many orders of magnitudes as shown in **Figure 5**, and the composite material becomes a conductive composite material [14, 15, 23]. For example, **Table 2** shows the percolation threshold of graphene reinforced different polymer resins, where the electrical conductivity of these composites increased significantly at this content of graphene. This behaviour is occurred because the conductor filler formed conducting path through the dielectric matrix material, this conductive path is formed when the conductor filler content is higher than the percolation threshold [23, 24].
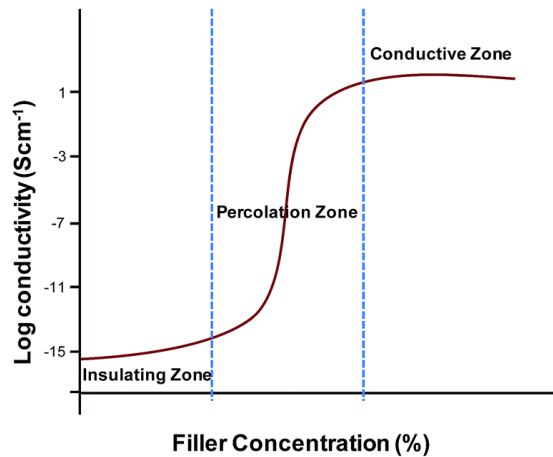
Figure 5: The percolation zone of conductive fillers reinforced polymers composites [23].

Table 2: The percolation threshold of graphene reinforced different polymer resins [14-24].

Material	Percolation Threshold (vol%)
Functionalized graphene filled epoxy composites	0.1
Neat graphene/epoxy nanocomposite	0.53
Graphene/ polyethylene composite	0.07
The graphene/polyethylene terephthalate (PET) nanocomposite	0.47
TRGO (Thermally reduced graphene oxide)	<0.5
Graphite	>2.7
PS/graphene composites	0.1
RGO(reduced graphene oxide)/ PVC/vinyl nanocomposite	0.15
RGO/PET nanocomposite	0.47

By adding conductive fillers to the polymers, materials can be produced and designed with certain properties that are suitable for each application (see **Figure 6**). The electrical conductivity value of polymers commonly can be between 10^{-14} and 10^{-17}. The electrical conductivity values of other materials are 10^2 for carbon black, 10^3 for Polyacrylonitrile, 10^4 for carbon fibers, 10^5 for graphite, and 10^6 for metals such as aluminium and copper. All electrical conductivity values are given in S/cm. The composite materials must have an electrical conductivity in the range of 10^{-12} and 10^{-2} S/cm to be used for conductive applications, such as electrostatic dissipation and shielding [15][23][24].
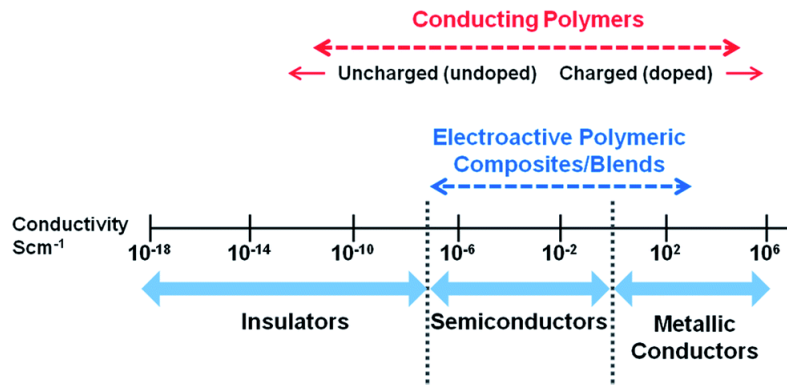
Figure 6: Conductivity range of conducting polymers and conductive polymeric composites [24].

The Electrical conductivity of conductive composites depends on filler type, shape, size, and filler dispersion and distribution in the composite. The conductive filler type has an important effect on the electrical conductivity of the composite. For example, Carbon based materials (CNTs, CB, graphite and graphene) can be used as conductive filler, but the electrical conductivity of each material is different, therefore, the value of the electrical conductivity of the composite will be related to the conductivity of the filler type used. The conductive filler size also effect the electrical conductivity of the composite, by changing the percolation threshold [14, 24]. For example, when the conductive filler shape is spherical and smaller in size, this will lower the percolation threshold. Also when the aspect ratio (ratio of length to diameter, l/d) is greater than one, this will lower the percolation threshold. The dispersion and distribution of filler in the matrix materials (such as filler orientation) also effect the electrical conductivity, of the composites [15]. A composite molding by extrusion and injection can align fillers that have an aspect ratio greater than one in a certain direction due to the flow through the nozzle of the different machines and the Mold. This alignment will produce anisotropic conductivity within the sample, meaning that conductivity will be greater in one direction over another. The surface properties of the filler and polymer also have a significant effect on the conductivity and the percolation threshold of the composite. For example, when the difference between the surface energy of the filler and matrix is small, the higher electrical conductivity of conductive composite is obtained [25, 26, and 27].

Conductive composite materials have been used to improve thermal management systems for transferring the heat. Thermal conductivity of these systems can be increased and enhanced by increasing the surface area and reducing the weight, this helps management systems to operate effectively in the difficult environmental conditions. For example, improved thermal management is needed to increase the power density of electronics and more effectively cool electronic enclosures that are envisioned for future aircraft, spacecraft, and surface ships. [25][28].The metallic materials are frequently used in thermal conductivity applications, such as heat dissipation for cooling semiconductor devices. For these applications, the metallic material typically is machined from bulk metals to form the desired configuration. However, these metallic materials are very heavy, costly to machine and causing corrosion. Therefore, the using of such metallic conductive materials adds severe limitations on heat sink design [29]. To improve heat sink geometries and design, many attempts is made to provide conductive composite materials that include conductive filler such as graphene, to provide the necessary

thermal conductivity as shown in the **Figure 7**. Using such conductive composites, it enables the design of heat sink with more complex geometries [29-31].
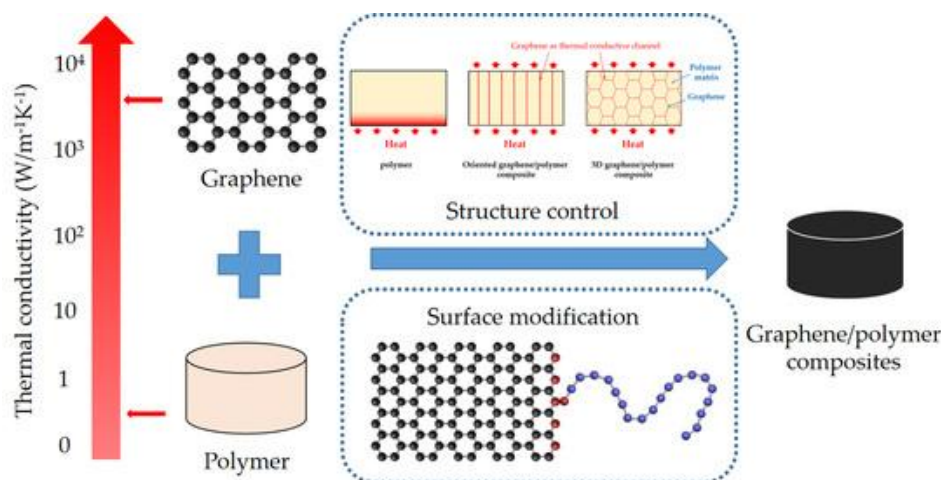

Figure 7: Using conductive composite materials, to provide enhanced thermal conductivity of polymers [30].

