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Review Article

Polymer/Graphene Nanocomposite: Preparation to Application

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Abstract

Graphene has emerged as a topic of huge scientific interest due to its high surface area, exceptional mechanical properties, electron transfer, and other physical properties. In polymers, this one-atom thick 2D crystal may significantly enhance the physical properties at very small loading level. In this review, essential characteristics of polymer/graphene nanocomposite have been discussed. Moreover, fabrication techniques (*in situ* method, solution route, melt technique) frequently employed for polymer/graphene nanocomposite have been discussed. Applications of these nanocomposites in Li-ion batteries, electronic devices, and solar cells have been conversed along with the current challenges associated with processing and scalability of these materials.

Keywords: Polymer; graphene; nanocomposite; Li-ion battery; solar cell

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1. Introduction

Polymers have been employed as recurrently active components in wide range of technical applications. The extensive use of polymers is entrenched in their versatile structure and properties such as lightweight, processability, strength, ease of fabrication, and low cost. However, technical applications of polymers have been limited due to deprived thermal, electrical, and mechanical properties [1]. In this regard, physical properties of polymers have been modified using reinforcement or second phase in the matrix [2]. Nanoparticles (graphite, graphene, carbon nanotube (CNT), carbon nanofilber) have been exploited as successful reinforcing agent for polymers compared with the traditional fillers. In polymer/nanofiller nanocomposite, final material properties usually depend on *(i)* nature and type of polymer; *(i)* nature and type of nanofiller; and *(iii)* nature of interaction between polymer and nanofiller [3]. Consequently, polymer nanocomposite reveals superior material performance than pristine polymer matrix and composite [4]. Incorporation of graphene nanofiller has gained immense research interest [4]. Graphene is composed of carbon atoms that are densely packed in honeycomb crystal lattice. Graphene is two-dimensional carbon nanofiller with one-atom-thick planar sheet of sp^2 bonded atoms. It is the thinnest known material found in the universe and has tremendous potential [5]. Graphene exhibits range of essential characteristics such as flexibility, transparency, high aspect ratio, surface area, tensile strength, thermal and electrical conductivity, electromagnetic interference shielding ability, and low coefficient of thermal expansion (CTE) [6]. Superior mechanical properties, high thermal conductivity, and outstanding electronic properties have been observed when compared with other conventional nanofiller such as carbon nanotube, montmorillonite (nanoclay), graphite, and exfoliated graphite (EG) [7]. The exclusive properties of graphene are further enhanced when combined with polymers to form polymer/graphene nanocomposite. Polymer/graphene nanocomposite has demonstrated superior mechanical, thermal, and electrical properties compared to neat polymers. Improvements in mechanical and electrical properties of polymer/graphene nanocomposite have been observed relative to nanoclay or other carbon filler-based composite. Recently, production of polymer/graphene nanocomposite has gained extraordinary research attention. Graphene has also been largely exploited as conducting nanofiller in electronic applications [8]. Due to the existence of isolated graphene sheets, thermal, mechanical, and electrical, and barrier properties of the resulting materials have been influenced [9]. Polymer/graphene nanocomposite systems are expected as potential contender in electronic equipment, electrode materials, electronics packing, chips, fuel cells, aerospace, sporting, goods, radar absorbents, corrosion resistant coatings, and other devices [10].

2. Graphene

Graphene is basic structure of several carbon nanofillers as graphite, CNT, graphene oxide (GO), and fullerene (Fig. 1).

The planar sheet of sp^2 bonded carbon atoms in graphene is densely packed In honey comb crystal structure [11-13]. Graphene own extraordinary properties. It has double surface area than that of a single walled carbon nanotube (SWCNT). Graphene also has tunable electronic band gap, ultra high mechanical strength, excellent thermal conductivity, and elasticity [14]. Consequently, graphene is a monatomic nonomaterial (2D) consisting of sp^2-hybridized single layer of carbon atoms with benzene as repeating structural monomer. Since the discovery of graphene, It has been employed to resolve several research deficiencies associated with polymer/graphite and polymer/CNT composite [15]. A single atomic sheet of graphite i.e. graphene is 200 times harder than steel and 30 times harder than

diamond [16, 17]. Graphene also has certain advantages compared with carbon nanotube *(i)* graphene synthesis from inexpensive graphite; *(ii)* low cost production than nanotube; and *(iii)* high purity as metallic impurities present are present in CNT [18]. Table 1 outline various processes used for graphene production. Graphene, due to excellent mechanical, electrical and thermal properties, has recently attracted immense research interest as reinforcing phase in nanocomposite materials [19]. Graphene also has intrinsic energy dissipating mechanism, sheet bending and sliding to improve toughness nanocomposite [20-23]. However, sometimes graphene sheets may restack to form graphite due to high aspect ratio and vander Waals interaction.

