

Graphene-based polymeric nano-composites: an introspection into functionalization, processing techniques and biomedical applications

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ABSTRACT

Although, there have been numerous efforts in synthesis of polymers, their mechanical properties have limited their applications. Graphene has been investigated for excellent properties such as superior mechanical properties, high thermal conductivity that has attracted the attention of scientific community to employ graphene as a filler material in polymeric matrices to form composites with multi-functional capabilities. The excellent properties possessed by Graphene has motivated users to fabricate flexible nanocomposites that can be used for applications requiring superior mechanical, chemical and thermal performances. Characteristics of both the components if explored synergistically through proper structural and interfacial organization. The investigation in this direction has resulted into combination of graphene with variety of polymeric materials and hence the development of different graphene-based nanocomposites. The present work reviews the application of graphene-based nanocomposites in the biomedical domain. With this objective, the polymeric matrices suitable for biomedical applications as well as the techniques of producing graphene polymeric nanocomposites have been discussed. Finally the application particularly in biosensing, wound healing and drug delivery system has been discussed.

Keywords: *polymer, graphene, mechanical properties, thermal conductivity composites nanocomposites graphene-based nanocomposites biomedical applications, bio sensing, wound healing, drug delivery system.*

1. INTRODUCTION

Nanocomposites developed from polymeric matrices consists of one or more than one polymeric matrices and reinforcing agents with nano dimensions below 100 nm [1]. Polymeric matrices have reasonable mechanical properties and can be processed as well as shaped conveniently. Furthermore, the control of surface as well as interfacial chemistry is much easier with the polymeric matrices. Typical reinforcing agents may be fibers, flakes and other particles with higher properties [2-4]. Nano fillers have large surface areas that is instrumental in enhancing the interfacial strength between the filler and the matrix. Moreover, superior composite properties are achievable at low reinforcement loads. As a result, the fabricated nanocomposites are expected to reach significant improvement in properties with controllable interfacial interactions [5].

The frequently used nanomaterial are carbon-based as for instance: carbon black, carbon nanotubes. Graphene has excellent

properties that have attracted the attention of academic and industrial personnel to employ it as one of the suitable reinforcing agents in polymeric matrices. In comparison to other carbon materials, graphene has higher surface-to-volume ratio and has superior properties. Therefore, graphene is one of the interesting reinforcing agents. However, there are critical issues in the production of functional nanocomposites: homogeneous distribution of graphene into polymeric matrices and interfacial bonding.

The present work reviews on graphene based nanocomposites, their properties and the techniques of processing. The scope of the article also encompasses discussion on surface modification of graphene. Main applications in biomedical applications are also discussed. The work finally concludes with the concluding remarks.

2. GRAPHENE AND SURFACE MODIFICATION

Graphene is structurally a planar sheet comprising of sp^2 hybridized carbon atoms that are arranged into a 2D hexagonal lattice. It is constituted by two sub-lattices of sigma bonded carbon, wherein each carbon showcases out-of-plane π orbital. Each atom of carbon is connected to three neighboring atoms through strong covalent bonds. Bond length of each covalent σ bond being 0.142 nm [15]. The van der Waals thickness of graphene sheet is around 0.34 nm making it the thinnest material on the planet till date. Graphene can be obtained synthetically

through oxidation of graphite and through exfoliation process of graphite. This process produces graphene in bulk at high yield.

Graphene exhibits high elastic modulus, high tensile strength, and ability to sustain high densities of electric current and very high thermal conductivity [6-7]. Graphene has higher impermeability to any gases and large specific area of 2630 m^2/g . Furthermore, graphene has a higher flexibility.

Different in vitro studies have been carried out in the past that reveal the biocompatibility of graphene and its derivatives. Moreover, it presents low toxicity with the various cells [8-9].

