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Electrospun Graphene Oxide-Based Nanofibres

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Abstract

Graphene and graphene oxide have been the forefront of research in the field of carbon nanostructures. In this chapter, the authors attempt to review and discuss works on electrospinning of graphene oxide-based nanofibre materials, mainly focusing on their combination with polymeric materials. Explanation and insights towards their method of preparation, properties and applications will be discussed. The conclusion section will give some closing remarks regarding the future direction of graphene-based nanofibres.

Keywords: graphene oxide, electrospinning, nanofibres, nanocomposites, nanomaterials, polymer matrices

1. Introduction

The discovery of graphene around this decade changed the focus direction of material science into its unique and magnificent characteristics. Researchers throughout the world are competing in finding ways to synthesis and explore the possible applications of graphene-based materials. Graphene oxide (GO) is an intermediate material that has better hydrophilicity and easier modification due to the presence of oxygen functionalities, compared to graphene. A broad range of applications have been reported over the years, including tissue engineering, sensor, power storage, coating and photonics. Electrospinning is one of the techniques that make use of GO materials in nanofibrous composites. This state-of-the art technique has been very

popular in facile 1-D nano-scaled fibres production and gives materials their excellent properties in relation to high specific surface area.

2. Graphene oxide

The novel nanostructure of graphene has been showing promising characteristics not only in fundamental science but also in variety of applications. Its remarkable chemical and physical properties urged into many attempts of synthesizing graphene in mass production. However, it fails to give adequate productivity for large-scale use [1]. Graphene is not easy to be handled when it comes to incorporating it with other materials and to transform it into other form. Current development in this field recognizes graphene oxide (GO) as the best alternative to alleviate this problem, by having a more solvent friendly structure [2]. The solubility of GO in water and other solvents is likely in favour to the large commercialization of this novel material.

2.1. Structure

GO is basically a graphene sheet with oxygen functional groups, either epoxy, carboxyl or hydroxyl. It can be easily exfoliated from graphite through oxidation and sonication processes. Since graphene is a one-atomic layer of graphite, the structure of GO also consists in exfoliated layers of graphite oxide. Simplistically, the structural process is shown in **Figure 1**. The carbon compounds of **Figure 1** are organized in a honey-comb structure. One sheet of graphene contains six-carbon-atom hexagons, bonded by strong π and σ bonds.
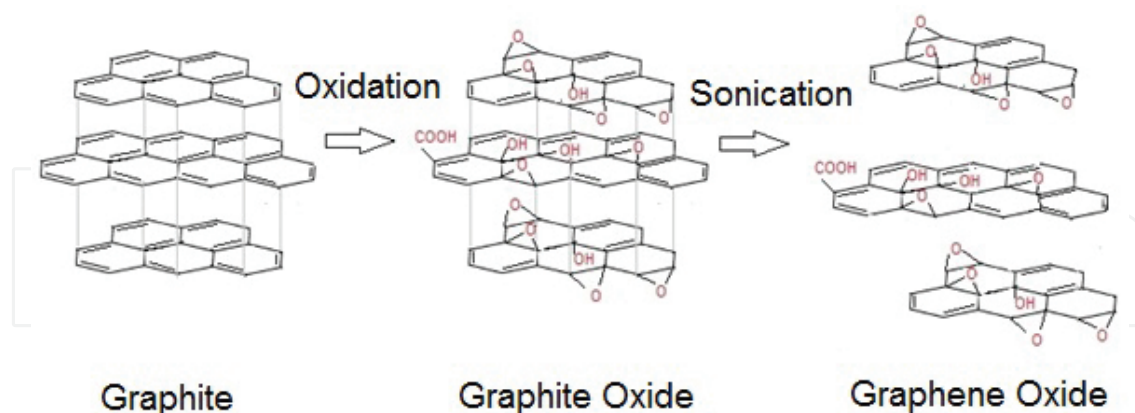
Figure 1. Structural process of graphene oxide.

The synthesis of GO involves oxidation of graphite into graphite oxide and exfoliation of graphite oxide into graphene oxide (**Figure 1**). There are several routes that can be utilized to oxidize graphite. The Brodie method, Brodie–Staudenmaier method and Hummers method are the earliest routes developed, but they lead to the release of toxic gases like NO_x and ClO_x [3]. Later, a typical modified Hummers method was introduced, and it has been used in many

reported works in literature [4–6]. In general, graphite–graphene oxide conversion process can be summarized down into three steps [7]. First, the conversion of graphite into graphite intercalation compound (H_2SO_4–GIC), then, as a second step, H_2SO_4–GIC is converted into pristine graphite oxide (PGO). Finally, the conversion of PGO into GO takes place by the reaction of PGO with water.

The structure of GO on its own has become a research area interest among material scientists. This is due to the complexity of its non-stoichiometric composition [8]. The most recent and acceptable structure is constructed by Lerf and Klinowski. The Lerf–Klinowski model ignores the lattice-based model and focuses on a nonstoichiometric-amorphous alternative. It contains unoxidized benzene rings and aliphatic six-membered rings with epoxy, hydroxyl, carbonyl and/or carboxyl groups on the graphene sheet or at the graphene edges. Each oxygen atom is covalently bonded to two carbon atoms, forming an epoxy ring. Reduced graphene oxide (rGO) structure contains both sp^2 and sp^3 hybridized carbons and a high carbon/oxygen atoms ratio (C:O). sp^2 carbons bind to three other atoms, sharing a double bond with one of them, while a sp^3 carbon binds to a four other atoms.