3. Graphene
3.1 Introduction
Graphene sheets (see **Figure 8**) — one atom thick, two dimensional layers of sp2-bonded carbon — is predicted to have a range of unusual properties as shown in **Figure 9**. Graphene has high in-plane values of thermal conductivity and mechanical stiffness ($\sim 3{,}000 \text{ W m}^{-1} \text{ K}^{-1}$ and 1,060 GPa, respectively). Its ultimate tensile strength value is about 130 Gpa, which is considered higher than steel material by 325 times. The research studies have found that graphene has very high electrical conductivity value. Therefore, it is possible to utilize unique graphene properties in some engineering applications by using them in the manufacture of composite materials. The graphene sheets must be distributed in the matrices homogenously, in order to form efficient composite materials [32]. Conductive graphene composites is made by adding graphene to matrix materials such as: polymers, metal and ceramic, using a certain method to produce conductive composite that have steady and sustained electrical conductivity. The conductive graphene composites are commonly used in different applications, such as electronic devices, energy fields, batteries, capacitors and chemical engineering. [32][33].
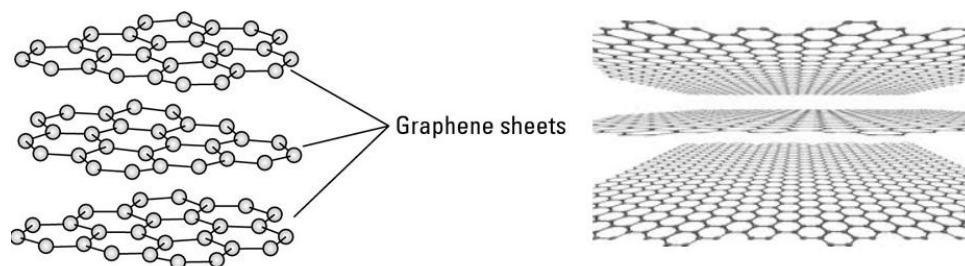

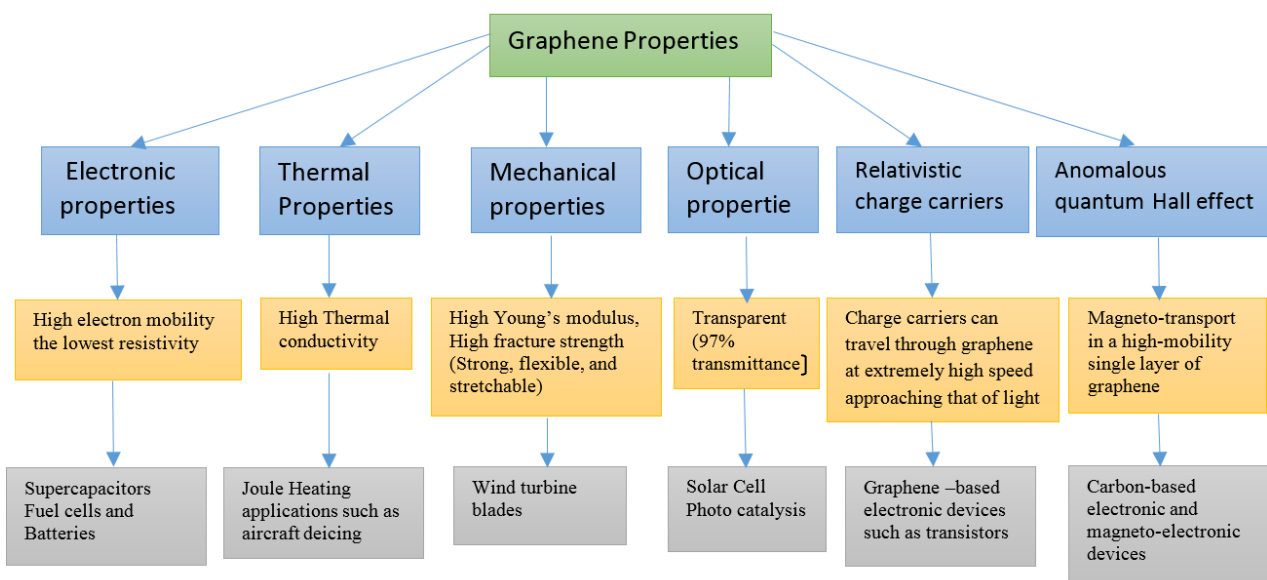**Figure 8:** Graphene sheets that exfoliated from graphite [32].

Figure 9: Graphene properties and applications [31-33].