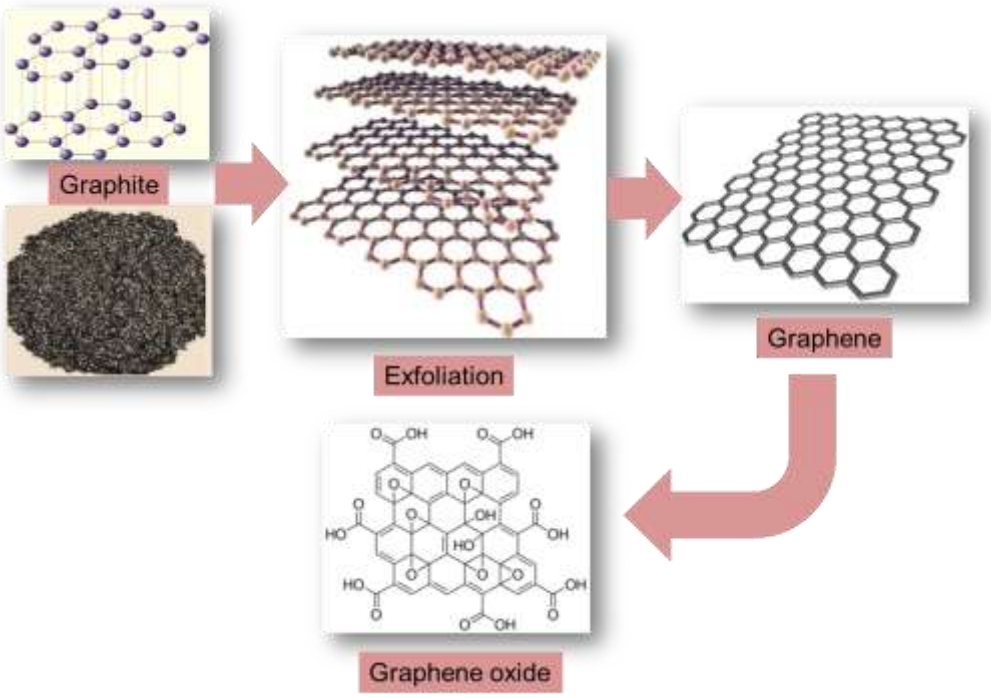

Fig. 1 Graphite, graphene, and graphene oxide.

Table 1 Processes for graphene production.

Method	Thickness	Advantage
Reduction of GO	Multi-layered	Good yield
From CNT	Multi-layered	Thickness control is tricky
Arc discharge	Single to few layer	Good yield
Chemical vapor deposition (CV	Single to few layer	Controlled thickness

