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Review

Nanocomposite Nanofibers of Graphene—Fundamentals and Systematic Developments

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Abstract: Research on polymer nanocomposite nanofibers has seen remarkable growth over the past several years. One of the main driving forces for this progress is the increasing applicability of polymer nanocomposite nanofibers for technological applications. This review basically aims to present the current state of manufacturing polymer/graphene nanofiber nanocomposites, using appropriate techniques. Consequently, various conducting and thermoplastic polymers have been processed with graphene nano-reinforcement to fabricate the nanocomposite nanofibers. Moreover, numerous methods have been adopted for the fabrication of polymer/graphene nanocomposites and nanofibers including interfacial polymerization, phase separation, freeze drying, template synthesis, drawing techniques, etc. For the formation of polymer/graphene nanocomposite nanofibers, electrospinning can be preferable due to various advantages such as the need for simple equipment, control over morphology, and superior properties of the obtained material. The techniques such as solution processing, melt spinning, and spin coating have also been used to manufacture nanofibers. Here, the choice of manufacturing techniques and parameters affects the final nanofiber morphology, texture, and properties. The manufactured nanocomposite nanofibers have been examined for exceptional structural, microstructure, thermal, and other physical properties. Moreover, the properties of polymer/graphene nanofiber rely on the graphene content, dispersion, and matrix–nanofiller interactions. The potential of polymer/graphene nanocomposite nanofibers has been investigated for radiation shielding, supercapacitors, membranes, and the biomedical field. Hence, this review explains the literature-driven significance of incorporating graphene in polymeric nanofibers. Conclusively, most of the studies focused on the electrospinning technique to design polymer/graphene nanofibers. Future research in this field may lead to advanced innovations in the design and technical applications of nanocomposite nanofibers. To the best of our knowledge, research reports are available on this topic; however, the stated literature is not in a compiled and updated form. Therefore, field researchers may encounter challenges in achieving future advancements in the area of graphene-based nanocomposite nanofibers without first consulting the recent literature, such as an assembled review, to gain necessary insights, etc. Consequently, this state-of-the-art review explores the manufacturing, properties, and potential of polymer/graphene nanocomposite nanofibers.

Keywords: graphene; polymer; nanofiber; nanocomposite; manufacturing; spinning; shielding; membrane

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1. Introduction

The nanofibers of polymer/nanocarbon nanocomposites have been effectively developed and reported in the literature [1]. Graphene has been considered an important nanocarbon nanomaterial [2,3]. The combination of polymer and graphene has improved the morphological, electrical, thermal, mechanical, and other essential physical properties of the nanocomposites [4]. Improvements in nanocomposite properties have been observed due to the synergistic effect between the polymers and nanofillers [5,6]. Consequently, the polymer/graphene nanocomposite nanofibers were developed. Fine polymer/graphene nanofibers have been manufactured using facile techniques and different polymeric matrices. Electrospinning has been identified as a prominent technique to develop polymer- and nanocomposite-based nanofibers [7,8]. Here, the electrospinning technique revealed various controllable processing parameters. The polymer/graphene nanocomposite nanofibers have been designed and fabricated for radiation shielding, energy, membranes, and biomedical applications [9,10].

This review article comprehends the fundamentals, characteristics, and applications of polymer/graphene nanocomposite nanofibers. The inclusion of graphene nanofiller in polymers and processing using facile manufacturing techniques have resulted in high-performance nanocomposite nanofibers. Enhancement in the physical features has been observed with the increasing nanofiller contents. Applications of the finely manufactured polymer/graphene nanocomposite nanofibers have been observed for supercapacitors, radiation shielding, membranes, and biomedical devices.