3.2 Synthesis of Graphene

Graphene is prepared for the first time in 2004, by Novoselov and Geim. They exfoliated graphene from graphite using adhesive tape, this method is called micromechanical exfoliation method. In this method, graphene is peeled into multiple-layer graphene on the tape, afterwards graphene flakes are attached to the substrate. The quality of graphene obtained from this method is very high with no defects, but it is difficult to produce large quantities of graphene by this method. Therefore, another method is used to obtain higher amounts of graphene, where the graphite is dispersed in an organic solvent, in order to detach a graphene layer from the graphite. Then the solution is sonicated for several hours. After the dispersion, the solution is centrifuged at certain speed to remove the thicker flakes. The quality of graphene from this method is high, but the graphene size is still very small. Graphite oxide exfoliation is another method used to prepare graphene from graphite oxide which obtained by treating pristine graphite with strong oxidizers (see **Figure 10**). Graphite oxide is mixed with water or organic solvents, then using sonication and stirring for a long time, it can be exfoliated into a few layers of graphene oxide (GO) sheets. After centrifugation for a certain period of time, the graphene oxide sheets can be reduced to either graphene or reduced graphene oxide by thermal or chemical methods as shown in the **Figure 8**. [34- 41].
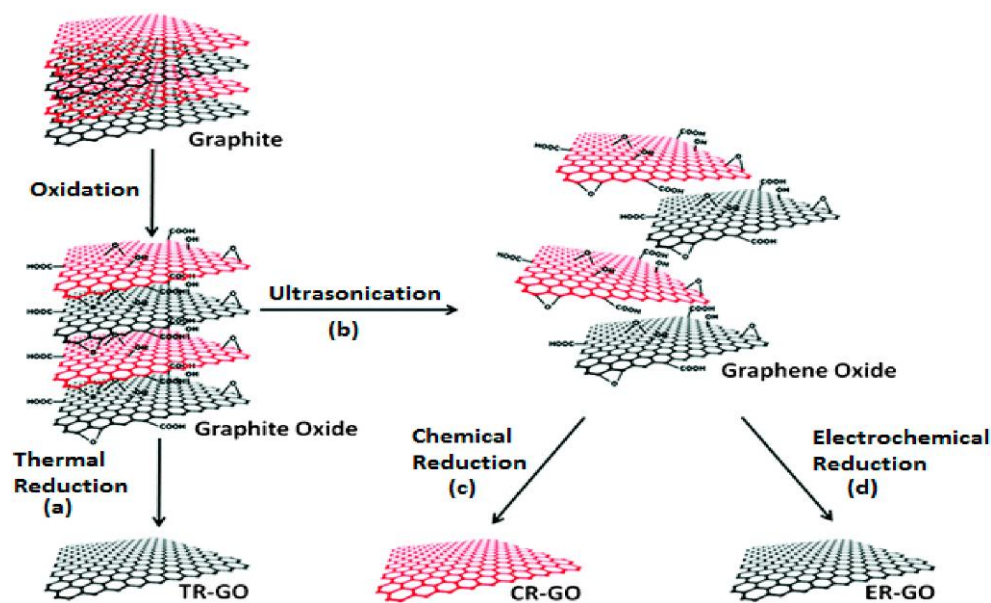
Figure 10: Schematic representation of the synthesis of chemically modified graphene [41].

Graphene also can be prepared by depositing dispersed graphene solution on the top of certain substrates using simple drop-casting method. The solvents are evaporated, leaving the graphene on the surface of substrate. There are another methods used to obtain graphene by growing it directly on a surface. The growth can occur in two different methods: epitaxial growth or chemical vapor deposition [42, 43]. **Figure 11** illustrates the epitaxial growth of graphene on silicon carbide substrate, where the growth of graphene can be achieved by heating the substrate to a high temperature, where this process allows evaporation of Si atoms, leaving the carbon atoms deposited on the substrate. Eventually, the deposited carbon atoms grow to form graphene. This method produce high quality graphene, and the layers of graphene can be controlled by controlling the temperature and pressure, but the amounts of graphene produced by this method is low, and the size of the homogeneous graphene layer is limited by the size of the crystal used [44-47]. While, chemical vapour deposition (CVD) is a well-known process in which a substrate (e.g. copper (Cu) or nickel (Ni)) or ceramic) is exposed to gaseous compounds (e.g. H_2, CH_4 and Ar) at high temperature about 1000°C . The carbon atoms carried by the carbon containing gas decomposed on the surface of substrate to produce the desired deposit, whereas the by-products evaporate by gas flow through the reaction chamber, leaving behind the carbon atoms. [47]. Afterwards, graphene starts to grow on the top of surface. A CVD growth graphene requires abrupt cooling after the exposure to carbon containing gas. The abrupt cooling will lead to high coverage of monolayer graphene. The slower the cooling process, the more graphene layers/stacking will be obtained. This method produces easily large amounts of high quality and harder graphene. However, this method is complicated, because it needs high temperature greater than 900 °C, and it is harmful to the environment (Producing corrosive and toxic gases) [48-50]. An overview of the different methods used to prepare graphene and their performances is given in the **Table 3**.
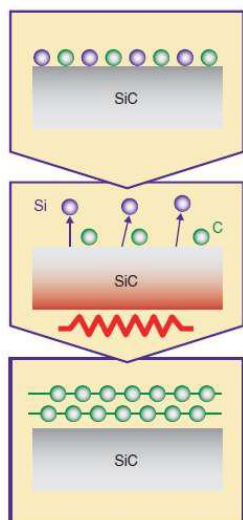
Figure 11: Schematic illustration of the epitaxial growth of graphene on SiC method [49].

Table 3: Overview of the performances of the different methods used to prepare graphene [41-50].

Method	Complexity	Advantages	Disadvantages
Adhesive tape	low	-Very high quality graphene	-Produc e low amounts of graphene
Liquid phase	low	-Very high quality graphene -produce large amounts of graphene	-the graphene size is still very small
Graphite Oxide Exfoliation	high	-Reduced graphene oxide is hydrophilic and can be solved in water by sonication or stirring.	-The reduced graphene oxide is of very bad quality compared to pristine graphene
Epitaxial Growth	high	-Graphene quality is high	- low amounts of graphene is produced - high cost -need high temperature
Chemical Vapour Deposition	Low	-Produced graphene layers can be very large and are easily obtained in large Amounts.	-expensive - Poor scalability

4. Conductive Graphene composite
4.1 Introduction

Graphene has been attracting great interest due to its unique structure and properties. To take full advantage of its properties for applications, integration of individual graphene in polymer matrices to form advanced multifunctional composites with significantly improved tensile strength, elastic modulus, electrical and thermal conductivity. This is one of the most promising routes, because graphene composites usually have exceptional specific modulus, specific strength, and wide application in aerospace, automobile and defense industries, etc. Moreover, graphene polymer composites can be easily processed and fabricated into intricately shaped components with excellent preservation of the structure and properties of graphene using conventional processing methods [51-57]. Graphene, a single layer of honeycomb lattice consisting of carbon atoms, has a unique form factor and exceptional physical properties which can be useful in electronic applications. The charge carrier mobility of graphene is 2-3 orders of magnitude higher than semiconducting silicon. When dispersed in a polymeric matrix, graphene sheets can provide conductive pathways for electron transfer similar to other carbon fillers such as CNTs and CB, thereby making the composites electrically conductive [58,59].