A study on atomic and electronic structure of GO using annular dark field (ADF) imaging and electron energy loss spectroscopy (EELS) was conducted by Mkhoyan et al. in 2009 [9]. They found out that oxygen atoms randomly attach to graphene sites and convert sp^2 carbon bonds into sp^3 bonds. By ab initio calculated structure, two carbon atoms bonded to an oxygen atom are pulled above graphene plane. This configuration is fairly stable with −3.12 eV absorption energy. The bond length between these two carbon atoms expands to 1.514 Å from graphene 1.407 Å [9]. This is close to the diamond sp^3 bond length of 1.54 Å. The sp^3 hybridization and π and σ bonds of graphene oxide contribute the most to its unique physical and chemical properties. The extent of polar groups on the surface and edges of the graphene oxide depends on the amount of the oxidizing agent and the time and temperature of the oxidation reaction.

2.2. Properties

Spotted and closely studied only since 2004, graphene has already caught wide attention in research history after one by one of its interesting properties were reported. Being a single atomic layer of graphite, graphene is very thin and light. Yet, it is the strongest material ever discovered with intrinsic strength of 42 Nm^{-1} [10], so robust that it is 200 times stronger than steel and can outperform the hardness of diamond. It is transparent, but it can absorb light [11]. Having Young's modulus to approximately 1 TPa [10], it is elastic and pliable but, at the same time, is impermeable to all gases and liquids, except water [12]. It was also reported that graphene possesses superior thermal conductivity in the range of ~$(4.84 \pm 0.44) \times 10^3$ to $(5.30 \pm 0.48) \times 10^3$ W/mK [13]. These values suggest that it can far exceed carbon nanotubes in conducting heat. Another important character of graphene is its exceptional electrical properties for having high intrinsic mobility of 2×10^5 cm^2 $v^{-1}s^{-1}$, compared to the highest known inorganic semiconductor (carbon nanotubes, with ~1×10^5 cm^2 $v^{-1}s^{-1}$) [14, 15].

Despite of being the building block of graphite layers, the synthesis of graphene is not a straight-forward exfoliation process. The method that discovered graphene sheets in 2004,

using cellophane tape, does not appear to be a readily scalable process. Graphite exists in large quantities and is not expensive, yet producing sufficient graphene sheets is still a huge challenge [3]. Even the use of chemical vapour deposition (CVD) to deposit graphene on arbitrary substrates is not possible at low temperatures and is incompatible for mass production processes. Another concern is that graphene is a difficult material to be incorporated into various polymer matrices. Although graphene has unique properties, due to the strong π and σ bonds, these also contribute to its weak reactivity and low solubility in common organic solvents.

In order to prepare graphene–polymer composites or graphene-based hybrid materials, stable dispersion of graphene with other matrices must be obtained. This is when GO becomes useful. The sp^3 hybridization of GO contributes to a larger amount of free bonds, therefore increasing reactivity [16]. When graphite is oxidized, the functional groups make its hydrophilic and improve its exfoliation in aqueous media or organic solutions. The surface functionalization and solvents are the critical factors in determining interaction with functional groups of polymer or other matrixes. Several studies were done to determine the behaviour of GO towards a wide variety of solvents. Paredes et al. [17] studied the dispersibility of GO in acetone, methanol, ethanol, 1-propanol, ethylene glycol, dimethyl sulfoxide (DMSO), N,N-dimethylformamide (DMF), N-methyl-2-pyrrolidone (NMP), pyridine, tetrahydrofuran (THF), dichloromethane (DCM), o-xylene and n-hexane. They have successfully identified some type of solvents (DMF, NMP, THF and ethylene glycol) that can maintain a stable GO solubility. Later, another similar study compared GO and rGO solubility in eighteen different solvents. That investigation suggests that polarity of solvents is not the only factor for good dispersibility [2]. NMP and ethylene glycol, besides water, were double-proved to be the best solvents for long-term stability.

The covalent character of C–O bonds in GO disrupts the sp^2 conjugation of pure graphene and gives GO its insulating properties. Electrical conductivity can be recovered by restoring the π–network. GO, due to the presence of abundant oxygen, can be easily modified. Control of oxidation can provide tunable electronic properties, including the possibility of accessing zero-band gap graphene via bonds. GO, which contains a C/O content ratio of 2/1, is non-conductive (less than 1 S/m). GO can be partially reduced to conductive graphene-like sheets by removing the oxygen-containing groups. Restoration of the π–network and conjugated structure can be obtained by reducing GO to 6/1 of C/O atomic ratio. GO reduction is a process that converts sp^3 to sp^2 carbon, and thus recovers the important electrical properties. Some researchers make use of the oxygenated functionalities to bind with functional groups of conducting polymers, such as polyaniline, to improve the conductivity.
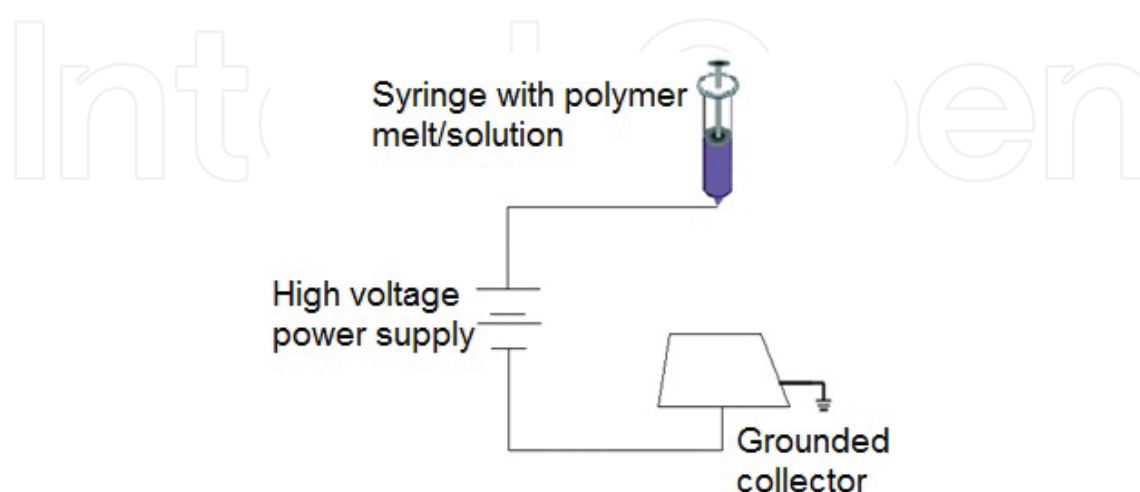
3. Electrospinning