Thus, this review is groundbreaking and pioneering to elaborate on the scientific advancements in the field of graphene-based nanofibers. This comprehensive review article states the fundamentals and technical features of polymer/graphene nanocomposite nanofibers. The incorporation of graphene in polymeric matrices using appropriate fiber processing techniques has produced high-performance nanocomposite nanofibers. The characteristics and performance of the nanofibers rely on the compatibility between the matrix–nanofiller, interactions, and consistent nanoparticle dispersal. High-competence nanocomposite nanofibers have been functional in technical fields ranging from energy to biomedical. To the best of our knowledge, this overview is novel and ground-breaking to highlight the field of graphene-derived nanofibers. Research has been reported on polymer/graphene nanofibers; nonetheless, the literature needs to be presented in an updated and compiled form. Hence, these polymer/graphene nanocomposite nanofibers will be helpful for the related scientists for essential future upheavals in graphene-derived nanofiber technologies. This novel article on polymer/graphene nanofibers will definitely be helpful for essential future innovations in nanofiber manufacturing.

2. Polymer and Polymer Nanocomposite Nanofibers

The polymer nanofibers are simply nanostructures with a diameter of a few nanometers and a length of up to several millimeters [11]. Various natural and synthetic polymers have been used to develop the nanofibers. The polymer nanofibers have superior physical properties and technical applications [12]. The polymer nanofiber diameter and characteristics have been dependent on the manufacturing techniques used. Accordingly, polymer nanofibers of different shapes have been manufactured, such as hollow nanofibers, ribbon-like nanofibers, wrinkled nanofibers, smooth nanofibers, and solid filled nanofibers.

Among different types of fibers, solid nanofibers with smooth surfaces have been preferred for various technical uses. In some cases, hollow nanofibers have been preferred according to the desired end application. Synthetic polymers including conducting polymers and thermoplastic polymers have been widely used to form nanofibers [13]. The solid round nanofibers with homogeneous surfaces have a large surface-area-to-volume ratio, optimum porosity, flexibility, toughness, and appreciable mechanical strength for applications such as membrane materials, packaging, and other technical applications. On the other hand, the soft porous and hollow nanofibers of natural polymers have found applications in wound dressing and biomaterials for tissue regeneration. Depending upon the polymer type and nanofiller used, the nanofiber properties such as mechanical properties, tensile strength, toughness, flexibility, electrical properties, optical properties, and chemical properties can be varied.

The polymer nanofibers have been manufactured through a range of techniques including solution drawing, melt extrusion, template technique, template-free process, and spinning techniques [14]. The polymer fibers and nanofibers have been produced through the traditional wet, dry, melt, or gel spinning techniques. The diameter of as-spun nanofibers may vary from nano- to a few micrometers. The traditional dry spinning process involves the use of one or more solvents to dissolve the polymers. This method is termed dry spinning because the solvents are removed through recovery procedures. In the wet spinning process, the fibers are placed in a coagulation bath, and solvent removal or evaporation techniques are not demanded. Due to the involvement of solvent extraction techniques, the dry spinning method has been found costly. The gel spinning technique has also been applied to the polymer fibers. In this method, the polymer gel is used for fiber spinning, and as-sun fibers are usually dried. The effectiveness of these techniques has been analyzed on the basis of surface area, porosity, flexibility, strength, and chemical resistance properties. The fabricated polymer nanofibers have been applied to membrane-based filtration systems, coatings, energy devices, electronics, tissue engineering, and drug delivery [15]. The conducting polymer nanofibers having a diameter of 200 nm and length of ~30 μm have been fabricated by the spinning method [16]. In particular, the polyaniline nanofibers have been processed by the spinning technique [17]. These nanofibers have important applications in electronics, supercapacitors, and sensors. Among thermoplastic polymers, polystyrene, polyethylene, polyamide, poly(vinyl alcohol), poly(ethylene oxide), Nafion, poly(acrylic acid), etc., have been used for the nanofiber manufacturing [18]. The properties of nanofibers have been found to depend on the polymer type and manufacturing method used [19].

Similar to polymers, the polymer nanocomposites have been processed to form nanofibers. Some of the important manufacturing techniques include interfacial polymerization, phase separation, freeze-drying synthesis, template synthesis, drawing method, and spinneret-based tunable engineered parameter (STEP) method.