The impact of graphene-based composites is set to reverberate throughout countless industries, enhancing performance and increasing application possibilities. [60] The manufacturing of such composites requires not only that graphene sheets be produced on a sufficient scale but that they also be incorporated, and homogeneously distributed, into various matrices [61-62]. The key to the full utilization of GNP (Graphene Nano plates) properties in composites is their uniform dispersion in and strong interfacial bond with the matrix. The restacking of flat graphene sheets during fabrication makes uniform dispersion difficult and limits the available surfaces to interact effectively with polymer matrix, deteriorating the reinforcing effectiveness. Strong interfacial interactions between graphene and the host polymers are other important factors to be considered in fabricating high performance composites . Make the graphene surface wrinkled and rough, can increase the mechanical interlocks which will enhance the interfacial interaction, but this process may degrade electrical and thermal properties. Therefore, the core issues such as homogeneous dispersion of graphene sheets, their connectivity and orientation, interfacial interaction with host polymer matrix still deserve further research. [63,64]. The graphene-based composites have several types such as Graphene/Polymer Composites, Graphene/metal Composites, Graphene/ceramic Composites, Graphene/MOFs Composites and Graphene/CNTs Composites, **Figure 12** shows the fabrication, properties and applications of each Composite Materials [65].
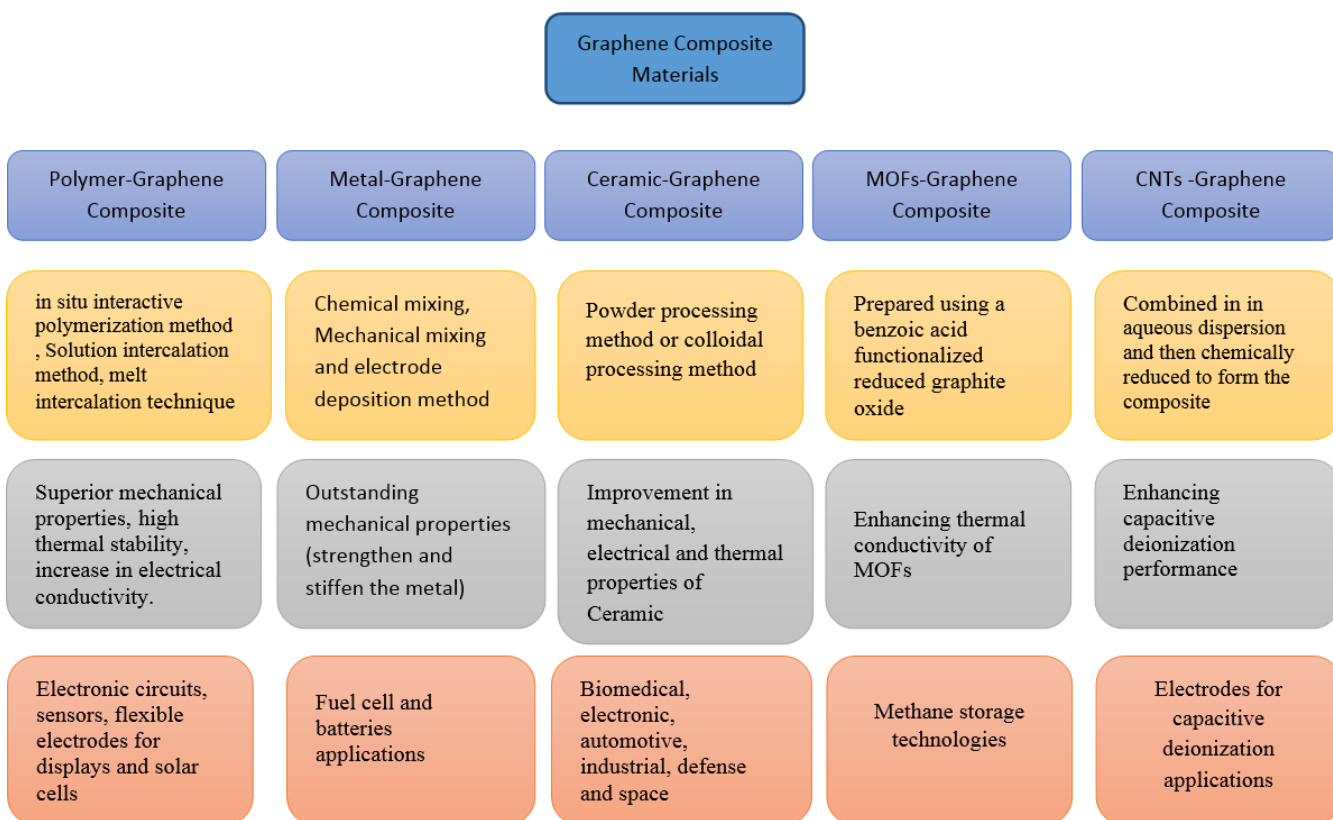
Figure 12: Types, fabrication, properties and applications of Graphene Composite Materials [60-65]

4.2 Manufacturing of Conductive Graphene Composite

Different methods are used nowadays to produce conductive composites materials. Composites with thermosetting resin use different techniques such as wet layup, resin transfer mold, vacuum infusion, autoclave and light resin transfer mold. The quality of filler dispersion is highly dependent on the proper manufacturing process [8, 66]. The combination of nano conductive fillers with polymer is more important to produce new composite materials with enhanced electrical properties using morphological modification or electronic interaction between the two components. Depending on the nature of the components used and the method of preparation, significant differences in composite properties may be obtained [67-69]. The incorporation process is obtained through melting or solution mixing. The dispersed materials are then molded to different parts by using readily available processing methods, such as injection molding, compression molding or extrusion, etc. A melt compounding method is more appealing than an in-situ polymerization method or solution mixing method, and its combination of traditional facilities, such as an extruder and a mixer, can give the production a greater diversity of polymers and fillers. Such a method is relatively economical and suitable for mass production and has already been successfully applied in the production of polymer conductive composites with the combination of conductive fillers of CNT, CB, expanded graphite, and graphene. During the molding process, it is possible to manipulate the mechanical and electrical properties of the final products by integrating different types, grades and quantities of conductive fillers. The main materials used as conductive fillers include (graphite, CNTs, CF, and CB) [8, 66, 70].

Physical and chemical properties of the graphene based polymer nanocomposite depend on the distribution of graphene layers in the polymer matrix and the interfacial bonding between the graphene layers and polymer matrix. However, pristine graphene does not form homogeneous composites, since it is not compatible with organic polymers. On the other hand, graphene oxide (GO) sheets which are heavily oxygenated graphene are compatible with organic polymers [71]. Therefore, GO is widely used as nanofiller for polymer nanocomposites. Unlike graphene, GO is electrically insulating and therefore cannot be used for synthesizing conductive Nanocomposites, therefore, graphene produced from the reduction of GO (i.e., the removal of oxygen) (see **Figure 13**) by thermal or chemical treatment can produce large quantities of reduced graphene oxide (RGO) platelets that have low sheet resistance which make rGO behave as a semiconductor with electrical conductivity of ~1000 S/m, which makes it an option for certain applications, such as antistatic coatings and semi-transparent electric circuits. There are two strategies to prepare graphene-based conductive film from this approach. The first is by having GO film previously prepared by vacuum filtration and subsequent thermal treatment or chemical reduction coupling with heat annealing .The second is a chemical reduction of GO in solution and then assembly to films by filtration [72,73].
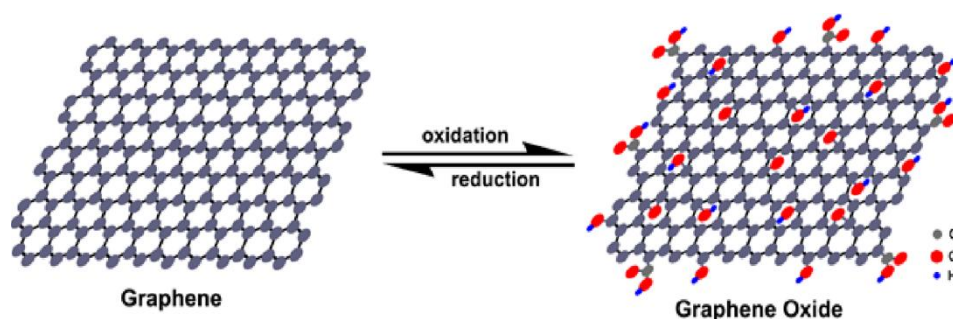
Figure 13: Scheme of structural model of graphene and graphene oxide (GO) [73-74].