3. Polymer/graphene nanocomposite

Graphene is a magnificent reinforcement for polymers owing to implausible physical properties such as high thermal conductivity ~5,000 W/mK, high electron mobility ~ 250,000 cm^2/Vs, and considerably high Young's modulus ~1TPa. Even very small amount of graphene has been found to improve mechanical, electrical, and thermal properties of nanocomposite [24-26]. Properties of graphene reinforcement have been compared with carbon nanotube and other conventional nanofillers [27]. A major problem with graphene reinforcement is poor dispersion in polymer matrices. For this purpose, surface modification of graphene has been performed. Functionalized graphene has bee found to easily dispersed and compatible with polymers. Fine distribution of graphene in polymer matrix in turn may enhance the mechanical, thermal, electrical, and gas barrier properties of final nanocomposite. Compatibility between polymer and graphene depends on the polar nature of the polymer as well as type of functional groups present on polymer backbone. In this regard, hydroxyl modified graphene, carboxyl modified graphene, amine modified graphene, as well as ionic liquid-modified graphene have been produced and studied for fine dispersion in polymer matrices [28]. Use of unmodified graphene has also been studied with polystyrene by *in situ* technique [29]. Poly(methyl methacrylate)/graphene nanocomposite has revealed applications in anti-corrosion, additive, coating, sealer, binders, optical fiber, and outdoor electrical applications [30, 31]. To improve thermal and electrical properties of the PMMA/graphene nanocomposite, functionalized graphene has been found more effective than non-functional form [32]. The PMMA/unmodified graphene nanocomposite was obtained through emulsion polymerization of methyl methacrylate monomer in graphene dispersion. Graphene has also been reinforced in polyethylene, polypropylene, and polybutylene matrices [33]. Polyvinyl alchol (PVA)-based graphene nanocomposite were also reported [34]. However, dispersion of graphene in polymer matrices is still a research challenge. Thermoplastic polyurethane (TPU), polyacrylic acid (PAA), polyacrylonitrile (PAN), polyester, polyolefin, and epoxy resin have also been considered as potential hosts for graphene [35-40]. Solution technique has been used in most of cases. In addition, latex technology has been found useful for dispersing graphene into a polymer matrix [41, 42]. Exfoliated polystyrene(PS)/graphene nanocomposite has been prepared [43]. Exfoliated polypropylene (PP)/graphene nanocomposite have also been prepared [44]. The storage moduli of PP/graphene nanocomposite was increased with increasing graphene loading up to 1.0 wt.%. Beyond this concentration, there was slight reduction in the storage moduli of nanocomposite. An increase in glass transition temperature (T_g) was observed with 0.1 wt. % of graphene. Thermal conductivity of 0.396 W/MK was observed with 2.1vol % of graphene [45, 46]. Thus, graphene is very promising as nanofiller to fabricate high-performance polymer nanocomposite [47, 48]. Multilayer graphene nanoplatelet (MLG) and polymer nanocomposite has also been fabricated from exfoliation of graphite. The affect of MLG on thermal conductivity and stability of polymer/MLG nanocomposite was explored. The thermal conductivity was found to increase with increasing nanofiller content.

4. Preparation strategies

Sonication, ultra-sonication, *in-situ* polymerization, and solution mixing have been used to form polymer/graphene nanocomposite (Table 2). Solution method is the low cost and facile technique in this regard (Fig. 2).

Choice of the technique basically depends on uniform dispersion of graphene, exfoliation, and complete intercalation with polymers [49]. However, all the mentioned techniques appear ideal to process polymer/graphene nanocomposite [50]. The coupled method of *in-situ* polymerization and solution mixing

has been used for the chemical interaction between the polymer and graphene. In solution route, serious problems are *(i)* use of hazardous chemicals and *(ii)* non-feasibility for industrial scale production due to low yield and high cost. Melt mixing technique is also in demand. However, direct melt-mixing of polymer and graphene is not adequate for appropriate filler dispersion. The poor dispersion in melt method may affect the quality and properties of resultant polymer/graphene nanocomposite [51]. Melt route has been modified to influence the dispersion quality. Therefore, a combined solution and melt route has been designed for better dispersion of graphene in polymer matrices [52]. Polymer/graphene nanocomposite obtained by combination approach has shown better mechanical and flexural properties due to better dispersion. Materials obtained by modified melt approach also show high electrical conductivity.

Table 2 Polymer/graphene nanocomposite.

Polymer	Reinforcement	Processing
Poly(vinyl chloride) (PVC)	Reduced graphene	Solution, *In situ* route
Poly(methylmethacrylate) (PMMA)	Reduced graphene	Solution, *In situ* route
Polyurethane	Graphene/Reduced graphene	Melt, Solution, *In situ* route
Polystyrene (PS)	Graphene/Reduced graphene/GO	Solvent method
Polyacrylonitrile (PAN)	Graphene/graphite/GO	Electrospinning, Solvent method
Polycarbonate	Graphene/Reduced graphene	Melt method
Rubber/natural rubber	Graphene/Reduced graphene	Melt method
Poly(vinylidine fluride) (PVDF)	Reduced graphene	Solvent, melt method