In the interfacial polymerization of polymer graphene nanocomposites, different monomers are usually dissolved in two different immiscible solvent phases and allowed to react at the interface [20]. Graphene can be introduced in one phase. The monomers are polymerized at the interface of the emulsion [21]. Using interfacial polymerization, the nanofibers can be fabricated due to the homogeneous nucleated growth process. Using this method, different types of polymers and nanocomposites can be produced [22]. In this technique, the concentrations of monomers in the immiscible solvent phases usually affect the molecular weight of the polymer formed and graphene dispersion.

In the phase separation method, the phases usually separate due to physical incompatibility [23]. The solvent phase is extracted in the solution, whereas the other phase remains intact. Another important factor of this method is the polymer dissolution in a solvent at room temperature or elevated temperature. Nanocomposite or nanofiber morphology is usually affected by a controlled gelation step. Here, the polymer concentration and gelation temperature also need to be controlled [24]. However, all polymers do not have a phase separation phenomenon to produce the nanofibers.

Freeze drying is an important method to form polymer/graphene nanocomposites and nanofibers [25]. This technique involves the solid–liquid phase separation and ice segregation-induced self-assembly processes. Initially, the solution is frozen at a low temperature (−70 to −80 °C), allowing ice-crystal growth and nucleation. Then, the drying process and removal of frozen samples through direct sublimation are carried out [26]. Rapid freezing and direct sublimation are important factors to avoid any chemical reactions or side products during the material formation.

The template method usually employs the chemical or electrochemical oxidative polymerization processes [27]. Moreover, the template or mold is used to obtain a desired nanocomposite or nanofiber material. In this technique, the nanofibers are formed by passing polymer solution through the nanopores of the template under water pressure on one side, which causes the extrusion of the polymer and the formation of nanofibers. The fibers solidify upon contact with the solidifying solution [28]. Thus, nanofibers of different diameters can be formed depending on the size of template nanopores. Thus, this technique can be advantageous to control the fiber diameter. However, this method produces nanofibers of only a few micrometers, and cannot produce long nanofibers.

Spinneret-based tunable engineered parameters (STEP) technique has also been used to form the nanocomposite and nanofibers [29]. This technique employs a micropipette spinneret and rotating substrate. The nanofibers of nano- to micrometers in diameter can be formed. Consequently, highly aligned and uniform nanofibers are developed on the substrate [30]. In the STEP methods, numerous factors (polymer type, molecular weight, solvent type, viscosity, etc.) govern the fiber diameter, length, porosity, and defects. Table 1 shows the essential properties and techniques applied for polymer and polymer nanocomposite nanofibers.

Table 1. Techniques, process parameters, and properties of polymer and polymer nanocomposite nanofibers.

Methods	Electrospinning	Solution Blowing	Template Synthesis	Phase Inversion	Freeze Drying	STEP Techniques
Nanofiber diameter range	40 nm to 2 µm	40 nm to several µm	40 nm to a few hundred nanometers	50 nm to 1 µm	50 nm–1 µm	50 nm to several µm
Production rate (injection rate)	5 µL/min	20 µL/min	-	-	-	15–100 µL/min
Influencing parameters	Voltages, viscosity, solution feeding rate, distance	Nozzle geometry, viscosity, solution feeding rate, gas pressure	Template shape, template pore size	Solvent properties, polymer concentration	Freezing rate, solvent features, polymer concentration	Polymer type, molecular weight, solvent properties, substrate, speed
Voltage requirement	10–40 kV	NO	~30 V (for electropolymerization)	No	No	No
Industrialization	Yes	Yes	No	No	No	No
Possibility of producing aligned nanofibers	Yes	Yes	Yes	Yes	Yes	Yes
Possibility for melt spinning	Yes	Yes	Yes	No	No	No
Possibility for spinning from highly concentrated polymer solutions	Yes	Yes	No	No	No	Yes
Production of core/shell nanofibers	Yes	Yes	Hollow	Yes	Yes	No
Production of polymer/composite fiber	Polyamide; polystyrene; polyaniline; polyamide/carbon nanotube; polystyrene/carbon nanotube; polyaniline/single-walled carbon nanotube nanofibers; polyaniline/zinc oxide; the nylon 6,6/zinc oxide; polyaniline/titania	Poly(vinyl alcohol); poly (lactic acid); poly(vinyl alcohol)/zinc oxide; poly(lactic acid)/titanium dioxide	Polypyrrole; polyaniline; polypyrrole/silica;	Poly(vinyl fluoride); aramid; poly(vinyl fluoride)/bentonite; aramid/zeolite	Poly(vinyl alcohol); poly(vinyl alcohol)/cellulose;	Polymer/titania; polymer/titania/alumina; hydroxyapatite/chitosan
Refs.	[31–36]	[37,38]	[39,40]	[41,42]	[43–45]	[46–48]