Graphene oxide can be functionalized (**Figure 14**), in many cases, to be more soluble, and stable, and enhancing the electrical conductivity, in order to be used in the electronic applications. Several methods for the preparation and functionalization of GO. This can be done using covalent bonding of the functional molecules between the basal planes and edges of GNPs or monovalent adsorption method. The predicted properties of graphene nanoplates (GNPs) can be improved and achieved by creating covalent hydroxyl (-OH) or carboxyl (-COOH) groups. Thus, the functionalized graphene is multifunction material that can be used on a large scale such as increasing the solubility of graphene in different solvents, enhancing the dispersion method in matrices, and improving the manipulation of graphene for the fabrication of different devices. [72-75].

Figure 14: Schematic representation of the graphene functionalization [72-75].

4.3 Electrical Properties of Graphene Conductive Composites

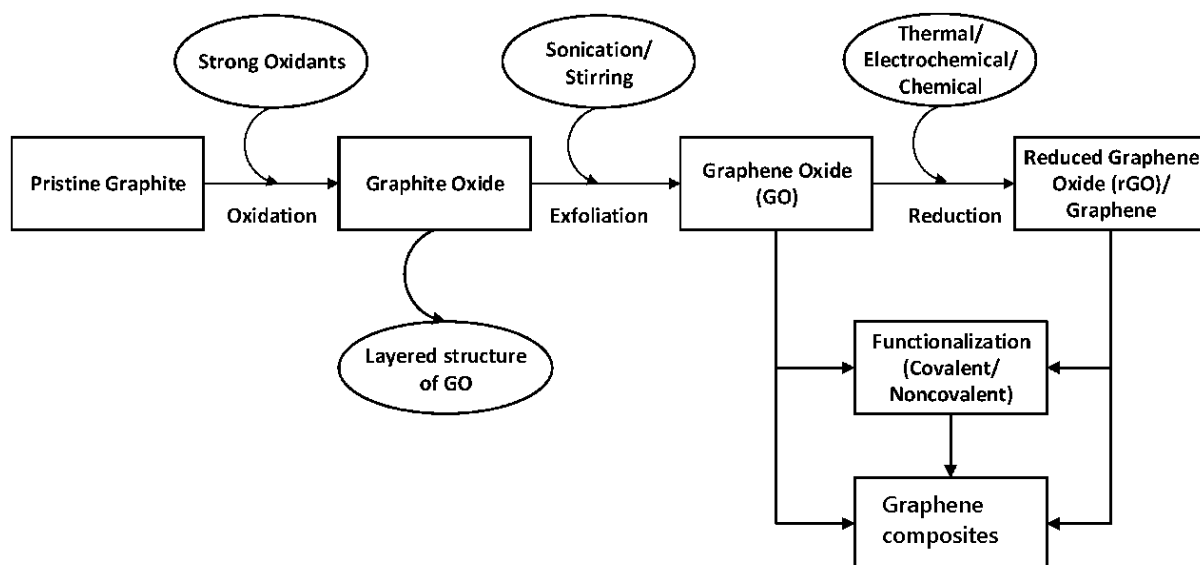Searching for materials that conduct electricity well has become essential for different applications. Carbon materials, which form a variety of allotropes occupy a unique place in terms of their electrical properties; because they conduct electricity very well. From these carbon materials is two-dimensional material graphene. Various efforts have been made to use the fascinating properties of graphene for macroscopic applications [76-78]. High electrical conductivity of graphene comes from its useful property zero-overlap semimetal. Because graphene has free electron in the outer shell that can move freely along the surface for electrical conduction. These electrons are called pi electrons which are overlapped to improve the bonding between carbon atoms in graphene [79, 80]. The researchers conducted transport experiments on graphene and they concluded that at room temperature graphene has high and magnificent electron mobility. Graphene has also ballistic transport properties, because the free electrons in the graphene can travel micro distances without being scattered. The resistivity of graphene is 10−6 Ω cm. Such a resistivity is lower than that of silver and graphene is considered the material with the lowest room temperature resistivity ever known so far [81-83].

Graphene has also extremely high thermal conductivities among other materials; most thermal properties of graphene are derived from those of graphite [84-87]. For example, sp^2 bonds between the carbon atoms are considered the strongest in the nature. On the other hand, the adjacent graphene sheets are bonded together by weak van der Waals interactions .The strong and anisotropic bonding and the low mass of the carbon atoms give graphene and related materials unique thermal properties [88-90]. Simulation work was firstly performed to predict the thermal conductivity of graphene and the value of thermal conductivity is found to be 6000 W m^{-1}K^{-1} at room temperature [91]. Later, another experiment is carried out to find the true thermal conductivity of the graphene and the thermal conductivity of graphene was found to be 2000 - 4000 W m-1K-1 [92, 93]. Furthermore, an optical method was used to measure the thermal conductivity of graphene and the reported value of thermal conductivity is about 5000 W m^{-1}K^{-1} [94-95]. There has been a growing interest in employing graphene as two-dimensional

heating elements on the macroscopic scale. Joule heating effect depends on the electrical and thermal conductivities of the conductive composite, which are affected by the properties of conductive filler.

Polymeric materials are naturally insulators, and the conductivity of composite materials depends on the content and properties of the conductive filler. In recent years, the polymer composites reinforced with carbon-based fillers have drawn the interest of the scientists and industry, because they are more effective for increasing and improving the electrical conductivity of nonconductive polymer matrix. These carbon-based fillers include fullerene, graphene, CNTs, CF, and CB [96-99]. Carbon nanotubes (CNTs) is highly effective to enhance and increase the electrical conductivity of polymer matrix. Due to the different structures and different properties of different kinds of polymers, when reinforced by CNTs, the nanocomposites will show different electrical percolation thresholds. The electrical percolation thresholds for CNTs-filled nanocomposites with thermosets such as epoxy resin and polyimide usually lie in the range from 0.1 to 1 wt%, while the values for thermoplastics such as polyethylene, polypropylene, polyamide, polycarbonate, poly (ethylene terephthalate) and polystyrene lie between 0.2 and 15 wt% [98,99]. E Kymakis , 2002 [100] investigated the interaction between carbon nanotubes and a soluble polymer (P3OT), it is found that the electrical conductivity of resulting film increased by five orders of magnitude.

