Graphene-based Polymer Nano-composites - A Review

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ABSTRACT

These days, different types of carbon nano-fillers are used widely as a reinforcement agent in polymer composites like fullerenes, carbon nanotubes, graphene nano-platelets, and graphite platelets. Moreover, graphene-based materials and their composites have shown promising characteristics for a wide variety of applications in nano-science and nano technology. Adding graphene as a reinforcing agent in a polymer matrix has improved the overall performance and properties of these substances. In this review, the general properties of the nano-particle in polymers have been studied. Also, the effect of these nano particles on the mechanical, thermal, and electrical properties of polymer composites has been investigated. It was demonstrated that filling graphene platelets in polymer materials improves their mechanical, thermal, and electrical properties.

Contents

1. Introduction

Recent researches in the field of polymer nano-composites (PNCs) have focused on the modification of nano-structured materials to improve their characteristics such as electrical, electrochemical, thermal, and mechanical properties [1-5] The discovery of graphene by the exfoliation of graphite has drawn enormous attention due to its unique properties [6-8]. Graphene is a 2D structure of carbon atoms with a hexagonal crystalline structure with sp2 bonds. It has a high surface area (highest, adsorption and surface reactions), electron mobility, thermal conductivity and mechanical strength. With the highest surface area compared to any other material of this kind, it enhances the interaction between the sheets and polymer material [2018]. The superior properties of graphene compared to polymers are also reflected in polymer/graphene nano-composites. Polymer/graphene nano-composites show superior mechanical, thermal, gas barrier, electrical, and flame retardant properties [12-14].

This article ectensively reviews the various investigations about the synthesis of graphene nano-platelets and the effect of these nano-particles on mechanical, thermal, and electrical properties of polymers.

2. General Properties of Graphene Nano-plateletes

Graphene is the thinnest two-dimensional atomic substance. It is a fundamental building block for 0D fullerenes [15-17], 1D carbon nanotubes [18-21], and 3D graphites [22-24]. This extaordinary compound is known as the thinnest and strongest ever measured material in the universe. It is a 2D structure of carbon atoms with a hexagonal crystalline structure with sp2 bonds, that holds unusual properties such as remarkable mechanical [25-29], thermal [30, 31], structural [32-34], and electrical [35-39] properties and also strong nano-filler-matrix adhesions [40, 41] that lead to strong mechanical strength [42-44]. These distinguished properties of graphene make it an ideal candidate to be used in the formation of polymer/graphene nano-composites (see Fig. 1).

Since its discovery in 2004 by Geim as a new allotrope of carbon, graphene has drawn a lot of attention from both theoretical and experimental scientists [11] because of its exceptional properties, such as high porosity [46], excellent electron

mobility [47,48], heat resistance [49] and high thermal conductivity of~5000 W m⁻¹K⁻¹ [50,51], that is comparable to current carbon nanotubes [52-55] and high optical transparency (97% Optical transmission) [56-60].

Moreover, monolayer graphene has a strength of about 200 times greater than steel which makes it the most durable material ever tested [61] and also has Young's modulus of about 1.0 TPa [62-65], and strength of 130 GPa [65] making it a promising reinforcing phase for ultra-strong structural composite materials [67]. Thus, the tensile strength of graphene is similar or slightly higher than CNT, but much higher than steel, kevlar, and natural rubber [68-70].

The theoretical surface area of graphene without overlap of sheets attains about 2630 m²/g with a layer thickness range of 0.35 to 1nm [71]. Although, in real and experimental systems, the observed layer surface is smaller due to the occurrence of overlap of the exfoliated sheets. In addition, graphene has a relatively high thermal conductivity in comparison with other materials. At the same time, the electrical conductivity of graphene is higher than most engineering materials except for steel [72, 73]. The considerable surface area of the graphene sheet provides a significant advantage in improving the mechanical, thermal, and electrical properties of the resultant composite [74, 75]. Due to all these reasons, graphene projects are rapidly rising on the horizon of materials science and condensed matter physics [76, 77].

Furthermore, graphene can be produced by exfoliation of graphite or its derivatives, such as graphen oxide (GO) [78]. The advantage of this approach is that it enables high yield production, and hence, it is a cost-effective and scalable process [72]. Therefore, this material is suitable for the production of polymer/graphene nano-composites. Graphene sheets are starting to play the role of an alternative to CNTs for preparing multifunctional polymer nano-composites, due to their high specific surface area, high aspect ratio, and layered structure [79].