At nanoscale, materials exist in 0, 1, 2, or 3 dimensions. One dimensional nanofibres can be synthesized by several techniques such as phase separation, self-assembly, template synthesis and electrospinning. Among those techniques, electrospinning has been recognized as a facile

process in fabricating polymeric fibres in nano-size [18]. It promptly gained popularity since its establishment, due to its potential implementation in various fields such as tissue engineering, chemical and biological sensing, energy generation and textiles.

3.1. Basics

The first observation on electrospinning dates back from the study of the electrospraying method used by Rayleigh (1897) and Zeleny (1914) [19]. Formhals, however, is the one who patented the electrostatic force experimental setup of polymer filaments production. In 1934, Formhals published a patent in which describing the production of textile yarn from cellulose acetate [20]. Since then, further research was carried out in order to assemble fibre processing using electrostatic force. Later, the study of electrically driven jets, by Taylor in 1969, became a strong foundation in understanding the electrospinning process. Only around 1994, the electrospinning term was used. Along with its popularity and rapid establishment in many fields, electrospinning has undergone many modifications and upgrading, in order to maximize its production and effectiveness in fabricating versatile nanofibres. There are several advanced techniques and setups of electrospinning instruments used favourably according to the required characterizations. Coaxial electrospinning, multilayer and mixed electrospinning, forced air-assisted electrospinning and air-gap electrospinning are a number of innovative processing techniques to enhance the function of electrospun fibres [21].

Basically, the typical basic components of any electrospinning setup are the high-voltage power supply, the spinneret (normally syringe with blunt-tip needle) and the collector (normally grounded). This basic experimental setup is shown in **Figure 2**. The polymer melt or solution placed into the syringe is held by retort stand (for vertical setups) or with a syringe pump (for vertical and horizontal setups). The tip of the syringe needle is connected to the positive electrode of the voltage supply. From here, the polymer liquid is pumped out of the syringe through the needle and brought to the grounded collector beneath the syringe or placed horizontally.

**Figure 2.** Electrospinning basic experimental setup.
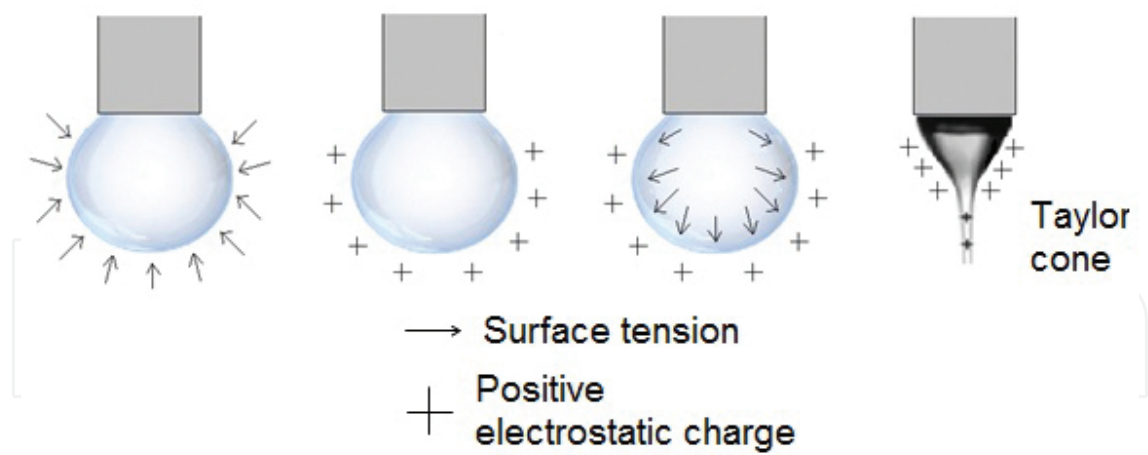
Figure 3. Description of electrospinning principle.

This unique approach for producing continuous fibres from polymer melt or solution, works by the principle of surface tension acting on the polymer drops. By the time, electricity is supplied onto the polymer, the fluid droplet will gain electrostatic charges, being positive charged. Due to the interaction between two forces, a Taylor cone is formed. The increase in high-voltage supply then eventually achieves a certain critical point, at which the electrostatic repulsion can overcome the surface tension holding the polymer droplet. It results in a liquid jet ejection targeting the collector. **Figure 3** shows a clearer depiction of the principle behind electrospinning technique.