Among the non-spinning methods, plasma-induced synthesis has also been used to form nanofibers [49]. The process consists of significant steps such as atomic vapor deposition, expansion of plasma, solution condensation, in situ oxygen reactions, and nanofiber growth. This technique may produce nanofibers of around 15–25 nm in diameter [50]. However, this is a sophisticated technique to form nanofibers and therefore less preferred.

As compared to plasma-induced synthesis, electrospinning has been more frequently used due to facile processing advantages.

Thus, for the nanocomposite nanofibers, various carbon and inorganic nanoparticles have been used [51]. Carbon nanotube-reinforced polymeric nanofibers have been developed [52]. Fajardo-Diaz et al. [31] reported the nanofibers of polyamide/carbon nanotube using a spinning process for reverse osmosis membranes. Amer Flayeh et al. [32] fabricated the polystyrene/carbon nanotube nanocomposite nanofibers using the spinning method. The nanofibers have been studied for fiber texture and morphology. Liao et al. [33] manufactured polyaniline/single-walled carbon nanotube nanofibers. The nanofibers revealed enhancement in the electrical conductivity from 10^{-4} S cm^{-1} to 10^{2} S cm^{-1}, with increasing nanofiller contents. At high nanofiller contents, the nanofibers revealed a non-homogeneous surface and beaded texture [53]. Inorganic nanoparticles have also been used as an effective reinforcement in polymeric nanofibers [54]. In poly(vinyl alcohol) and cellulose matrices, silica nanoparticles and clay nanoplatelets have been reinforced to form nanocomposite nanofibers [55]. Titania nanoparticles have also been filled in the polymer nanofibers using the sol–gel and spinning methods [56]. The template method has been used for manufacturing polyaniline/titania nanocomposite nanofibers [36]. Patil et al. [34] fabricated the polyaniline/zinc oxide nanofibers through spinning. These nanofibers have uniform surfaces and morphology. The nanofiber diameter was ~200–300 nm. Kayaci et al. [35] manufactured the nylon 6,6/zinc oxide nanofibers through the spinning and atomic layer deposition techniques. The core–shell nanofibers have uniform thickness. Thus, the diameter, properties, and uniformity of the polymer nanocomposite nanofibers rely on the choice of polymer, nanofiller selection, and the fabrication method used.

The choice of manufacturing technique and related parameters definitely affect the final nanofiber material properties:

i. In the electrospinning technique, the feed rate can influence the polymer solution delivery speed and jet intensity [57]. Consequently, the feed rate has been found to affect the diameter and morphology of the nanofibers. Increasing the polymer solution feed rate may enhance the fiber diameter, whereas slow flow may form thin fibers.
ii. Distance between the spinneret tip and collector has been found to affect the morphology and fiber diameter [58]. The optimum distance between the tip and collector provides sufficient time to dry the nanofibers and avoid bead formation. Furthermore, the increasing distance results in more round solid fibers.
iii. The sufficiently high voltage between a needle and metal collector is important to overcome the surface tension holding a drop of liquid at the needle tip [59]. Consequently, a thin fluid jet is projected out. The solvent is easily evaporated during the trajectory between the needle and collector. Hence, fibers with homogeneous surfaces and small diameters have been developed. The low electric field cannot provide enough jet elongation to generate uniform fibers.
iv. The pumping pressure also influences the flow of polymer solution during the electrospinning process [60]. Very low pumping pressure may increase the nanofiber diameter and bead formation.