Nowadays, the interest in nanoparticles such as graphene is being broadened considerably, regarding its' other interesting physical properties such as high levels of thermal conductivity [80], stiffness, and strength [81], and being coupled resulting in impermeability to gases [82]. It is proved that graphene and graphene-derived layered materials are also potential candidates for gas barrier membranes [83]. Also, according to recent studies, the production costs of graphene in large quantities are much lower than carbon nanotubes [84, 85]. The price of natural crystalline graphite is quite low, around 1.5$/lb, and the cost of exfoliated graphite nano-platelets is about 5$/lb or less [86]. These prices are significantly less than CNTs (100$/g) [87].
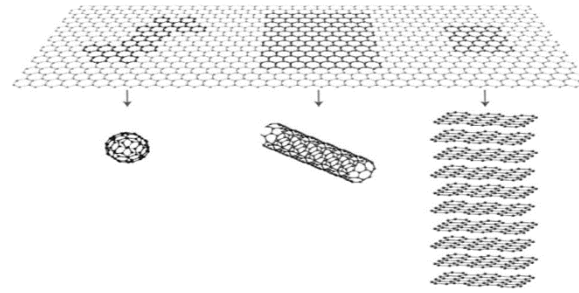
Fig. 1. Schematic representation of graphene, as the fundamental starting material for a variety of fullerene materials such as buck balls, carbon nanotubes, and graphite [45].

Thus far, graphene based nano-composites have been explored in a myriad of fields, including nano-electronics [79, 88, 89], intercalation materials, drug delivery systems [90] and catalysis. It has also shown potential applications in many technological fields, such as, single molecule sensors [91-94], biosensors [95,96], electrochemical sensors [97], molecular sensors [98,99], batteries [100-104], liquids crystal displays [105], light emitting devices [106], nano-composites [107-109], high frequency analog electronics [110], supercapacitors [63,111], optoelectronic devices [112], solar cells, transparent and flexible electrodes for displays [95,113,114] hydrogen [115] and energy storage devices [116]. Additionally, incorporating graphene sheets in a composite material is one of the practical methods to exploit their enhanced properties for real-world applications [77]. Substantial property enhancements of Polymer nano-composites in comparison with polymer composites with conventional micron-scale fillers (such as glass or carbon fibers) have been observed and have led to higher loading tolerating capacities, which ultimately result in a lower component weight [117].

Also, graphene can be used as an additive to delay the flammability of polymers due to its unique two-dimensional atomic carbon sheet structure [118-120]. The nano-filler network structure of graphene increases the heat transfer to the surrounding environment and thus retards the flame [121,122].

3. Synthesis of Graphene

Four different methods have been used to synthesis graphen [123]. The first method (which can be used for the preparation of graphene nano-platelet) is micromechanical exfoliation of graphite, which is also known as the 'scotch tape' or 'peel-off' method [124,125]; the second method is chemical vapor deposition (CVD) or thermal, chemical decomposition of graphitic materials [126,127]. Epitaxial growth on electrically insulating surfaces or electrostatic deposition of graphene [128] is known as the third method. And finally, the fourth method is the solution-based reduction of graphene oxide or chemical exfoliation [129-131].

The chemical exfoliation of graphite is the typical process for the bulk synthesis of graphene, in which graphite is oxidized to graphite oxide by oxidation with $K_2Cr_2O_7$ [132], $KMnO_4$ [132], and $KClO_3$ [134]. Furthermore, interest in the chemical synthesis of graphene nano-sheets is increasing due to the mass production of graphene nano-sheets and facile functionalization through oxygen (containing functional groups) [135,136]. The synthesis of graphene nano-sheets makes the graphene nano-sheets capable of being used in the areas of hybrid nano-composites, which shows promising applications in high capacity storage materials, and especially electronic devices such as batteries [137,138]. This method, which was initially developed by Stankovich [139], has been used to fabricate solution-processable functionalized graphene in two stages:

1- Water-soluble graphite oxide (GO) was prepared from graphite using the Hummers method [60] and dispersed in water with appropriate sonication to yield a dispersion containing mainly individual graphene sheets.

2- The exfoliated GO was reacted with an alkylamine to obtain a solution-processable functionalized graphene.