The polymer jets onto grounded collector from the spinneret can be divided into two segments. There are streaming jet and whipping jet (**Figure 4**). The streaming jet can be seen with appropriate lighting, while the whipping jet is invisible to the naked eye. The flow of the streaming jet may give a wise initial prediction on the morphology of collected nanofibres. Leach et al., in 2011, indicated that streams behaving inconsistently, non-uniformly, in short
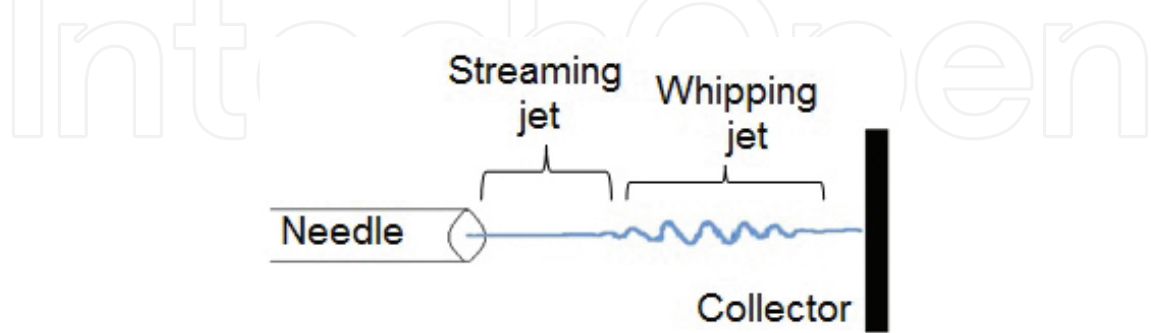
Figure 4. Streaming and whipping jets.

and oscillating, might produce fibres with poor alignment, beading, wavy pattern and splattering [22]. Therefore, researchers engaging in electrospinning studies need to carefully study various parameters (which will be further explained in the next section) before the optimized nanofibre formation procedure can be achieved.

3.2. Working parameters

Electrospinning deals with many working parameters in order to fabricate the required morphology and properties in the as-spun nanofibres. Other than the electrospinning experimental setup, like having different kind of collectors or spinnerets, electrospun nanofibres are also mainly affected by working parameters. The factors to be adjusted are the properties of polymer solution or melt, operational properties and ambient parameters [18, 22]. The electrospinning solution and operational properties are among the major parameters that influence electrospun nanofibres traits. Solution viscosity, surface tension and conductivity, along with electric field strength, distance between spinneret tip with collector and also the flow rate, are thoroughly investigated by researchers to maintain optimized fibre formation. Meanwhile, most researchers prefer to keep ambient parameters such as solution temperature, humidity and surrounding air velocity as constants throughout their studies. **Table 1** displays some factors taken into account for tuning the electrospun fibre morphology. Note that whenever finer or smaller diameter fibres are spun, they tend to get beading. Therefore, it is important for researchers to recognize their requirements and determine the optimized conditions for electrospun fibre formation. There are also some parameters that need minimum requirements to be executed in order to induce electrospinnability. The applied voltage and tip-to-collector distance, for example, are some of those parameters, having certain minimum

Parameter	Effect on fibre morphology
Viscosity	Fibre diameters increase with solution viscosity. Also, higher viscosity results in more beading
Concentration	High solution concentration results in larger fibre diameter
Applied voltage	Higher-voltage supply jets more fluid and cause larger fibre diameter. Increase in electrical potential also results in rougher fibres
Surface tension	Reduction in surface tension minimizes fibre beads
Feed-rate	Low feeding rate: decrease in fibre diameter. High feeding rate: increase in bead generation and Taylor cone instable
Tip-to-collector distance	Minimum distance required for dry fibres. Excessively far distance will lessen the deposited amount of fibres on the collector
Conductivity	Decrease in fibre diameter with increase in conductivity, bending instability and diameter distribution

Table 1. Effect of parameters on fibre morphology [19, 23].

values which differ for every electrospinning material. Other parameters that rule electrospinning process are the molecular weight of the material used, types of collector and of solvent used, and ambient parameters [18, 23].

4. Electrospinning of GOs with polymers

The establishment of GO incorporation into polymeric electrospun nanofibres was only initiated around this decade. Since 2010, at least 34 literature works were published involving about 14 types of synthetic and natural polymers. There are some distinct reasons to add GO and its derivatives into electrospun nanofibres. The major purpose is to improve the mechanical and electrical properties of the electrospun material. Others include the morphological enhancement to meet certain diameters or porosity of the spun fibres. **Table 2** summarizes the reported polymer/GO combinations.

Graphene oxide	Polymer	References
Graphene oxide	Poly (vinyl acetate) (PVAc)	[56]
Graphene oxide	Polycaprolactone (PCL)	[42, 56]
Functionalized graphene oxide	Poly (lactic acid) (PLA)	[26, 41]
Graphene oxide	poly(lactic-co-glycolic acid) (PLGA) and collagen (Col)	[40]
Graphene oxide	poly(lactic-co-glycolic acid) (PLGA)	[32, 40]
Graphene oxide	PLA and polyurethane (PU)	[38]
Graphene oxide	Polyurethane (PU)	[38, 53, 55]
Graphene oxide	Polyethylene oxide (PEO)/chitosan (CS)	[44]
Graphene oxide	Poly (vinyl alcohol) (PVA)	[31, 35, 44, 48]
Reduced graphene oxide	Poly (vinyl alcohol) (PVA)	[24, 27]
Graphene oxide	PVA and chitosan CS	[33, 47]
Graphene oxide	Nylon 6-6	[52]
Reduced graphene oxide	Nylon-6	[57]
Graphene oxide	Nylon-6	[34]
Reduced graphene oxide	Poly(carbonate urethane) (PCU)	[30]
Reduced graphene oxide	polyvinyl pyrrolidone (PVP)	[25]
Reduced graphene oxide	polyacrylonitrile (PAN)	[25, 29, 36]
Functionalized graphene oxide	polyacrylonitrile (PAN)	[28]
Graphene oxide	Polyacrylonitrile (PAN)	[28, 50]
Reduced graphene oxide	Polyvinyl Butyral (PVB)	[46]
Graphene oxide	Polyacrylic acid (PAA)	[54]

Table 2. Reported literatures on electrospun GO.