However the results present a gloomy picturesque. Decreased cell viability has been observed by various in scientific community with increased content of graphene and its interaction time. Furthermore, the toxicity associated with functionalized graphene is more than that associated with the non-functionalized one. Therefore functionalized graphene a better prospect for improved biocompatibility [8]. The levels of toxicity are also reduced when the fillers are included in biomaterials. However, one of the major drawbacks related to graphene is its ability to agglomerate to form graphite. Graphene has poor dispersion in aqueous as well as organic solvents. However, this drawback could be overcome through functionalization process.

To produce graphene-based nanocomposites exhibiting higher performance, it is essential to disperse graphene sheets in polymeric matrices. Also, the establishment of better interaction/interfacial bonding is another essential parameter for producing high performance nanocomposites [10-11]. The application of graphene has been restricted however due to its hydrophobic behavior. Hydrophobicity of graphene can be improved through the addition of graphene oxide or through its functionalization. Functionalization can be achieved through attachment of functional group to the graphene surface that presents reactivity to certain compounds or molecules [11-12]. The improved hydrophobic graphene disperses stably within the polymeric matrices [6, 13]. As a result, the nanocomposites produced possesses higher structural properties even at lower reinforcement loadings. There are certain solvent-assisted techniques that have been used to promote chemical functionalization of graphene. This technique includes filtration, spin-coating and layer-by-layer assembly. There are different covalent and non-covalent modification techniques for graphene [1, 14-15].

Oxidation of graphite and hence graphene oxide formation leads to covalent functionalization of graphene. The covalent functionalization involves oxidation of edges of graphene nanosheets as well as that of the associated basal planes. As a result of oxidation, strong and stable interfaces are established with the polymeric matrices. There are other functionalization process that modifies graphene covalently that includes dienophiles and free radicals covalent functionalization of graphene oxide.

Solubility characteristics of graphene with other materials have been reported to be enhanced through free radical reactions. Free radicals in large quantum are generated when heating benzoyl peroxide and react with graphene to establish covalent bonds [16, 14]. Diazonium salts were reported to chemically functionalize graphene nanoribbons [17]. In this case, graphene nanoribbons were made to react with 4-nitrobenzene diazonium tetrafluoroborate. The reaction was found to be rapid and it also altered the electrical properties of devices based on graphene.

A band gap was engineered by Niyogi et al. [14, 18] through covalent functionalization approach. The reaction takes place rapidly by transfer of electron to p-nitrobenzenediazonium from graphene. This results in attachment of nitrophenyl group onto the graphene sheets. A band gap of around 0.4 eV was introduced due to the hybridization of carbon graphene to sp^3 . Another research work that could efficiently functionalize

graphene was carried out by Fang et al. [19]. The graphene surface was covalently attached to hydroxylated aryl groups. Diazonium addition reaction was carried out to transfer atom from polystyrene and hence resulting in radical polymerization. The polystyrene was able to effectively bond with the graphene atoms. Therefore, as a result of effective transfer mechanism it was possible to develop nanocomposites with enhanced mechanical properties. Reaction of graphene sheets and benzoyl peroxide is another alternative approach to carry out free radical addition reaction. A photochemical reaction between benzoyl peroxide and graphene was reported by Liu et al. [20]. Graphene sheets were deposited on substrate of silicon and the combination was immersed in toluene solution. Ar-ion laser beam initiated the reaction. The major reason behind radical generation mechanism as described in the work was the hot electron that initiates the transfer of electron to benzoyl peroxide from graphene that was photo excited. Other research studies in the domain of free radical addition reaction for graphene functionalization were reported by Wang et al. [21] and Bekyarova et al. [22].

Employability of dienophile reaction is one of the covalent modification mechanism. The reaction between sp^2 carbon atoms with dienophiles is prominently discussed in the domain of chemical modification of graphene. Within this class the most successful reaction is 1,3 dipolar cycloaddition of azomethine ylides. This reaction is applied to the functionalization of fullerenes and carbon nanotubes. This most widely used reaction can be employed for a number of organic derivatives.