The technologies for producing nanofibers have been in progress, since the 20th century [61]. The developments in the 21st century (since the 2000s) have made fiber processes more commercially feasible. Recent developments must focus on enhancing the current manufacturing technologies to produce nanofibers with high product uniformity and process speed. Consequently, a large number of contract manufacturers can trade successfully. For efficient trading and commercial viability of nanofibers for technical applications, fibers having fine diameters must be efficiently produced and employed.

Other spinning-based processing techniques have also been seen in the literature to form nanofibers. The solution blow spinning technique has been adopted to form nanofibers [62,63]. This technique compensates for the essentials of both the electrospinning and melt-blowing methods [64]. This technique may be helpful to form the non-woven micro- and nanofibers. The nanofibers of poly(methyl methacrylate), poly(lactic acid), and poly(vinyl alcohol) have been effectively formed using solution blowing [65]. The choice of polymer and concentration has been found to influence the fiber diameter [66]. The solution blow spinning setup has a syringe pump, concentric nozzle, high-velocity gas flow, and collector [67]. The nanofiber size and diameter have been found comparable to the electrospinning method. Moreover, the fiber production rate was high. Still, electrospinning has been widely used for nanofiber formation.

The centrifugal jet spinning method has been effectively used to form micro- or nanofibers [68]. It is an advantageous method due to the low cost, efficiency, and high throughput to form the fibers. Moreover, this technique has the advantages of the precise handling of centrifugal forces, viscoelastic properties, and mass transfer features of the desired solution used for the nanofibers [69]. The centrifugal jet spinning setup contains a DC motor, a spinning chamber, and multiple fiber collectors [70]. For solution-processed nanofibers, centrifugal jet spinning has been found to be as beneficial as the electrospinning technique [71].

Electrohydrodynamic direct writing (with a mechano-electrospinning process) has been successfully applied to form the micro- or nanofibers [72]. This technique involves the use of a mechano-electrospinning process for the constant/programmable direct writing of the fibers [73]. The method has combined the electrical and mechanical forces to form viscous ink for the large-scale production of the fibers [74]. The main fabrication steps contain the alteration of the electrical field and mechanical drawing force to control the nanofiber size and morphology, changeable nozzle-to-substrate distance, and adjustable applied voltage [75]. However, electrospinning has been more frequently used due to the simple equipment and easily controllable processing conditions for nanofibers.

3. Graphene

Graphene is a two-dimensional nanostructure consisting of sp^2 hybridized carbon atoms [76]. In 2004, Andre Geim and Konstantin Novoselov prepared and reported single-layer graphene [77]. However, theoretically, graphene was reported earlier in 1947 (P. R. Wallace) and then experimentally explored in 1962 [78]. Graphene has been synthesized using various techniques such as mechanical cleavage of graphite, exfoliation of graphite, plasma and laser techniques, chemical vapor deposition, and chemical routes [79]. Graphene revealed excellent structural and physical properties. Graphene is a very thin material and is one atom thick [80]. Graphene has a high electron mobility of ~200,000 $cm^2\ V^{-1}s^{-1}$. In addition, graphene has a high thermal conductivity of ~3000–5000 W/mK [81]. Young's modulus of graphene has been found ~1 TPa, which means that it is 200 times stronger than steel [82]. Due to van der Waals forces, graphene nanosheets may possess wrinkling features [83]. To avoid crumpling problems, graphene has been functionalized through various facile techniques [84].