A fundamental study by Park et al. in 2013 described the effect of GO distribution and its location in 1D polymer composites. By controlling the distribution and location of the embedded nanomaterials in 1D nanoscale composites, the macroscopic properties can be enhanced and thus enable many practical applications. In their study, three types of polymers and two different solvents were used. The type of polymers and solvents are two processing conditions to manipulate the distribution and location of the GOs. Homogeneous colloidal dispersions of GO in the polymer solutions were successfully electrospun into polymer nanofibres, without the formation of beads or aggregates [24]. The location of GO was influenced by phase separation during the electrospinning process, which was in turn dependent on the compatibility between the two components and the vapour pressure of the solvents. The thermal and mechanical properties of polymeric composite nanofibres were effectively improved by adding GO as nanofiller.

GOs have a curved morphological structure and form powder materials after exfoliation from graphite oxides. This powder structure can be used in paints, adhesives, coatings and lubricants, where GOs act as multifunctional additives. However, for applications where structured thickness is a limitation, the levelling of the graphene sheets in a plane is required. Thus, an effort to study the levelling of graphene sheets through electrospinning was recently carried out [25]. Those authors used PAN and PVP as examples of polar polymers for nanofibre matrices. GO was reduced using different agents, but later it was confirmed that the curved features of GO sheets cannot be prevented using arGO coating strategy. Nonetheless, the strategy of using electrospinning to extend single GO sheets seems effective. Levelled graphene sheets were successfully developed and showed good conductivity in the order of 10^3 Sm^{-1}.

The effect of GO loading into PCL was investigated in presence of different oxidation levels [26]. GO dispersibility, wettability, polarity and charge relaxation time, which assess the fibre diameter, increase with oxidation level. Surface tension of PCL solution is invariable in the presence of GO, resulted in diameter variations of the spun fibres. The diameter of PCL nanofibres decreases with GO loading, due to the reduction of solution viscosity and increase in the conductivity. Another morphological study attempted to blend rGO into PVA [27]. The addition of a nanofiller seems to increase the viscosity and conductivity, but the nanofibre exhibited smaller diameters than those without nanofillers. Some GO particles were also observed to be protruded from the composite nanofibres under transmission electron microscope (TEM).

The manufacturing of commercial high-performance carbon nanofibre (CNF) has been carried out using PAN as precursor, due to its high carbon yield and superior fibre strength. It includes a typical process of oxidative stabilization and carbonization steps. Nevertheless, most continuous CNFs produced to date have relatively poor graphitic structure, which is important for higher elastic moduli and thermal and electrical conductivities. This can be solved by increasing the carbonization temperature, because high temperature graphitization significantly improves the size and alignment of the graphitic crystallites in PAN-based CNFs. This high-temperature condition is, however, unlikely since fibre processing at such conditions requires a specialized expensive equipment. Therefore, Papkov and co-

workers, in 2013, carefully made an effort to maximize the orientation of the polymer chains inside the polymer precursor fibre by incorporating oriented inclusions that had high surface area and could strongly interact with polymer chains [28]. GO was used since it can both organize the polymer chains and simultaneously serve as a 'templating' agent for the formation of graphitic crystallites during carbonization. Their close examination revealed the presence of crumpled GO particles inside the PAN matrix. The irregular shape of the radially crumpled sheets closely matches the popular model of the high-strength carbon fibre structure, which consists of irregular, longitudinally wrinkled turbostratic graphitic stacks with preferred axial orientation. It is very useful to represent a natural template to achieve similar high-strength CNFs structure in economic manufacturing protocol. In addition, the crumpled graphene oxide particles may provide multiple nucleation sites for graphitic crystal growth during carbonization, especially because the carbon atoms along the crumpled folds are more reactive.

The paramount role of graphene-based embedded in polymer matrix via electrospinning is to enhance the mechanical properties of the polymeric nanofibres. Wang et al., in 2015, examined PAN/GO composite nanofibres with lateral force microscopy and force-distance curves, in order to assess the surface properties like friction force and elasticity. The experiment results indicated that, with increasing GO concentrations, both the surface friction force and adhesive force increased [29]. A mechanical characterization of high-performance GO incorporated into an aligned fibro-porous PCU membrane was made, under static and dynamic conditions [30]. The electrospun membrane indicated that 55% of the tensile strength increased, a 127% rise in toughness, and the achievement of maximum strength reinforcement efficiency was reported at 1.5 wt.% GO loading. An even smaller loading of GO (0.02%) in PVA diluted aqueous solution was able to produce elevation of tensile strength of electrospun nanofibres by 42 times [31].