Pyrrolidine groups have been used to decorate the graphene sheet through the condensation of sarcosine and 3,4-dihydroxybenzaldehyde [23]. Dihydroxyl phenyl groups were added to the graphene sheets owing to the formation of pyrrolidine derivatives. As a result of introduction onto the graphene sheets, the stability was enhanced when introduced in polar solvents such as N,N-dimethylpyrrolidone and ethanol.

Graphene Dispersion and its exfoliation in N-methylpyrrolidone resulting in few-layer graphene were functionalized by Quintana et al. [24] using 1,3-dipolar cycloaddition reaction carried out with azomethine ylides. Condensation of paraformaldehyde with alpha-amino acid aided in functionalization of graphene. The condensation reaction was followed with deprotection of t-butyl carbamate group.

Another strategy of covalent functionalization of graphene was reported based on nitrenes being generated from the azide groups through irradiation process or thermolysis mechanism [25]. 2+1 cycloaddition of nitrenes was followed to introduce a variety of polymeric chains and functional groups onto the graphene sheets. Functionalization of graphene with phenylalanine was reported by Strom et al. [26]. This was carried out through 2+1 nitrenecycloaddition. Enhanced water solubility as well as biocompatibility of graphene surface have been reported with the addition of amino acid groups [14, 26].

Another popular technique of producing graphene is through the preparation of graphene oxide its exfoliation and finally its reduction [25]. However, the properties of graphene oxide in reduced form are affected to a great extent owing to the presence of oxygen groups and structural defects. The mechanical as well as electrical properties were the majorly affected ones.

Numerous approaches have been reported to produce graphene oxide: Brodie method [27], Staudenmaier method [28] and modified Hummers methodology [29]. The produced graphene oxide is converted to graphene oxide in reduced form which may be carried out using various techniques: electrochemical, chemical and thermal.

Graphene oxide consists of different functional groups such as lactone, carboxyl, carbonyl, ether and hydroxyl that have their specific chemical reactivity and can be used to establish covalent bonds with other molecules. The covalent bonding is formed through different reactions such as amidation, cycloaddition, esterification and silanization [30].

Ligands, biomolecules, chromophores and polymers have been reported to be attached with graphene oxide through the amidation reaction between amine groups and the associated carboxyl group of graphene oxide. Natural amino acids that are biocompatible were used for carrying out covalent functionalization of graphene sheets [31]. A promising composite material for drug delivery application was developed by attaching amine-terminated polyethylene glycol to the graphene oxide nanoplatelets [32]. Amidation reaction was reported between carboxylated graphene oxide [16] and casein phosphopeptides to produce bifunctionalized graphene.

Another approach for modification of graphene oxide is through esterification reactions. The reaction involves the reaction between carboxylic group present in graphene oxide and the alcoholic groups of the molecules to be bonded. Graphene oxide and polycarbonate were bonded effectively through esterification process. The development was useful for tissue engineering applications [33]. Lonkar et al. [16] reviewed the alternative methods for carrying out functionalization of graphene oxide, in particular the esterification process.

Another prominent method of graphene functionalization involves the non-covalent modification process. This involves the adsorption of specific molecules onto the graphene surface. π - π interactions form the basis of the adsorption process. The adsorption process results into immobilization of the different molecules: biomolecules, aromatic molecules, surfactants etc. the main advantage of non-covalent functionalization process is that the process doesn't affect the sp^2 carbon hexagonal lattice associated with graphene [12, 33, 34]. Georgakilas et al. [35] presented a review report on modification of graphene oxide and graphene non-covalently. Restacking of graphene sheets have