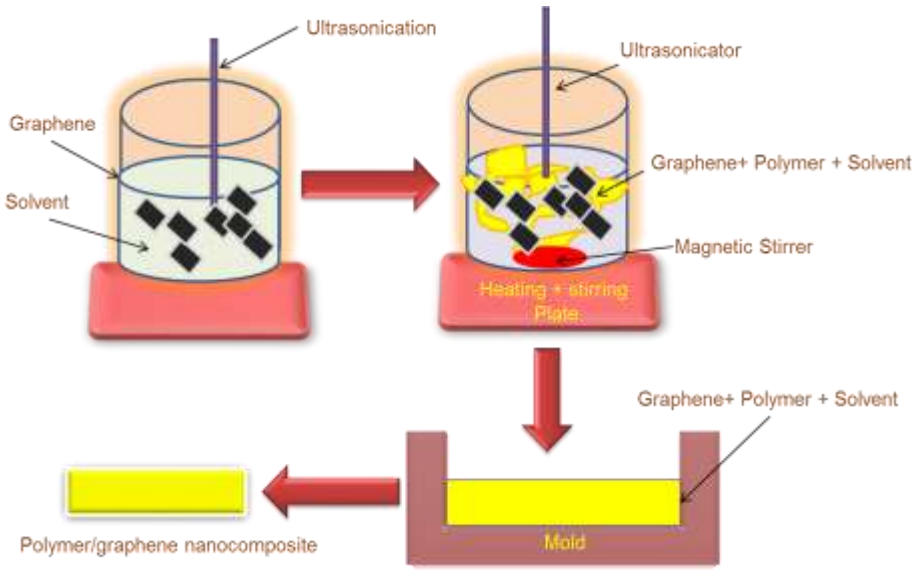

Fig. 2 Conventional solution technique for graphene dispersion in polymer.

5. Application of polymer/graphene nanocomposite

Graphene has superior properties compared with polymers which are also reflected in polymer/graphene nanocomposite. These nanocomposites show superior mechanical, thermal, gas barrier, electrical and flame retardant properties relative to neat polymers. Polymer/graphene has also established high electron mobility at room temperature. The probable uses of polymer/graphene nanocomposite include energy storage devices such as lithium ion batteries, conducting electrodes, electronic devices, and solar cell and dye-sensitized solar cell.

5.1. Electronic devices

With the advancement in experimental nanosciences, the electronic properties of graphene nanosheets have gained attention [53, 54]. Field-effect transistor (FET) is an excellent example of polymer/graphene nanocomposite-based electronics (Fig. 3).

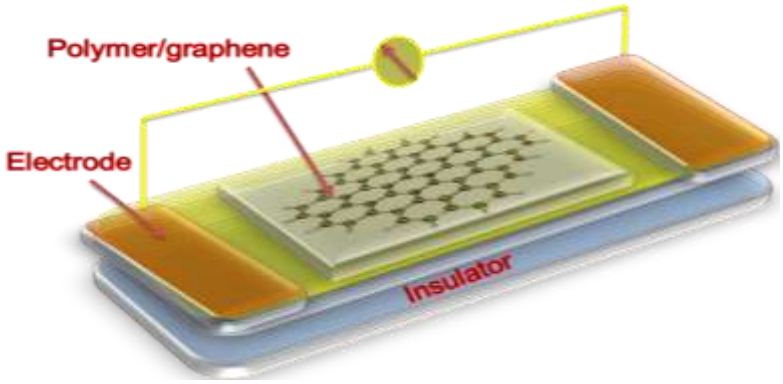

Fig. 3 Field-effect transistor.

Fabrication of ultra-sensitive sensors has also been effectively established. Various techniques for micromechanical cleavage have been developed to achieve fine single layer graphene nanosheets [55]. Yield of the methods have also been tried to improve. Among greater yield methods, chemical reduction of graphene oxide to graphene is successful. However, these graphene sheets have basal plane of carbon atoms decorated with epoxide and hydroxyl groups [56]. Due to hydrophilic character, the nanosheet with functional groups may decrease the interplane forces [57]. At present, graphene is more preferred in electronic devices compared with other thermally conductive fillers such as graphite, carbon nanotube, carbon black, silicon carbide, silicon nitride, and nanodiamond. Thin films of polymer/graphene nanocomposite have shown cost efficiency, elasticity, transparency, and electric and magnetic properties [58]. In electronic applications, mostly thermoset polymers (epoxy, phenolic) are favored because they do not change their form or do not melt at high temperature. However, thermoplastic polymers are least recommended for prolonged device stability.

5.2. Li-ion battery

Application in Li-ion battery is a greatest achievement of polymer/graphene nanocomposite. Initial efforts involve the replacement of liquid electrolyte in battery with solid polymer (Fig. 4). The battery electrolytes were first established in 1970s [59].

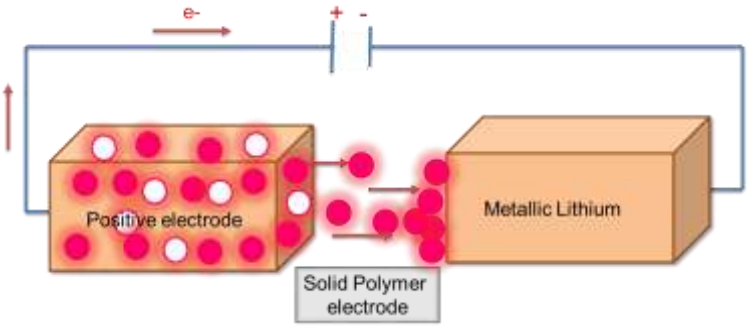

Fig. 4 Schematic representation of solid polymer lithium batteries.