Another study on significant improvements in thermomechanical and surface chemical properties of poly(D, L-lactic-co-glycolic acid) (PLGA) upon addition of GO nanosheets was published in 2011 [32]. A significant enhancement of storage moduli and glass transition temperature of the PLGA/GO (2 wt.%) was presumably reported to be caused by the enhanced chemical bonding between the GO oxygenated functional groups and the polymer hydroxyl groups. This nanocomposite fibre was more hydrophilic than the pristine PLGA fibre. Biocompatibility tests showed that the enhanced surface chemical properties of the PLGA/GO (2 wt.%) effectively improved neuronal cell proliferation and viability, indicating enhanced biocompatibility of PLGA nanocomposite nanofibres by the addition of 2-D GO nanofillers. Novel electrospun PVA embedded with glycine-modfied GO (GMGO/PVA) and PVA with nanogold deposited GO (AuG/PVA) was fabricated and its biocompatibility was critically analysed [33]. GMGO was synthesized by reducing GO with a biocompatible amino acid, glycine, for conductivity improvement. Then, AuG was modified from GMGO, by mixing with hydrogen tetrachloroaurate ($HAuCl_4$). Cellular toxicity tests of these nanofibres showed almost no cytotoxicity to L929 cells, most likely due to nontoxicity of PVA nanofibres and the biocompatibility of modified GO.

There were also some experiments carried out with produce electrospun GO/polymer nanofibres for morphological modification, in order to meet certain requirements for applications. A spider-wave-like nano-net was fabricated from GO blend with nylon-6 which holds polyelectrolytic and polymorphic characteristics [34]. The amide (CO-NH) group of nylon-6 molecule can easily interact with GO by means of hydrogen bond formation or donor-acceptor complexes. The strategy was to incorporate foreign materials into nylon-6 in order to change the morphology and crystallization of nanofibres. The pristine nylon-6 electro-spun mat appears well defined with no spider-wave-like nano-net. As GO was added into the polymeric solution, a double-layered morphology (bimodal fibre diameter distributed mat) was noticeable. Further increase of GO in the polymeric solution not only decreased the spider-wave-like structure but also decreased the spinnability and formed a film-like structure.

The widespread use of electrospinning technique in tissue engineering application is no longer new. Many researchers have been exploring this subject. GO/polymer composites also found their way through this area, since they showed potential biocompatibility and noncytotoxici-ty characteristics. Scaffolds play an important role in tissue engineering as they not only provide support for cell attachment, but they also allow cell growth into tissue until the tissue is able to sustain itself. There are several types of GO/polymer composites been developed for tissue engineering scaffolds, such as PVA/GO [35], PAN/rGO [36], PVA/CS/GO [37], PLA/PU/GO [38], PLA/hydroxyapatite (HA)/GO [39], GO/PLGA/Col [40], GO-g-PEG/PLA [41] and PCL/GO [42]. The polymeric scaffolds attain better thermal, mechanical and electrical properties, due to the intrinsic properties of GO. Cells adhesion and proliferation onto these nanofibrous scaffolds have been tested morphologically and quantitatively. Cells like mouse osteoblastic cells (MC3T3-E1), adipose-derived stem cells (ADSCs), skeletal myoblasts (C2C12), mouse marrow mesenchymal stem cells (MSCs) and low-differentiated rat pheo-chromocytoma cells (PC12-L) were cultured onto the samples. They confirmed that a small amount of GO did not restrain the proliferation and viability of cells, which demonstrated the appreciable cell affinity of GO. Cells are likely to spread on the composite scaffolds and some even enhance the cell growth by having similar rate of cell proliferation to that of tissue culture plates.

Tissue engineering is very important for regeneration and replacement of damaged tissues in the human body. Scaffolds for musculoskeletal and skin tissues have caught the attention of researchers in the last years, aiming to help many patients with injuries. On the other hand, tissue engineering for organs or tissues without regenerative abilities is an extensive area that needs serious exploration. Vascular tissue engineering is one of its branches, and it aims to fabricate functional vascular grafts to be used in vivo, in order to replace blood vessels and help their regeneration. Current commercial synthetic grafts used by surgeons are woven poly (ethylene terephthalate) (Dacron) and extended polytetrafluoroethylene (ePTFE). Although the aim was to substitute large diameter blood vessels, they are not suitable for small diameter grafts due to thrombosis and intimal hyperplasia risks. A thermoplastic polyur-ethane (TPU)/GO scaffold was fabricated using grounded rotation mandrel as electrospin-ning collector to obtain fibrous small diameter tubular scaffolds [43]. The tensile strength

(Young's modulus) and scaffolds hydrophilicity increased as GO amount increased. The suture retention strength and burst pressure of tubular TPU/GO scaffolds containing 0.5 wt. %. GO exceeded the mechanical requirements of human blood vessels. In addition, results of platelet adhesion tests indicated that a low loading of GO is unlikely to cause thrombosis on the scaffolds. Endothelial cells could form a layer on the inner surface of the tubular scaffolds which resembles the native blood vessel structure, indicating that the TPU/GO electrospun scaffolds, with low GO loading level, have the potential to be used in small diameter vascular graft applications.

Another impressive function of scaffolds that needs to be expanded is the ability to **control drug release** onto specific tissues or organs. A targeted drug delivery system that can efficiently transport drugs is a way for a scaffold to improve the efficacy of the drug and to reduce the systemic toxicity. Electrospun nanofibres are very important materials due to the high surface area and high porosity of the nanofibres, which are able to provide higher drug encapsulation efficiency and better stability, compared to other systems. An electrospun scaffold for anti-cancer drug delivery was tested using a polyethylene oxide (PEO)/chitosan (CS)/GO nanocomposite [44]. In the nanofibre carrier system, GOs act as nanocarriers which control doxorubicin (DOX) release, an anthracycline antibiotic used in cancer chemotherapy intravenous administration. DOX and GO surface can make a strong bond through π–π stacking interactions as a non-covalent type of functionalization that may provide controlled release of drug. GO also contains epoxy and carboxylic functional groups that can react with the amine groups of CS. The π–π stacking interaction between DOX and GO with the fine pores of nanofibrous scaffolds allowed high drug loadings (98%).A strong pH dependence was shown by the DOX release result at pH 5.3 and 7.4. At pH 5.3, faster drug release can be seen which may resulted from the unstability of hydrogen-bonding interaction between GO and DOX.