On the other hand, exfoliation of graphite or its derivatives, such as GO is an alternative method for producing graphene that is used nowadays [78]. Generally, these methods are suitable for large-scale production for polymer composite applications. Usually, the Staudenmaier [111,140] or Hummers [141] methods use different variations for production of GO where graphite is oxidized using strong oxidants such as $KMnO_4$, $KClO_3$, and $NaNO_2$ in the presence of nitric acid or its mixture with sulfuric acid.

Also, graphene sheets can be attained by graphite exfoliation, including mechanical cleavage of graphite [142-145], chemical exfoliation of graphite [104,146], thermal-induced exfoliation [147], and direct synthesis, such as epitaxial growth and bottom-up organic synthesis (see Fig. 2) [148]. In terms of the yield and production cost of graphene, the solution phase preparation technique has gained the most attention in recent years [149].

3.1. Characterization of graphene nano-platelets

One of the main problems in graphene related research is the difficulty of accurate counting of the atomic layers in samples obtained by either mechanical exfoliation from bulk graphite or grown by other means [112]. Atomic force microscopy (AFM) may not identify the number of graphene layers individually, where there are other alternatives, including transmission electron microscopy (TEM). A significant disadvantage of these tests is the lengthy preparation process of the involved experimental samples. The X-ray diffraction (XRD) test method has also been used for the characterization of the structure of nano-composites [150]. Recently micro-Raman spectroscopy has become a conventional technique for the identification and characterization of graphene layers [151-153]. Since, the Raman spectrum of graphene is very sensitive to the number of atomic layers and the existence of disorders or defects, which allows for better graphene characterization.

4. Intercalation and exfoliation of graphene in polymers

The intercalation of graphite increases its interlayer spacing, weakening the interlayer interactions, and facilitating the exfoliation of the graphite intercalation compounds by mechanical or thermal methods [104]. Dispersion of graphene nano-sheets in to the polymer hosts is a challenge in the development of high-performance graphene-based nano-composites due to the strong interlayer cohesive energy and surface inertia [154,155]. Also, graphene nano-particles are highly hydrophobic, and thus, it is challenging to disperse them in solvents [156,157]. These subjects limit the application of pure graphene sheets in nano-composites. To improve the dispersibility of graphene in the matrix, both the surface modification and in situ polymerization approach have been tried [158,159] (see Fig. 3). Furthermore, expanding the application of graphene between graphene sheets and aromatic organic molecules has been carried out based on π-stacking [130, 97], which could improve its solubility [160]. Recently, it has been found that the functionalization of graphene with a polymer is an efficient method to enhance its dispersibility and solubility in the aqueous solvent [161]. Furthermore, the insolubility and re-aggregation tendency limiting the manipulation and processing of graphene results in the non-uniform dispersion of graphene in the polymer matrix. Much effort has been dedicated to the chemical functionalization of graphene to improve the solubility and compatibility with polymers by changing its surface properties [162,163]. Graphene platelets, in their intact form, are characterized by low surface energy, and therefore, poorly wetted by most polymer matrices. Consequently, the dispersion is often poor and leads to reduced mechanical properties of the composite [151]. In most cases, conventional organic modification of graphene sheets improves their dispersion in the polymer matrix [164, 165]. The melt rheological property also provides a convenient way to evaluate the dispersion state of the nanofillers in the polymer matrix [14].

Additionally, the final properties of the graphene reinforced polymer nanocomposites are dictated by interfacial bonding between graphene and the host polymer [166].
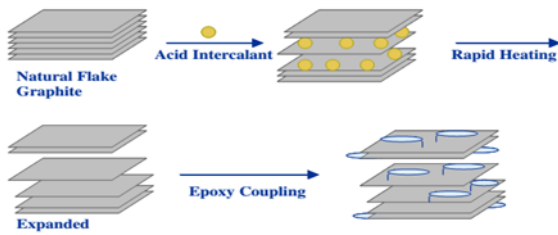
Fig. 2. Schematic picture of graphite expansion and functionalization process [148].
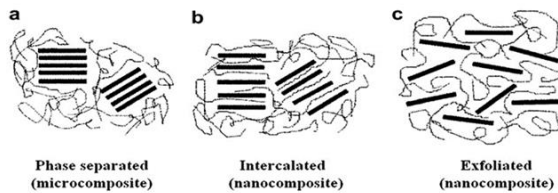

Fig. 3. Schematic figure of three morphological states (originally suggested for layered silicate fillers) (a) phase separated, (b) intercalated, (c) exfoliated [158].