Graphene has been oxidized to introduce the carbonyl, epoxide, hydroxyl, and carboxylic acid groups on the surface. The oxidized form of graphene is commonly referred to as graphene oxide. Among modified forms of graphene, graphene oxide has a unique structure with hydrophilic functional groups [84]. For synthesis, Brodie's method has used a mixture of potassium chlorate and nitric acid to form graphene oxide from graphite [85]. Hummers and Offeman methods have been used to form graphene oxide from graphite using sodium nitrate, sulfuric acid, and potassium permanganate [86,87]. Both graphene and graphene oxide have been used as nanofillers in polymeric nanocomposites and nanocomposite nanofibers for high-performance technical applications. Figure 1 depicts the structures of graphene and graphene oxide nanosheets.

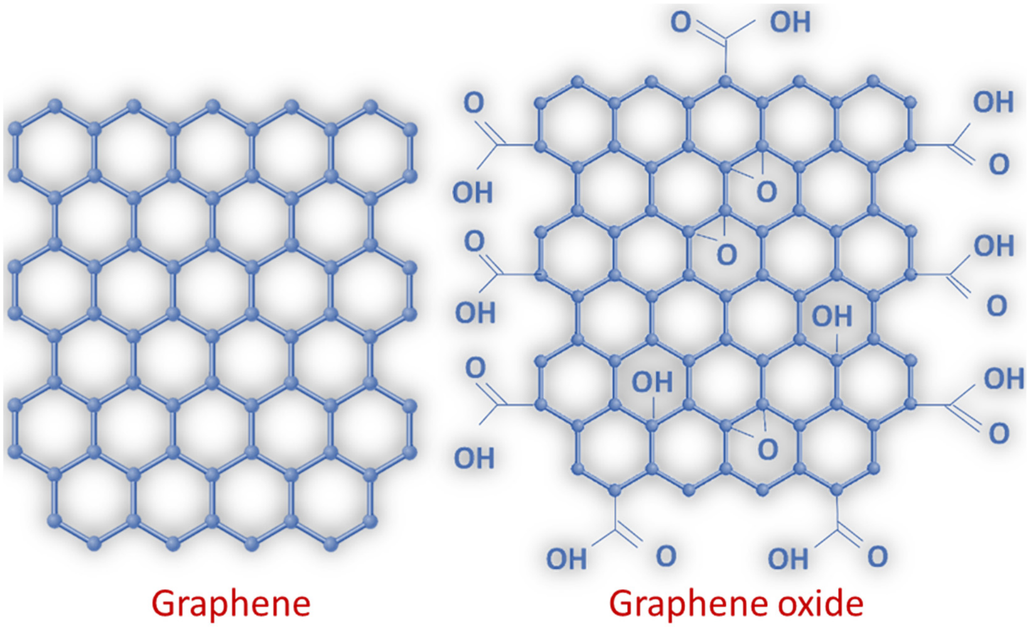

Figure 1. Graphene and graphene oxide.

Graphene and graphene oxide reveal superior heat stability, electrical conductivity, thermal conductivity, mechanical strength, and chemical stability properties [88]. These features of graphene have been explored for the development of high-performance nanocomposites [89,90]. Graphene and modified graphene nanomaterials have found applications in aerospace/automotive [91], energy devices [92], electronics [93,94], sensors [95], membranes [96], and several other technical fields. Table 2 shows important graphene-based nanocomposite nanofiber systems prepared using varying techniques and related physicochemical properties and parameters.

Table 2. Properties and fabrication techniques for polymer/graphene nanocomposite nanofibers.