Along with the development of tissue engineering field, the **fabrication of scaffolds** is also advancing and succeeding in providing a mimic environment to the real body system. Currently, combining biomaterial with living cells for tissue scaffolds is a critical approach. Luo et al., in 2015, have reported a fabrication of GO-doped ply (lactic-co-glycolic acid) (PLGA) nanofibrous scaffold mixed with human MSCs for osteogenic differentiation [45]. The porous structure and the diameter of produced fibres are similar to the topological structure of natural extracellular matrices (ECM), and therefore facilitating cell attachment and proliferation. GO plays two roles in this scaffold. First, it enhances the hydrophilic performance and protein- and inducer-adsorption ability of the nanofibres. Second, it accelerates the human MSCs adhesion, proliferation and differentiation towards osteoblasts.

Graphene-based materials have extraordinary electronic transport properties, strong mechanical strength, excellent electrocatalytic activities, high surface area, ease of functionalization, and thus have contributed to ion detector and **biosensing** applications. A sensor for Cu (II) detector based on rGO/polyvinyl butyral (PVB) nanofibres was fabricated by Ding and co-workers in 2015. Electrospun GO/PVB on glassy carbon electrode (GCE) was reduced through an electrochemical method, to form reduced graphene oxide (RGO)/PVB nanofibres [46]. Cu (II) is one of the heavy metals that contaminate water sources through casual discharge from mining, machinery manufacturing and metal smelting. Although copper is an essential

element in human body, the excessive intake of copper might results in Wilson's disease and Menke's syndrome [46]. The rGO/PVB nanofibres modified glassy carbon electrode (GCE) were used for Cu (II) detection by differential pulse anodic stripping voltammetry (DPASV) [46]. The as-fabricated sensor based on rGO/PVB nanofibres showed good analytical performance with the linear range of 0.06–22 μM, a low detection limit of 4.1 nM (S/N=3), high sensitivity of 103.51 μA •μM^{-1} cm^{-2}, good selectivity and excellent reproducibility (RSD = 0.49%) [46].

A novel platform for enhanced biosensing of electrospun PVA/CS/GO was also engineered [47]. Authors used glucose oxidase (GOD) as a model enzyme. Nanofibres produced from electrospinning are an ideal biosensing platform because of the large surface area they provide, leading to high enzyme loading capacity and low hindrance for mass transfer. Electrospinning is also a simple, durable and adjustable prove to provide stability and reproducibility of biosensors. GOs can bind enzyme molecules well, through electrostatic interactions, and thus avoiding the leaching of enzymes from modified electrodes. Nanomaterials are needed in nanofibre platform to bind the enzyme, since accessibility of the substrate to enzyme will be inhibited when the enzyme is confined inside the nanofibres. The fabricated biosensing platform displayed optimal oxidation and reduction capabilities towards electro-active species, even when compared to the bare Pt electrode, due to the synergy effects of PVA/chitosan nanofibres and GO nanosheets [47].

Power generation is one of the essential needs of human daily life. The increasing demand for green and rechargeable energy sources to power up many electronic devices urged into a variety of power storage system alternatives. Knowing the superior properties of graphene and high specific surface area of CNFs, a number of attempts have been made to incorporate these materials into the fabrication of electrodes for Li-batteries and supercapacitors. Current freestanding anode for Li-ion batteries of graphite has limited theoretical capacity (372 mA h g^{-1}). Xu et al., in 2014, proposed the use of Si, a nontoxic and abundant element, as the anode, due to its highest ever-known theoretical capacity up to 4200 mA h g^{-1} [48]. However, Si has its limitations, as it can undergo up to 280% volumetric expansion upon lithiation, which leads to electrode pulverization and loss of electrical contacts, resulting in severe capacity fading. The solution to this problem is to convert bulk Si into particles of low dimensions and to uniformly disperse Si in an inactive and conductive matrix, like GO. Co-axial nozzle was used to electrospun Si/CNF/GO anode, in order to avoid agglomeration during the spinning process. The resulting electrode achieved a high rate capacity of 567 mA h g^{-1} at 1 mA h g^{-1} current density. It obtained excellent cyclic performance with a discharge capacity of 872 mA h g^{-1} after 50 cycles and 91% capacity retention. Another approach is the synthesis of core-shell structured nanofibres that consist of rGO/PAN (shell) and ZnO/carrier polymer (core) [49]. The current use of ZnO has actually gained a good theoretical capacity of 978 mA h g^{-1}. Unfortunately, it also carries a huge volume expansion for approximately 228%, which causes damage of mechanical integrity. Considering the same solutions, Shilpa et al., in 2015, encapsulated ZnO nanoparticles in the hollow core of glassy carbon–reduced graphene oxide (C–rGO) where the void spaces should provide a buffer zone, accommodating the volume changes associated with lithiation/delithiation of the metal oxide and prevent attrition of nanoparti-

cles by detachment and fragmentation. The outcome of this study gained capacitance of 815 mA h g^{-1} at a current density of 50 mA g^{-1} with a capacity retention of almost 80% after 100 cycles [49]. A freestanding electrode eliminates the use of current collector, which is an inactive material that only adds to the battery mass and volume.