Pristine graphene is not compatible with organic polymers and does not form homogeneous composites. Hence, it is difficult to obtain an acceptible dispersion of graphene in polymer matrices. Instead of using pure graphene, the functionalization of graphene (FG) is one of the most common methods to solve the problem [167]. The functionalized groups enable graphene to disperse uniformly and interact with polymer chains producing strong interfacial adhesion [168]. Oxidization is a typical way to functionalize graphene, which is denoted as GO. GO consists of graphene lattices that are chemically functionalized with hydroxyl, carbonyl, and epoxide groups [169]. This additional carbonyl and carboxyl groups take place at the edge of the sheets and make graphene oxide sheets, strongly hydrophilic, allowing them to swell and disperse in water [170].

5. Effect of graphene on thermal properties of polymer nano-composite

As electronic products continue to move toward miniaturization and high-performance, one of the critical challenges is dramatically increasing energy dissipation [115] (see Fig. 4). If this problemis not solved, the energy dissipated in the form of heat will significantly reduce the speed of development of new technologies. Recently, for this purpose, thermal interface materials have been used. These materials have been applied to connect different thermal elements to ensure efficient transfer of heat. They can be inserted between a chip and a heat sink or between a heat sink and a heat spreader in the electronic device [171]. Graphene-filled polymers are considered to be up-and-coming candidates for high performance thermal interface materials and expected to solve the challenge of increasing heat dissipation from electronic devices (see Table. 1 for some instances). The glass transition temperature (T_g) of nano-

composites has been found to increase by filling graphene nano-platelets [182]. In addition, increasing the number of filled graphene, results in the decrease of T_g due to the agglomeration of graphene nano-particles. The introduction of filler components into organic polymers can improve their thermal degradation stability; in other words, graphene adds thermal stability [170,183] to nano-composites because of the thermal isolation effect of the graphene sheets and the mass transport barrier that they provide to the volatile products which are generated during thermal decomposition [184]. The test results indicated that the inclusion of graphene into composites resulted in low coefficients of thermal expansion (CTEs) [182], and increasing graphene fraction reduced CTEs significantly [112,185]. Some computational results suggested that in-plane expansion, bond stretching, and bond bending effects in the graphene sheets counteract each other out, leading to a negative thermal expansion coefficient in the plane graphene sheets below the temperature of 470 K [166,186]. Due to its planar structure, thermal contraction in graphene sheets was more evident than other carbon structures, such as graphite, carbon nanotubes and, diamond [150,166].

The preliminary measurement of thermal conductivity also indicated that graphene composites significantly improved the thermal conductivity of the polymer matrix [77].

Moreover, since the shrinkage of resin below the glass transition temperature (T_g) increases linearly with reduction of temperature; thus, the difference between the operating temperature and Tg is significant. The magnitude of the resulting internal residual stresses depends on T_g, CTE, and the elastic modulus of the resin, as well as the curing conditions and the degree of dimensional constraint in the forming process [150].

6. Effect of graphene on mechanical properties of polymer nano-composites

To enhance the mechanical properties of polymeric nano-composites, substances such as carbon nano-tubes, intercalated clay, graphene, and graphene oxide are added as high-performance reinforcing nano-fillers [65]. The mechanical properties of composites depend not only on the dispersion and mechanical properties of the nano-filler in the polymer but also on the features of the polymer–particle interface [187]. On the other hand, it has been demonstrated that the enhanced mechanical performance of the polymer nano-composites depends on the inherent properties of the nano-filler and also, more importantly, depends on the nature of the bonding at the interface and the mechanical load transfer capability from the matrix to the nano-fiber [40].
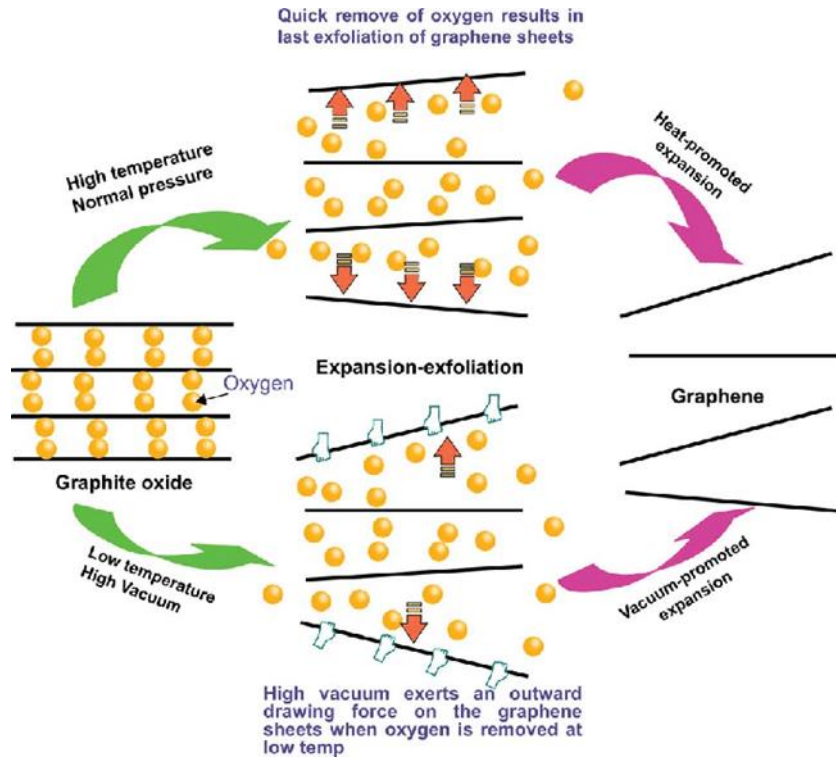
Fig. 4. Schematic representation of thermal exfoliation of graphene [165].