Polymer/ Nanocomposite	Technique	Diameter/ Size	Solvent/ Concentration	Flow Rate	Voltage Requirement	Physical Properties	Ref.
Polyaniline/poly (methyl methacrylate)/amino-functionalized graphene.	Electrospinning	35–133 nm	Dimethyl formamide	0.3 mL/h	18–20 kV	Thermal stability	[97]
Poly(ε-caprolactone)/ graphene oxide	Electrospinning	201–264 nm	Glacial acetic acid; 1.5 *w*/*v*%	1 mL/h	12 kV	Increment in tensile stress by 189%	[98]
Poly(ε-caprolactone)/ graphene	Electrospinning	121–154 nm	Dichloromethane/methanol; 10–12 wt.%	0.8–1 mL/h	15–17 kV	Young's modulus tensile strength of 3771 MPa and 56.08 MPa, respectively	[99]
Poly(ε-caprolactone)/ reduced graphene oxide	Electrospinning	100–130 nm	Glacial acetic acid; 1.5 *w*/*v*%	1 mL/h	12 kV	Tensile strength increase by 304 %	[100]
Polyamide/graphene	Electrospinning	76–338 nm	Hexafluoroisopropanol; 0.005–0.01 wt.%	0.05 mL/h	8–10 kV	Increase in tensile strength, Young's modulus fracture, toughness by 56%, 113%, and 250%, respectively	[101]
Polyethylene/graphene	Drawing	-	Xylene; 0.1 wt.%	Draw ratio 30–70	-	Thermal conductivity ≈ 75 W m^{-1} K^{-1} ρ^{-1}	[102]
Polypyrrole/graphene	Template method	80–100 nm	NaOH and acidic solutions	-	-	Specific capacitance 466 Fg^{-1}; energy density 165.7 Wh/Kg	[103]
Polypyrrole/graphene	Wet spinning method	40 µm	Acidic solution; ethanol: water; ~10 mg/ml	-	-	Tensile strength 364.3 MPa; specific capacitance 334 mF cm^{-2}	[104]
Aramid/graphene	Co-axial spinning	~8 nm	Dimethyl sulfoxide	2, 4, 6 mL/h	-	Increase in ultimate tensile stress by 700%	[105]
Polyethylene terephthalate/graphene	Dry-jet wet spinning	-	Methanol; 3 mg/mL	Air gap ~3 cm; pressure 25.0 psi	-	Percolation threshold 0.2 S/cm	[106]

4. Manufacturing and Properties of Polymer/Graphene Nanocomposite Nanofibers

The polymer/graphene nanocomposite nanofibers have been manufactured through various facile procedures [107]. The commonly adopted techniques include wet spinning [108], melt spinning [109], electrostatic spinning [110], and other solution and chemical methods. Spinning methods have been considered advantageous due to less graphene agglomeration in the resulting nanofibers [111]. Additionally, the spinning processes have numerous controllable fabrication parameters and facile processing [112]. Among spinning methods, electrospinning has been widely used to manufacture nanocomposite nanofibers [113,114]. Accordingly, electrospinning has been effectively applied to form polymer/graphene nanocomposite nanofibers [115]. These nanofibers have been investigated for superior conducting, mechanical, thermal, and other enhanced physio-chemical characteristics [116]. Consequently, polymer/graphene nanocomposite nanofibers have been investigated for industrial-level manufacturing and uses. The electrospun graphene nanofibers have been found promising for advanced sensor technologies [117–119]. Electrospinning is considered a simple, inexpensive, and multipurpose technique to manufacture nanofibers having diameters down to nanometers [120]. Figure 2 shows a simple electrospinning setup.