been avoided through the employability of stabilizers in the form of aromatic molecules: pyrene, porphyrin derivatives, ionic liquids, coronene, triphenylene, anthracene, quinolone and fluorinated benzene derivatives and pyridine. Assistance of sonication process is used to attach these aromatic molecules onto the graphene surface. Single and several layered graphene sheets can be functionalized through pyrene derivatives [36]. Pyrene derivatives have been used in order to investigate the π - π interactions between aromatic molecules and reduced graphene oxide [37-38]. Employability of aluminum phthalocyanine in sulfonated form was analyzed to delineate the ability of reduced graphene oxide, single wall carbon nanotubes and graphene and to undergo π - π interactions. Wang et al. [39] studied the adsorption capacity of graphene oxide in reduced form and graphene using pyrene derivatives, phenanthrene, naphthalene and aromatic hydrocarbons. It was reported that reduced graphene oxide had more adsorption capability in comparison to graphene oxide.

Graphene presents enhanced interactions with hydrophobic molecules such as ionic liquids and surfactants. This is due to the reason that graphene exhibits hydrophobic character. These reactions are useful in mediating graphene in both the organic as well as aqueous media [35]. Amphiphilic character is exhibited by surfactants [40] and can be classified into non-ionic, zwitterionic, ionic on the basis of polar head group [35]. Therefore, ionic interaction is only the medium for surfactant aided functionalization of graphene sheets [41]. Some of the popularly used graphene stabilizers and dispersants are amphiphilic molecules and macromolecules such as sodium dodecylbenzenesulfonate, sodium cholate, albumin, lignin and cellulosic derivatives [35]. It was observed and therefore revealed that the best performance in dispersing graphene was achieved with surfactants having hydrophobic tails and π - π interaction capabilities [42]. Dispersion of graphene is enhanced when polar part interacts with polar solvents while the aromatic part interacts with the reduced graphene oxide or graphene [35, 41]. Fernandez-Merino et al. [43] investigated the stabilization of graphene oxide sheets by using different surfactants under various operating conditions. Paper-like films were produced with two different surfactants and the film prepared with poly sodium 4-styrenesulfonate and polyoxyethylenesorbitanmonooleate were revealed to have higher performance in terms of capacitance in comparison to that produced without the aid of surfactants [43].

3. METHODS FOR PREPARATION OF GRAPHENE BASED POLYMERIC NANOCOMPOSITES

Significant improvement in mechanical properties has been reported with graphene-based polymeric composites in comparison to other conventional polymers [1]. However, the improvement in mechanical properties is dependent to a greater extent on the processing conditions used in their development [2]. The degree of graphene dispersion dictates the effectiveness of mechanical improvements. The level also dictates the interfacial strength, spatial organization which in turn dictates the strength, stiffness, elongation and toughness of polymeric composite materials [2]. There is a number of factors that are to be considered while selecting a suitable method for preparation of graphene based polymeric composites: hydrophobicity, polarity,

molecular weight and type of surface modification process used for graphene. The three most commonly used methods for graphene based polymeric composite materials are in situ polymerization [44-46], melt blending [47-49] and solution mixing [33, 50-51, 52-58]. Layer-by-layer technique of preparation is another occasionally reported method of preparing graphene-based nanocomposites [2].

Solution mixing method has been used mainly for producing graphene based polymeric nanocomposites for biomedical application. The process facilitates in the separation of graphene sheets [59]. The filler dispersion in matrix is maximized as the process of solution mixing uses pre-suspended graphene

sheets with a single layer configuration. Solution casting method is accomplished via three stages: dispersion and dissolution of filler in a suitably chosen solvent, inclusion of polymeric matrix and distillation process to remove the solvent matter [6]. During the development process, graphene sheets are coated by the polymer and reassembling of graphene sheets on evaporation of solvent. This results in interconnection with the polymeric matrix and hence formation of nanocomposites. Good dispersion is achieved if the polymeric material as well as filler have excellent solvent compatibility. Commonly used solvents are toluene, dimethyl formamide, chloroform and water acetone [1, 6]. Some of the advantages associated with the process include: fast fabrication rate, simplicity, higher control on the behavior of the constituting components and the effectiveness of dispersion. The disadvantages are the challenges associated with finding common solvents, difficulty associated with solvent removal, thin-film limitations, solvent evaporation and related issues of aggregation [2, 60-61]. Some of the examples of nanocomposites prepared include graphene reduced-poly(vinyl alcohol) nanocomposites and poly(vinylidene fluoride)-thermally reduced graphene nanocomposites [62].