In other research study done by J.K.W. Sandler, 2003 [101] studied the percolation threshold in carbon nanotube reinforced epoxy composites, the results are compared with previous studies, and the electrical conductivity is enhanced, because the percolation threshold in carbon nanotube epoxy composites was the lowest . Also Wei Liu, 2016 [102] used large size carbon nanotube buckypaper, which is used to reinforce the epoxy polymer, With 26.87% weight fraction of carbon nanotubes, the composites show enhancement in electrical properties with a conductivity of 220 S/cm . On other hand, the drawback of using carbon nanotube in composites that CNTs is expensive due to the less production. Carbon black (CB) is cheaper than CNTs [103], but the electrical conductivity of CB/polymer composites can be increased by adding high concentration of CB [104]. The addition of high CB contents improves the thermal stability of CB/ polymer composites. However, the disadvantages of using CB that some CB/ polymer composites shows wear as compared to raw particulate composite and it also shows semi ductile type failure .

Recently, graphene has become a favourite material for the scientists as a nanofiller for polymer matrices [105-109]. Graphene sheets can increase the electrical conductivity value of polymers several times at low concentration, more than carbon black. Therefore, graphene conductive composites can be used extensively in large scale of applications such as: electromagnetic shielding, electronic components, capacitors, electrodes for rechargeable batteries, sensors and actuators, and ice protection systems in the aircrafts [110-111]. Organic materials cannot be used alone for high performance applications because they have limited properties. Therefore, organic/reinforcing filler composites are frequently employed in order to overcome the limitation [112]. One of the widely used organic/reinforcing filler composites is an epoxy/ carbon-based nanofiller composite. Epoxy resins as organic material that have high strength, good stiffness, good thermal stability, excellent heat, moisture and chemical resistance; therefore, they are applied in the field of coatings, adhesives, casting, potting, composites, laminates and encapsulation of semiconductor devices [113]. The use of graphene as a conductive nanofiller in

the preparation of graphene/epoxy nanocomposites enhanced the electrical and thermal conductivities of the composite [114-117]. The variation in electrical conductivity with various forms of graphene and graphite reinforced epoxy composites are summarized in **Table 4**. The electrical properties of epoxy composites are compared in terms of reinforcement content (w %) and dispersion method. The maximum improvement in electrical conductivity was observed in the case of dip coating and mold casting fabrication methods, which is used by Embrey et a., 2017, [118] to disperse 1.91 wt % of graphene foam in the epoxy, where the electrical conductivity is improved and reached to 500 S/m. Ming et al., 2015, [126] used high concentration of graphene foam (about 80 wt.%) to modify the epoxy resin to improve it's electrical conductivity. There are another dispersion methods such as ball milling and mechanical stirring used to obtain high values of electrical conductivity. Therefore, both thermal and electrical conductivities improved in the case of mechanical stirring. Epoxy resin without any conductive filler is essentially insulative with an electrical conductivity of (10^{-14} S/cm), while the GNPs was reported to have excellent electrical conductivity (10^6 S/cm). The research studies found that electrical and thermal properties of graphene/epoxy composite, depend on the graphene content dispersed in the epoxy matrix [118-137].

Table 4: The variation in electrical conductivity with various forms of graphene and graphite reinforced epoxy composites.

Sr.	Authors	Year	Reinforcement (wt %)	Dispersion method	Enhanced Electrical Conductivity (S/m)	Ref.
1	Embrey et al.	2017	GrF (1.91 wt %)	Dip Coating and Mold Casting Fabrication Methods	500	[118]
2	Imran et al.	2017	GNPs (4 wt %)	Three-Roll Milling	1.5×10^{-3}	[119]
4	Raji et al.	2016	GNPs (5 wt %)	Mechanical Stirring	100	[121]
5	Zou et al.	2016	EGr (0.1 wt%)	Shear Mixing and Sonication	1×10^{-5}	[122]
			GNPs 0.1 wt%		5×10^{-3}	
6	Wan et al.	2016	GO (1.00 wt %)	Sonication and Ball Milling	2×10^{-8}	[123]
			RGO (1.00 wt %)		7×10^{-4}	
7	Prolongo et al.	2016	CNTs (0.1 wt %)	Direct Mixing and Sonication	4×10^{-1}	[124]
			GNPs (5 wt %)		7×10^{-1}	
8	Wu et al.	2015	GNPs (3 wt %)	Three Roll Milling and Sonication	8×10^{-3}	[125]
9	Ming et al.	2015	GrF (80 wt %)	-	5.1×10^{3}	[126]
10	Ghaleb et al.	2014	GNPs (1.1 wt %)	Sonication	6×10^{-3}	[127]
			MWCNTs (1.9 wt %)		7×10^{-4}	
11	Dou et al.	2014	Ag-G (25 wt %)	Sonication and Mechanical Stirring	4.6×10^{1}	[128]
12	Tang et al.	2014	GO-D2000 (4.8 vol %)	Sonication and HSM	0.16	[129]
13	Monti et al.	2013	GNPs (3 wt %)	Sonication and Mechanical Stirring	5×10^{-4}	[130]
			GNPs (0.24 wt %)		1.2×10^{-9}	
14	Al-Ghamdi et al.	2013	FGNs (40 wt %)	Centrifugal Mixing	8×10^{-2}	[131]
15	Kim et al.	2012	GO (3 wt %)	Mechanical Stirring and Magnetic Stirring	9×10^{-8}	[132]
			AL-GO (3 wt %)		7×10^{-8}	
16	Heo et al.	2012	Al2O3 (80 wt %) / GO	Three Roll Milling	1×10^{-6}	[133]
			Al2O3 (80 wt %)/ AL- GO		6×10^{-7}	
17	Tien et al.	2011	Gr (14 wt %)	Sonication	8×10^{-1}	[134]
18	Fan et al.	2009	GNPs (4.5 wt %) / CB (0.5 wt %)	Sonication and Mechanical Stirring	6×10^{-4}	[135]
			EGr (8 wt %)		1×10^{-6}	
19	Liang et al.	2009	FGNPs (15 w %)	Sonication	0.1	[136]
20	Li et al.	2007	MWCNTs (20 wt %)	Sonication	5×10^{-3}	[137]