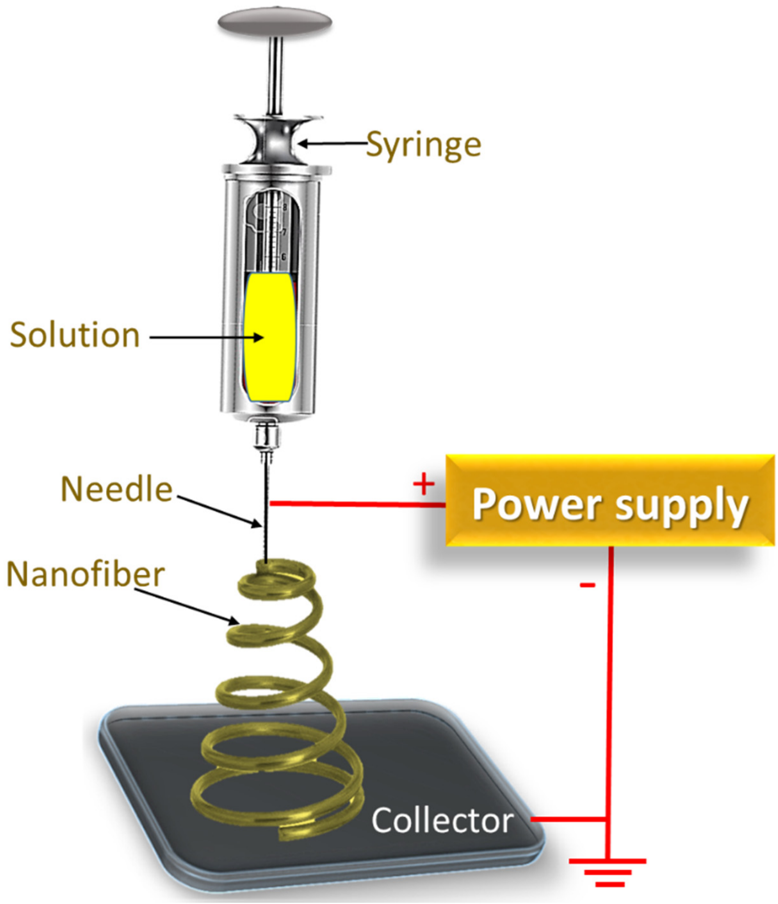

Figure 2. Electrospinning setup.

In this technique, electric force has been used to draw charged filaments of polymer solution or polymer melt [121]. Usually, a characteristic electrospinning setup has three gears, including (i) a syringe with a pumping system; (ii) a high-voltage power supply for charging the polymer solution; and (iii) a grounded collector to collect the nanofibers. During this process, the syringe was used to pump the polymer solution at a constant rate. Consequently, the polymer solution was extracted from a syringe needle. Under applied voltage, the polymer solution or melt gets charged and stretched to form the fibers due to electrostatic repulsions and surface tension [122]. The polymer jet is usually elongated through the whipping process. Consequently, the charged polymer jet travels to the grounded collector, and nanofibers get deposited. The thin nanofibers having uniform texture and nanometer-scale diameter have been developed using this technique [123]. Two types of electrospinning processes have been seen in the literature including the vertical and horizontal electrospinning systems. The difference in vertical and horizontal electrospinning systems caused changes in the fiber orientation, fiber surface texture, and fiber morphology. Typically, horizontal electrospinning has resulted in random fiber orientation. On the other hand, vertical electrospinning revealed uniform fiber alignment and smooth texture. Moreover, the vertical electrospinning system has facile parameter optimization and process monitoring. The resulting nanofibers have uniform surfaces and less beaded texture. This set up has been used for lab-scale nanofiber processing. For industrial-level electrospinning, several design challenges need to be overcome. Melt electrospinning has also been used for polymer and nanocomposite nanofiber formation. Similar to solution-based electrospinning, the melt spinning setup consists of a needle, syringe, heating system, tubular collector, high-voltage power supply, and polymer for melt formation. Most fiber processing conditions of melt and solution electrospinning have been found similar. In the case of high molecular weight polymers with less solution solubility, melt spinning has been found useful. However, this technique may result in nanofibers of large diameter. There are some differences between the electrospinning techniques for the polymer melts and polymer solution processing. For example, in the case of melt electrospinning, a continuous heat supply has been required to keep the polymer in molten form for fiber formation. Therefore, this technique uses more power and cost. For efficient melt-based fiber processing, the distance between the needle tip and collector must be reduced. Similar to polymer, the nanocomposite solution/melt can be electrospun to form the nanofibers. The resulting nanocomposite nanofibers have high surface area, controllable surface configuration, porosity, and uniform texture [124–126].

One-dimensional nanostructures of polyaniline (a conducting polymer) have been designed with varying shapes and unique characteristics [127–129]. These nanostructures include polyaniline nanotubes, nanowhisker, and nanofiber forms. