



Review

Review of functionalization, structure and properties of graphene/polymer composite fibers



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ABSTRACT

Fibrous materials usually have good mechanical, heat-resistant, acid-resistant, alkali-resistant and moisture regained properties which originate from its composition, condensed structure and crosslinking styles. However, these materials often lack of good electrical conductivity, flame retardance, anti-static and anti-radiation properties which are desired for varied specific applications. Graphene, as a new emerging nanocarbon material, has some unique properties including superb thermal and electrical conductivity, strong mechanical and anti-corrosive property, extremely high surface area etc. Therefore, graphene has attracted extensive interests in recent years. Upon modification with graphene, fibers exhibit a number of enhanced or new properties such as adsorption performance, anti-bacteria, hydrophobicity and conductivity which are beneficial for broader applications. In this review, the strategies to modify the fibers with graphene and the corresponding effects on the fibers as well as the relevant applications in varied areas were discussed.

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Contents

1. Introduction	30
2. Methodologies to modify graphene	30
2.1. Modification via non-covalent interactions	30
2.1.1. Van der Waals force	30
2.1.2. Electrostatic interaction	31
2.1.3. Hydrogen bonding	31
2.1.4. Coordination bonds	32
2.1.5. π - π stacking interaction	32
2.2. Modification via covalent interactions	32
3. Preparation of graphene/polymer composite fibers	32
3.1. Solvent mixing	33
3.2. Melt processing	34
3.3. <i>In situ</i> polymerization	35
3.4. Coating	37
4. Properties of graphene/polymer composite fiber materials	38
4.1. Mechanical properties	38

Abbreviations: 0-D, zero-dimensional; 1-D, 1-dimensional; 2-D, 2-dimensional; CNTs, carbon nanotubes; GO, graphene oxide; CVD, chemical vapor deposition; RGO, reduced graphene oxide; PVA, poly(vinyl alcohol); pTSA, para toluene sulfonic acid; GN@Pani, graphene/polyaniline; PAA, polyacrylic acid; PAN, polyacrylonitrile; NaAlg, sodium alginate; RAFT, reversible addition fragmentation chain transfer; SWCNT, single walled carbon nanotubes; CA, calcium alginate; GNPs, graphene nanoplatelets; PS, polystyrene; RC, regenerated cellulose; CMG, chemically modified graphene; PA12, polyamide 12; PP, polypropylene; CB, carbon black; SSM, solid state mixing; PET, polyethylene terephthalate; PA6, poly(ϵ -caprolactam); PGO, PAA and GO composite; Py, pyrrole; ODCB, O-dichlorobenzene; MM, traditional melt mixing; G/PPy, graphene/polypyrrole; PLGA, poly(DL-lactic-co-glycolic acid).

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4.1.1.	Solvent processing to mix graphene into fiber	38
4.1.2.	Melt mixing graphene into fiber	39
4.1.3.	<i>In situ</i> polymerization to mix graphene into fiber	39
4.1.4.	Spraying graphene onto fiber	40
4.2.	Electrical conductivity	40
4.3.	Thermal properties	41
4.4.	Other properties	43
5.	Application of graphene/fiber composites	43
6.	Conclusion	44
	Acknowledgements	44
	References	44

1. Introduction

It is well known that zero-dimensional (0-D) nanoparticles, 1-dimensional (1-D) nanotubes or nanowires, and 2-dimensional (2-D) nanosheets have been explored to improve the properties of composite nanofibers, especially carbon-based 1-D nanomaterials such as carbon nanotubes (CNTs) [1–4]. CNTs have proved to be efficient nanofillers to significantly improve nanofibers' mechanical and electrical properties due to their highly conjugated hollow structure, high specific surface area and good conductivity. However, the cost of producing high-quality CNTs in large quantity is high, which limits their wide-spread commercial applications. The latest immersed nanocarbon allotrope, graphene, can be the solution to this bottle-neck. Graphene is a highly conjugated, two-dimensional and single-atom layered carbon nanomaterial, whose research history can be traced back to 1859. The term graphene was coined as a combination of graphite and the suffix-ene by Hanns-Peter Boehm, who described single-layer carbon foils in 1962 [5,6]. A dramatically increasing interest has been paid to graphene after Novoselov and Geim reported on the unusual electronic properties of single layers of the graphite lattice in 2004 [7–9].