GrF: Graphene Foam, GNP: graphene nanoplates, MWCNT: multi wall carbon nanotube, DGEBA-RGO: , GO: Graphene Oxide , RGO: Reduced Graphene Oxide , Ag-G: Silver plated graphene, GO-D2000: Polyetheramine

refluxed Graphene Oxide , FGNs: Foliated graphite nanosheets, Al-GO: , Gr : Graphite , CB: carbon black, EGr: Expanded graphite , FGNPs: Functionalized graphene nanoplates , Al_2O_3: Aluminum Oxide , HSM: Sonicated human serum microspheres

Graphene reinforced epoxy composites have attracted the interest of researchers in the aerospace field, especially in solving some problems that faced the aircrafts in the air, such as: lightning strike and ice accumulation on the aircraft surface [138-139]. Carbon fiber (CF) reinforced polymer composites (CFRPs), are used extensively in aircraft applications such as fuselages, leading edges and wing surfaces, due to their improved in-plane mechanical properties such as high tensile strength, high elastic modulus and excellent stiffness. Therefore, these composite materials eliminate the using of metallic meshes in aircrafts, as a result reducing the weight and the costs of installing these metals in structure. Using of CF composite in aviation, can reduce consumption of fuel, and improving the efficiency. On the other hand, CF composites are poor conductors of electricity, therefore, they are prone to damage from lightning strike [140-142]. Therefore, there is need to improve the electrical conductivity of carbon fiber reinforced epoxy composite, in order to withstand high electrical currents coming from lightning strike. Carbon fiber composites can be modified using carbon based materials such as graphene and carbon nanotubes to improve its electrical conductivity. **Table 5** shows a review of some researches done by some authors for the enhancement of electrical conductivity of modified CF /epoxy composite using graphene nanoplates (GNPs). As shown from the table, the electrical conductivity is enhanced and increased by 145 %, when graphene nanoplates or carbon nanotubes added to CFRPs. But the electrical conductivity of CFRPs is increased by 257 %, when the silver nanoparticles are added to CFRPs, however, using silver will add more weight to the composite, and this is not preferable in the aviation. Graphene were found to be very successful in conversion of electric energy to thermal energy in a so-called electro thermal effect (joule heating), and this allows the scientists to build new de-icing heater layer composite made of graphene nanoribbons films to be used in ice protection systems. [147-149]

Table 5: The enhancement of electrical conductivity of modified carbon fiber /epoxy composite (CFRPs) using graphene. AgnP: silver nanoparticles.

No	Author	year	Composite	Matrix/fiber	Filler	Electrical Conductivity (S/m)	Ref
1	Li et al.	2017	CFRP/GNPs	CF/Epoxy	GNPs	0.6	[143]
			CFRP	CF/Epoxy	__	0.03	
2	Kandare et al.	2015	CFRP	CF/Epoxy	__	0.07	[144]
			CFRP/GNPs	CF/Epoxy	GNPs	0.17	
			CFRP/AgnP	CF/Epoxy	AgnP	0.25	
3	Qin et al.	2014	CFRP/GNPs	CF/Epoxy	GNPs	7	[145]
			CFRP	CF/Epoxy	__	2.5	
4	Bekyarova et al.	2007	CFRP/CN	CF/Epoxy	CN	1.3×10^4	[146]
			CFRP	CF/Epoxy	__	6×10^3	

4.4. Conductive Graphene Ink

Ink is a liquid ,paste , or powder that contains pigments or dyes and is used to color a surface, Inks can be produced as printing inks and writing inks. Conductive inks are composed of ingredients that provide the function of electrical conductivity. Conductive inks (**Figure 15-a**) are useful for a range of applications, including printed and flexible electronics. Therefore, they can be used to produce cheap and effective electronic devices. The components of conductive ink are non-toxic and friendly to the environment, these components include conductive fillers such as silver, carbon-based material like graphite, reduced graphene oxide, and graphene nanoplates, or metal base material such as silver. The selection of conductive inks depend on the market, to be used in many applications such as: electronics, sensors, touch screens, printed heaters and more [150,151]. Ultra-Thin layers (< 1μm) of Conductive inks can be deposited on different types of substrates using Inkjet printing technology, as shown in **Figure 15-b**. This technology helps the engineers to design flexible and electronic components that would withstand considerable amounts of mechanical stress [152,153].
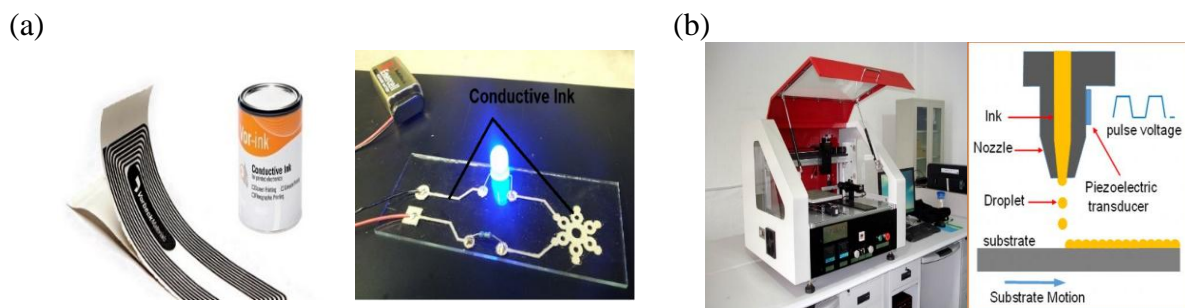
(a) (b)

Figure 15: (a) conductive ink. (b) Inkjet printing technology [154] [155].

Graphene based ink have magnificent characteristics for printing electrical components on different types of substrates [156]. Graphene conductive inks are found to withstand high values of repetitive mechanical stresses without breakage. As a result, there has been a global effort to develop practical methods to utilize graphene films for device fabrication. Unfortunately, pristine G is hydrophobic and cannot be uniformly dispersed in water to create a functional ink [157]. In addition to that, printable graphene inks are often synthesized using environmentally toxic organic solvents such as N-Methyl-2-pyrrolidone (NMP) and Dimethylformamide (DMF). Therefore, several surfactants have also been used, with varying degrees of success, to disperse G in an aqueous solution, one of these surfactants is a nontoxic hydrophilic cellulose derivative, ethyl cellulose (EC) and carboxymethyl cellulose (CMC), which used to create a conductive water soluble graphene (G) ink to help the naturally hydrophobic G sheets suspend in deionized water [158,159].