Polyethylene oxide (PEO) was complexed with alkali metal salts, and the electrolyte showed high ionic conductivity around 10^{-3} Scm^{-1}. The polymers electrolytes are classified as pure solid polymer electrolyte and gel polymer electrolytes. Poly(methyl metacrylate), polyacrylonitrile, and poly(vinylidene fluoride), and PEO has been successfully used as polymer electrolytes. In solid polymer electrolytes, lithium salts ($LiPF_6$, $LiBF_4$, $LiCF_3SO_3$, $LiClO_4$) have been dispersed in polymers. These solid polymer electrolytes show ionic conductivity in the range of 10^{-8} to 10^{-4} Scm^{-1}. Gelled polymer electrolyte consists of plasticizer or solvent in polymer matrix. These electrolytes have ambient ionic conductivity of ~10^{-3} S cm^{-1}. The ionic conductivity of these electrolytes also depends on the molecular weight of polymers. Consequently, ionic conductivity, mechanical, and physical properties of solid polymer electrolyte are less than gelled polymer electrolyte [60]. Reduced graphene oxide (RGO) and thermally reduced graphene oxide (TRGO) have also been used in Li-ion batteries. Incorporation of silica/titania particles have also been used to improve the efficiency of nano-hybrid [61]. In PEO and ethylene glycol, GO nanosheets have been directly converted to graphene by simple sonication method [62]. Graphene has tendency to chemi- and physisorb oxygen from water, oxygen and CO_2 species [63, 64].

5.3. Solar cell

In photovoltaic devices, graphene has excellent application due to low-cost, transparency, flexibility, and wonderful electron-transport properties and very high exporter mobility (Fig. 5).

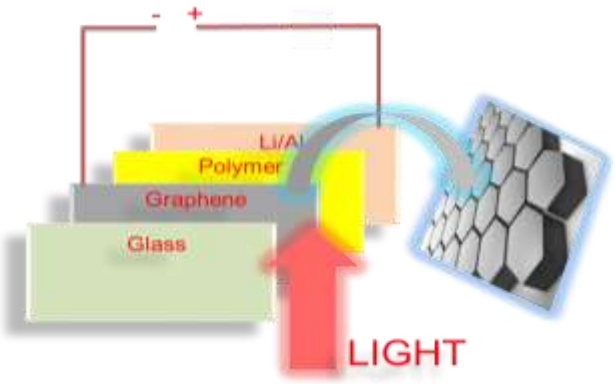

Fig. 5 Polymer/graphene solar cell.

Recently, several graphene-based solar cells have been reported [65]. In electrodes, graphene is a perfect 2D material to form films with good transparency, high conductivity, and low roughness. Graphene oxide reduction to graphene has been employed to form large-area, transparent, and conductive thin graphene films. Such graphene films have thickness of 10 nm, conductivity ~550 Scm^{-1} and transparency of 70%. In dye-sensitized solar cells, graphene materials have been used as window electrode [66]. In electrodes, polymer film width was attuned as 20 mm. Later the graphene particles were dropped on the polymer layer. Vacuum evaporation technique has been used to form solar cell structures. The polymer/graphene layer is usually coated on glass substrate. In bulk hetero-junction (BHJ) solar cells, graphene nanocomposite may act as a hole-extraction layer [67-70].

6. Conclusions

In this review, properties of graphene as reinforcement have been discussed. Graphene has been argued with reference to polymer property improvement including mechanical, thermal, and electrical. The superimposing effect of graphene and polymer in refining the thermal, conducting, and physical properties of polymers has been included. Considerable improvement in mechanical, electrical, and thermal properties was observed in polymer nanocomposite reinforced with graphene. The conductivity predisposes to high solar cell efficiency of polymer/graphene materials. These nanocomposite have been prepared using variety of techniques such as *in situ* polymerization, melt method, solution route, latex technology and other methods. Generally, dispersion of graphene is poor in polymer matrices. For this purpose, surface-modified graphene has been employed to form polymer/graphene nanocomposite. Functional graphene may easily disperse in organic polymer matrices. The polar interaction between graphene and organic polymers are accountable for better filler dispersion in the polymers. Potential application of polymer/graphene nanocomposite has been discussed in electronic devices, batteries, and solar cell.

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