Another CNF composite in form of fibre mats was prepared by electrospinning blend of GO and PAN in *N,N*-dimethylformamide (DMF) for supercapacitor electrodes [50]. Porous carbon material of CNFs was further improved by graphene-based electrical conductivity, through carbonization, to obtain CNF/graphene composites. GO concentration was the major factor responsible for the morphology and pore structures, where the optimized-specific surface area and conductivity were achieved at 5 wt.%. The electrode material (5 wt.% GO) exhibited enhanced electrochemical performance with a highest specific capacitance of 146.62 F g^{-1}, and highest energy density of 14.83–19.44 W h kg^{-1} in the power density range of 400–20,000 W kg^{-1}. Over the last decades, there was another upcoming power storage system that has been successfully applied in numerous fields. **Latent heat storage** using phase change material (PCM) makes use of the solid–liquid phase change of a material in order to store thermal energy. Fatty acid esters showed potential significant energy storage ability, as organic PCMs, with satisfactory thermal properties, thermal conductivity and thermal reliability. Nevertheless, the use of organic PCMs may undergo energy leakage. The synthesis of PCM with a supporting material is crucial to overcome the leakage problem and improve its thermal conductivity. Ke et al., in 2014, report on the preparation of methyl stearate (MES) fatty acid ester with PAN/GO by electrospinning [51]. GO was selected as the heat transfer/diffusion promoter and also as supporting material in the phase change composite. The results suggest that by adding GO, beneficial effects on mechanical properties, thermal stability and thermal energy storage/release performance were gained.

A work has been published dealing with the use of nylon 6-6 excellent mechanical resistance, with functionalized GO as a **corrosion protection** composite coating [52]. 2% GO loading in the electrospun nylon 6-6 possessed coating capacitance in the order of 10^{-7} F/cm^2, compared to 10^{-4} F/cm^2 for coating prepared by deposit. This value suggests charge storage capacity for the coating condition. Thus, the efficiency of deposited coating is better than electrospun coating. The well-known physicochemical properties of PU have been widely used in coating technology. However, its low hydrophilicity and mechanical properties are the main setbacks for its application. Meanwhile, GO-polymer composite fibres emerged as the subject of enormous interest because polymer nanocomposites, with graphene materials as fillers, have shown dramatic improvements in such needed properties. Electropinning of in situ synthesized PU/GO has been conducted for surface coating of metallic stents [53]. This study showed that the incorporation of small amounts of GO sheets through PU nanofibres could enhance the stability of coated layer on the surface of nonvascular stent. The grafted PU chains on the surface of GO can facilitate the beneficial stress transfer from the PU matrix to the GO sheets. Moreover, proper incorporation of GO sheets into the polymer matrix can improve the hydrophilicity of the materials, without any adverse biological effect. These improved properties of PU mat, induced by GO, make it a potential candidate for surface coating of medical devices.

In order to fabricate **water-swellable rubber** (WSR), a multi-scale hybrid nanofibre with enhanced elasticity, resilience, toughness and water swelling ability was electrospun [54]. WSR can be produced by chemical and physical methods. The chemical method is more complicated and more environmentally unfriendly than the physical method. In the physical method, common hydrophobic rubbers are mixed with various water-absorbent materials, known as superabsorbent polymers (SAPs) (hydrogel form). Hydrophilic super water-absorbent resin does not disperse well in hydrophobic rubber, expecting easy break off of physical-synthesized WSR. Eventually, swelling ability will lose, strength is remarkably decreased, and water is polluted. The addition of hydrophilic fibres can fix then as they can act as internal water channels in hydrophobic materials for superabsorbent nanocomposites. These fibres can help to transfer water from the surface of the rubber matrix to the hydrophilic SAP particles, as well as between encapsulated SAP particles in the rubber matrix, enhancing the water swelling ability of WSR composites. Fabrication of PAA/ethylene glycol (EG)/hyperbranched bis-MPA polyester-64-hydoxyl (HB)/GO showed synergistic effects on water swelling ability compared to without HB and GO. The electrospun fibre mats were added into conventional rubber by simple physical mixing, and the rubber composites showed considerable enhancement of water swelling ability, mainly due to the water channels built.

Functional **shape memory** composite nanofibres with GO have also been fabricated with PU shape memory [55]. Shape memory PU/GO (SMPU/GO) has better shape memory effect (SE) and lower thermal shrinkage, compared to SMPU nanofibre mats. When the loading amount of GO increased to 4 wt.%, the thermal shrinkage ratio of the composite nanofibrous mats reached as low as $4.7 \pm 0.3\%$, while the average fixation ratio and recovery ratio were as high as 92.1 and 96.5%, respectively. The study indicates that GO is a desirable reinforcing filler for preparing shape memory nanofibres with improved properties. Electrospun polymer with GO and modified GO fillers have been extensively investigated for a wide range of applications. Apart from all applications mentioned, electrospinning of polymer/GO also stands out as an optical element in fibre lasers [56] and photocatalysts [57], showing their important role in the area of photonics.

5. Conclusions

Graphene oxide and its derivatives show comparable properties as alternatives to graphene. Their outstanding performance can easily be manipulated into a wide variety of applications. An important highlight is that several researchers have shown that the biocompatibility of produced composites is still preserved, with no significant cytotoxicity appearing at low GO loading. Utilizing electrospinning to fabricate polymer/GO composites is a wise choice to obtain high specific surface area fibres at the nanoscale. It displays a promising advantage in achieving optimum properties out of the electrospun composites. Nonetheless, further research towards commercializing polymer/GO electrospun fibres in applications and improvement on electrospinning machines needs to be done, so that one can make use of their outstanding advantages to a larger extent.

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