As a result, the superiority of graphene platelets in terms of mechanical properties enhancement may be related to their high specific surface area, enhanced nano-filler matrix adhesion/interlocking arising from their wrinkled (rough) surface, as well as the two-dimensional (planar) geometry of graphene platelets [188]. furthermore, the maximum strength of nano-composites increases with the addition of graphene up to a critical graphene loading point, and then decreases above that critical content [189]. A similar trend is observed for the initial modulus. This phenomenon occurred mainly because of the agglomeration of graphene sheets above the critical graphene loading, as previously described. Also, the improvements in the tensile mechanical properties depend on the interactions between the polymer molecules and the layered graphene, as well as on the rigidity of the graphene sheets [190-192]. Moreover, the enhancement of the toughness of thermosetting materials by the incorporation of graphene has been studied [78]. Thermosetting materials are generally cured with high crosslink density to attain excellent mechanical properties. However, it almost always achieves weak fracture resistance [193]. The conclusion may review the main points of the author's work. Also, it could include the application of the proposed method and suggestion for future research. The percentage of improvements in tensile strength and tensile modulus are shown in Fig. 5. The maximum improvement in tensile strength is as high as 108% [194] and in the tensile modulus up to 103% [195].
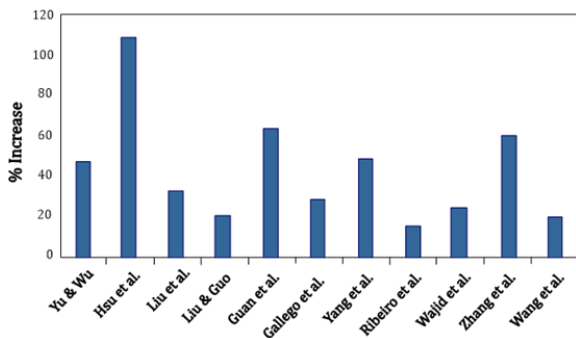
7. Effect of graphene on electrical properties of polymer nano-composites

Recently, graphene nano-sheets, as a new kind of carbon material, have captured much attention due to their excellent electronic conductivity [108, 206, 207], and their very high electron mobility [208,209]. Because of this high conductivity and electro-catalytic activity, graphene is an ideal material for the preparation of electrochemical sensors and biosensors [210-213]. Furthermore, these sheets have attracted enormous attention due to their potential application in liquid crystal and nano-electronic devices, as well as in super-capacitors and field emitters [214]. This nano-particle is superior to other conventional carbon materials due to the ease of synthesis, cost-effectiveness, remarkable mechanical stiffness and, large surface area [215]. This nano-particle and its derivative, graphene oxide (GO), have been substantively used as the main framework for dispersing or building nano-architectures because of their desireable properties in electronics and catalysis [132]. It is extremely promising for graphene to become the nano-scale building block of new nano-composites because of its unusual nano-structure and extraordinary electronic properties [216,217]. Also, a nano-composite of graphene/MnO2 nano-platelets has been prepared as high lithium capacity electrical batteries. The superior lithium storage capability can be attributed to the open structure: the large effective surface area and short diffusion paths [218].