There are many techniques used to produce graphene films such as: chemical vapor deposition and epitaxial with high conductivity, but their application in electronic inks are limited by high cost and low production. Therefore, there are other methods used to exfoliate graphene nanoribbons chemically from graphite, to be used in producing of conductive graphene inks like oxidation/reduction method which used to exfoliate the graphite to nonconductive graphene oxide (GO) using Hummers method, then reducing GO into conductive reduced graphene oxide, but this method suffers from boring multiple-step chemical reaction processes and employment of explosive or toxic reagents. Therefore, there are other techniques used to produce conductive graphene or GNPs in large scale with high conductivity and low cost as shown in **Figure 16**. These methods include: electrochemical intercalation, bubbling exfoliation and liquid sonication, but the dispersion concentration of graphene in the solvent still not high, as well as the electrical conductivity of the solution need more improvement. Therefore, there is another excellent method, called ball-milling treatments (see **Figure 17**), this method allows the simultaneous exfoliation and surface modification of graphene, and this efficiently improves production yield and dispersion ability. Microfluidisation process used to produce high quality conductive graphene inks with high concentrations using ultrahigh shear forces to exfoliate graphene flakes from graphite for conductive inks, avoiding the need for centrifugation and reducing the time taken to produce a usable ink [160,161].
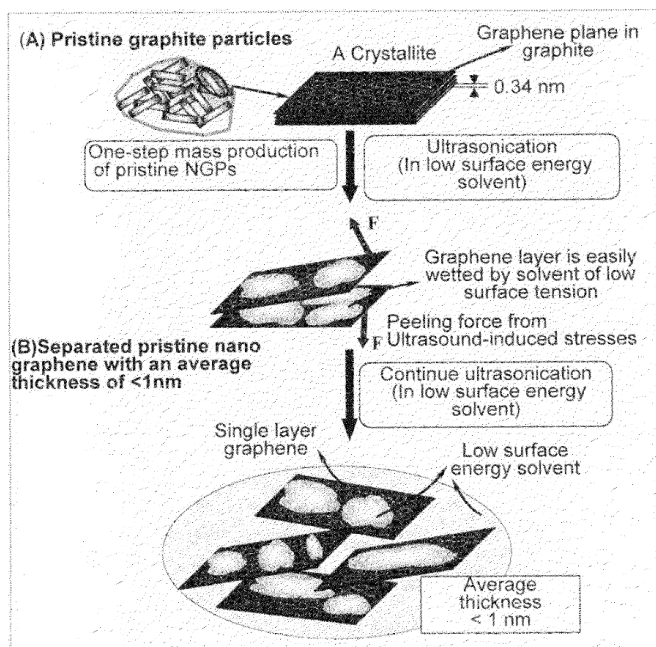

Figure 16: The essential procedures used to produce single-layer or few-layer [160] [161].
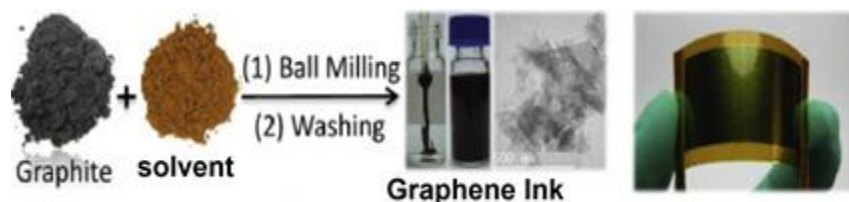
Figure 17: The processes that have been used to prepare graphene ink [161]

Table 6, shows the electrical conductivity values of conductive graphene-based inks, which are prepared by different scientists from 2011 to 2017, using different methods. The electrical conductivity of graphene-based inks prepared from graphene nanoplates, graphite, reduced graphene oxide, and functionalized graphene oxide exhibited magnificent values that is ranging from 1.2×10^2 (S/m) to 3.4×10^4 (S/m). On the other hand, adding some metals nanoparticles such as silver (Ag) with graphene to the inks, improved and increased the electrical conductivity to higher levels than using graphene alone. For example, Zhu et al and Jabari et al have found that in separated studies, the electrical conductivity of conductive inks, which prepared by adding silver nanoparticles and graphene, reached to 33.3×10^6 (S/m), which is an excellent value that can be utilized in more applications in the industry.

Table 6: Electrical conductivity values of graphene inks prepared using different methods.

No	Author	Year	Graphene Materials	Electrical Conductivity of ink (S/m)	Solvent	Synthesis Of Ink Method	Ref
1	Zhang et al	2017	GNP	3.5×10^4	Water + DY50	Direct mixing + ball milling +sonication	[162]
2	Michel et al	2017	Gr	909	T	Direct mixing + sonication	[163]
			Gr	104	NMP +EC		
3	Knopf et al	2016	GNPs	1.4×10^4	DI water + butoxyethanol+ CMC	magnetic stirring +Sonication +centrifugation	[164]
4	Zhu et al	2016	GNPs + AgnP	33.3×10^6	Ethanol+ PEG + glycerol	Direct mixing +sonication + mechanical stirring	[165]
5	Jabari et al	2016	Gr + AgnP	9.3×10^5	EC	Direct mixing + sonication, + ultra-centrifugal process	[166]
6	Majee	2016	Gr	10^5	MC+ saturated Br2 water + DI water	Direct mixing + sonication, + ultra-centrifugal process	[167]
7	Zhang et al	2016	GO + AgnP	2.0×10^3	DI water + ethanol + EG	magnetic stirring +Sonication +centrifugation	[168]
8	capasso	2015	GNFs	1.2×10^2	NMP	Direct mixing + ultra-centrifugal process	[169]
9	Huang et al	2015	GNFs	4.5×10^4	NMP	binder-free technique	[170]
10	Huang et al	2011	GO	900	DI water	Sonication + Centrifugation	[171]
			FGO	540			

NMP: N-Methyl-2-pyrrolidone, GNFs: graphene nanoflakes, FGO: Few layers graphene oxide   DY50: yellow 50 modifier, EC: ethyl cellulose, T: Terpineol, CMC: carboxymethyl cellulose, PEG: polyethylene glycol, MC: methyl cellulose, EG: ethylene glycol

6. Conclusion

Conductive composites include any composite having significant electrical conductivity, the electrical conductivity of a composite can be increased and enhanced by adding conductive fillers to the matrix such as carbon-based materials, when the filler content in the composite is increased, the conductive fillers particles start to contact each other, and form a continuous path that make the free electrons travel easily and conduct the electricity. Conductive composite materials have been used also to improve thermal management systems for transferring heat in the aerospace vehicles. The required enhancements include more thermal conductivity, large surface area, reduced weight/volume, as well as operability in harsh environments, such as durability under high flow rates, vibrations, stress, elevated temperatures, and oxidative environments.

Conductive polymer composites have a higher electrical conductivity and better mechanical properties than intrinsic conductive polymeric materials. These powerful properties make the conductive polymer composites replace the metallic parts in many applications. More significant research is carried out to investigate the addition of graphene into epoxy matrices, to improve the electrical, mechanical and thermal properties. Recently, graphene have become the favorite candidate for scientist to be utilized as nanofiller for these polymer matrices.

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