In situ polymerization process on the other hand requires the employability of a high temperature reactor, an initiator and

monomers. The technique of production incepts with the dispersion and dissolution of graphene or its functionalized form in a liquid monomer. Next, an initiator suitable selected is diffused and the polymerization process is initiated by radiation or heat. Various condensation reactions aid in homogeneous dispersion and dissolution of nanofillers from graphene in polymeric matrix and hence formation of covalent bonds between them. One of the major limitations associated with this process is the higher cost of monomer and higher vessel temperature.

Another prominently used approach for production of graphene-based nanocomposites is that of melt-mixing. The process is solvent-free and entails mixing of graphene or modified graphene under conditions of high mechanical shear. This is achieved either through a blending mixer or through screw extruder [2, 5]. Polymeric material liquefies owing to the higher temperature which also results in intercalation or dispersion of reduced graphene sheets or graphene oxide. The aforementioned technique is applicable to both the polar as well as non-polar polymeric materials [62]. Nanocomposites can be produced on large scales and are also economical in comparison to the solution mixing process. However, the effectiveness of dispersing graphene is lesser in case of melt-mixing process owing to the higher associated viscosity.

4. APPLICATIONS OF GRAPHENE-BASED NANOCOMPOSITES IN BIOMEDICAL DOMAIN

Graphene-based nanocomposites have been employed in diverse engineering applications as sensors, energy technology and nanoelectronics. Other than the aforementioned applications, biomedical domain is another interesting wherein the graphene based nanocomposites find application. Several biomedical applications include antibacterial materials, cancer photothermal therapy, drug delivery [8-9, 63], bioimaging and biosensing [12]. Regenerative medicine as well as biotechnology are other prominent application areas besides the aforementioned biomedical applications. Different properties as thermal, mechanical and electrical properties of biomaterials are enhanced through the graphene-based composites and also aids in promoting growth and cellular attachment in surface of biomaterials. These also result in the production of efficient biosensors.

Graphene-based nanomaterial for drug delivery.

Availability of large surface area of sp^2 hybridized carbon atom of graphene is one of the major characteristic features that make graphene-based nanocomposites suitable for drug delivery applications. Planar aromatics are showcased by the majority of drugs such as doxorubicin when complexed with graphene. This is because of the π - π interactions between the graphene surface and aromatic rings [64]. Delivery of Ibuprofen was reported to be delivered by employing CHI-grafer graphene oxide [65]. Significant enhancement in anticancer efficacy was reported for sequential delivery of siRNA and doxorubicin with the employability of graphene oxide-PEI nanocarrier.

Much interest has been received by gene therapy that aids in the treatment of a number of diseases as for instance cancer and Parkinson's disease. Generation of safe gene vector is one of the major roadblocks associated with gene therapy. The purpose of safe gene vector is to protect the degradation of DNA. Studies have revealed the binding of graphene to single-stranded DNA.

However, the studies also revealed infectivity of graphene to bind with double-stranded DNA. Reports and studies have also been published on the ability of graphene to protect oligonucleotides from enzymatic cleavage. Therefore, graphene oxide in combination with PEI has been normally explored for delivery of plasmid DNA [9, 66-67].

Tissue engineering application.