Table 1. A brief record of graphite and/or graphene-based nanocomposites studied for improvement in thermal conductivity values.

No.	Authors, Year	Reinforcement (wt %)	Dispersion method	% Increase in thermal conductivity	Remarks	Ref.
1	Kandare et al. 2015	GnP (1.9 wt %) SnP/(0.09 wt %) SnW/(0.09 wt %) GnP (1.9 wt %), SnP (0.09 wt %) GnP (1.9 wt %), SnW (0.09 wt %)	Sn	9 18 8 38 40	The simultaneous inclusion of GnPs and SnP/SnW at a combined loading of 1 vol % resulted in about 40% enhancement in the through-thickness thermal conductivity, while the inclusion of GnP at the same loading resulted in only 9% improvement. A higher increment with simultaneous addition of GnP and SnP/SnW can be attributed to synergistic effects.	[172]
2	Fu et al. 2014	Graphite (44.30 wt %) Graphite nanoflakes (16.81 wt %) Graphene sheets (10.10 wt %)	MS	888.2 982.3 2258.8	The maximum improvement in thermal conductivity was observed in the case of graphene sheets with a thickness of 1.5 nm.	[173]
3	Im and Kim 2012	Thermally conductive graphene oxide (GO) (50 wt %) Thermally conductive graphene oxide (GO) (50 wt %), MWCNTs (0.36 wt %)	Sn	111 203.4	The thermal conductivity decreases after 50 wt %, which can be attributed to residual epoxy that forms an insulting layer on reinforcement. MWCNT helps the formation of 3D network structures.	[174]
4	Pan et al. 2015	Perylene bisimide (PBI)-hyper-branched polyglycerol (HPG) modified reduced graphene oxide (RGO), (PBI-HPG/RGO) (1 wt %)	Sn	37.5	The filler was observed to be uniformly dispersed, resulting in strong interfacial thermal resistance.	[175]
5	Zhou et al. 2015	Multi-layer graphene oxide (MGO) (2 wt %)	Sn	104.8	The thermal conductivity decreases after 2 wt % MGO.	[176]
6	Wang et al. 2015	GNPs (8 wt %)	MS	627	The thermal conductivity increases with GNPs at the loss of Vickers microhardness after 1 wt % of GNP.	[177]
7	Tien et al. 2011	Graphene flakes (12 wt %)	Sn	350	The thermal conductivity increases exponentially with increasing wt % of graphene flakes.	[178]
8	Chandrasekaran et al. 2013	GNP (2 wt %)	3RM	14	The thermal conductivity increases with increasing temperature.	[179]
9	Chatterjee et al. 2012	Amine functionalized expanded graphene nanoplatelets (EGNPs) (2 wt %)	Sn + 3RM	36	The EGNPs form a conductive network in the epoxy matrix allowing for increased thermal conductivity.	[180]
10	Gallego et al. 2011	Functionalized graphene sheet (FGS) (1 wt %) in nanocomposites	ShM	63.6	The layered structure of MWNTs enables an efficient phonon transport through the inner layers, while SWNTs present a higher resistance to heat flow at the interface, due to its higher surface area. The f-MWNTs have functional groups on their surface, acting as scattering points for the phonon transport.	[181]

Fig. 5. The % increase in tensile properties of epoxy/graphene nanocomposites [194–205].

8. Conclusions

In recent years, graphene materials have been of great attention due to their desireable mechanical, electrical and thermal properties. Besides, their large surface area as compared with other nanostructural materials, have made it an ideal substance used as nano reinforcement for many polymer-based composites. As a result, general properties and synthesizing of graphene nano platteletes were studied in the first section of the presented research. It was concluded that two-dimensionality and nanoscale thickness of the graphene platelet causes a high aspect ratio of this nano particle due to significant mechanical, thermal, and electrical properties. The effect of the addition of these nanoparticles on mechanical, thermal, and electrical properties of polymer composites was investigated in other sections of this research. The results of the investigations show that the mechanical properties of polymer composites highly improve by dispersion of these nano particles because of the superior mechanical properties of these nano particles. On the other hand, outstanding thermal properties of graphene platelets cause the enhancement of thermal properties of polymer composites such as increasing the thermal conductivity and decreasing of CTE of polymer-based composites. Furthermore, the high electron mobility of these nano particles causes elevated electrical conductivity of nano-polymer composites in which it was dispersed. Finally, to determine the number of atomic layers and assess the quality of the graphene materials, sufficient information was given by Raman spectral features.

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