The unique plane structure and geometry of monolayer graphene contribute its super properties, including high Young's modulus (~1100 GPa), high fracture strength (~125 GPa), excellent electrical (~ 10^6 S/cm) and thermal conductivity (~5000 W/mK), fast mobility of charge carriers (~200,000 cm²/Vs) and large specific surface area (theoretically calculated value, 2630 m²/g) [10,11]. These remarkable properties of graphene provide infinite possibilities for various applications in many areas such as electronics, energy storage and conversion, biotechnology and especially improvement in composite fiber materials. Graphene can be prepared with various methods, among which the mostly applied methods include exfoliation of graphite, reduction of graphene oxide (GO) and chemical vapor deposition (CVD). However, one major problem that needs to be tackled for graphene before its applications, is the low dispersibility in common organic and inorganic solvents. In some cases the good dispersion of graphene in common solvents is a crucial move toward the formation of homogeneous nanocomposites. Therefore, modification of graphene to tailor its solubility at will is critical for varied commercial applications.

Graphene can usually be modified with covalent and non-covalent methods. Non-covalent methods include π - π stacking interactions, electrostatic interaction, hydrogen bonding, coordination bonds and van der Waals force. This modification method can maximally preserve graphene's natural structure, however, the interactions between functionalities and graphene surface are relatively weak; therefore it is not suitable for some applications where strong interactions are required. Covalent methods can be used to create the composites with strong interactions between graphene and the modifier, however, graphene's original structure

is usually destructed, leading to compromised electrical conductivity and mechanical properties.

Polymeric fibers, including natural (cotton, linen, silk, and hair) and synthetic fibers (polyester, polyamide, polyacrylonitrile, polyethylene and polypropylene etc.), are a very important part of modern life and extensively used in varied areas such as textiles, packaging and medical technology. Their wide-spread application is not only largely due to their high strength and low density but also due to their durability, abrasion resistance, and chemical and environmental stability. In addition, conductive, thermostable properties as well as toughness can also be required for some special applications. Incorporation of nanofillers such as CNTs and graphene, into these fibers can improve the mechanical properties and enable the multi-functionality required for electrical energy storage, sensing and actuation [12–14]. Many of the possible applications would greatly benefit from the increased fiber toughness, which is the ability to absorb mechanical energy before rupture. Graphene being used to reinforce the property of fabric polymer materials has been studied by more and more researchers [15,16].

Composite fibers can be prepared mainly in two processes, mixing and spinning as shown in Fig. 1. Graphene can be added during the mixing process in three different ways: solvent mixing, melt processing and *in situ* polymerization. Some researchers also tried to coat graphene onto the fiber surface after spinning. There are also different spinning methods to prepare graphene/polymer fibers: wet-spinning, melt-spinning, and electrical-spinning. In this review, we mainly discuss the preparation, properties and applications of graphene reinforced polymeric fibers. The methods for the modification of graphene are also briefly highlighted.

2. Methodologies to modify graphene

2.1. Modification via non-covalent interactions

2.1.1. Van der Waals force

Initial methods for preparing graphene/polymer composites were based on simple physical mixing, where the graphene sheets worked as conductive fillers to produce nanocomposites with very low percolation concentrations. Percolation concentration refers to an onset concentration at which the conductive filler forms an infinite network of connected paths through the insulating matrix and direct or alternating current electrical conductivity of composite materials takes place [17–19]. Melt processing, solvent processing and *in situ* polymerization are the mostly adopted methods to achieve physically mixed graphene composites [20]. Solvent processing can make graphene disperse in polymers uniformly [21]. Furthermore, mixing GO with polymers in proper solvent followed by addition of reductant is another good way to prepare homogeneous graphene/polymer composites [22]. *In situ* polymerization can be employed to prepare more uniform graphene/polymer composites with stronger interaction between graphene and polymer

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