Graphene-based nanocomposites can play a prominent role in enhanced differentiation, proper adhesion and sustained proliferation in the domain of tissue engineering owing to the properties as flexibility, adaptability to irregular or flat faced surfaces, high elasticity [9]. Reports have been published on the biocompatibility aspect of graphene-based nanocomposites [8]. Graphene oxide/calcium carbonate was synthesized by Kim et al. [68] that revealed to showcase better compatibility with osteoblast cells and also enhanced bioactivity associated with bone. Enhanced proliferation and improved cellular attachment were observed with CHI-Graphene oxide scaffolds. Decreased rate of chitosan degradation was also reported which is one of the major issues for designing of scaffolds for tissue engineering application [69]. Complete wound healing was reported with the employability of CHI-PVA membrane consisting of graphene [64].

Molecular imaging.

Expression of biomarkers is measured through the introduction of molecular imaging agents through molecular imaging. The expressions are measured throughout the different stages of diseases. The research community has recently focused on the applicability of graphene, reduced graphene oxide with different molecular imaging techniques as photoacoustic, radionuclide-based imaging and magnetic resonance imaging. Recently, nanoscale graphene oxide was functionalized covalently with PEG

resulting in PEGylated graphene oxide exhibiting photoluminescence in both the infrared and visible ranges. Graphene oxide/PEI composite was used as a bioimaging tool [66].

Biosensing application.

Graphene-based nanocomposites have shown potential application to be used effectively as biosensors. Biosensors aid in selective, sensitive and accurate detection of different biomarkers. These biosensors have the potential ability to detect glucose [70], adenosine triphosphate, nicotinamide adenine dinucleotide, cholesterol, hemoglobin, uric acid [71], ascorbic acid, heavy metal ions [1, 72] etc. Graphene-based biosensors were reported for cancer-cell detection by Feng et al. [73]. Graphene/PET electrodes were developed to enhance cell-to-cell interactions [74]. Some of

5. CONCLUSIONS

Present work delineates the different aspects associated with graphene-based polymeric nanocomposites: functionalization, processing techniques and biomedical applications. Graphene has excellent properties as mechanical, thermal, electrical, higher surface-to-volume ratio. As a result of these excellent properties, nanotechnology realm has been revolutionized. Stable dispersions have been obtained with graphene through the functionalization process. Functionalization process has also resulted in enhanced compatibility with polymeric matrices. Enhanced graphene is being used for fabrication of varied nanocomposites. Different processing techniques can aid in controlling the properties and characteristic features of graphene-based nanocomposites.

Graphene has been used for biomedical application that has the capability to regenerate some specific tissues and detect biomarkers. Graphene composites have been used for drug delivery and wound healing owing to their inherent antibacterial capabilities and affinity to certain drugs. The enhanced

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the major contributions related to the application of graphene-based polymeric composites for biosensor application have been tabulated in Table 1.

Table 1. Biosensor applications of graphene-based polymeric nanocomposites.

Reference/s	Type of biosensor	Graphene polymeric composite
73, 74	Cancer cells	Graphene/PET Graphene/PTCA
71	Uric acid	Graphene/CHI
75-76, 70	Glucose	GOD/Pt/graphene/CHI Graphene/GOD/CHI Graphene/PEI/GOD
77	Temperature	Graphene/PVDF/ZnO
78	Hemoglobin	HPCD-GO/TPP
79	Guanine and Adenine	Graphene/Nafion/GC

performance of nanocomposites is due to their good dispersion tendency within the polymeric matrix. Higher specific interfacial area results because of excellent interfacing between polymeric matrix and graphene.

Some of the successful biomedical applications of nanocomposites based on graphene and its derivatives include tissue engineering, cancer therapy, drug delivery, bio imaging and biosensing. Results indicate excellent cytocompatibility, adhesion and proliferation tendency. However, studies concerning the in vitro and in vivo tests remain to be inconclusive. Graphene nanocomposites have shown their acceptability for a diverse biomedical as well as other engineering fields. As for instance, graphene based materials have been used prominently for water treatment especially through membrane separation [80-85]. Energy industries are also reaping the benefits of graphene based polymeric composite materials [86-90].

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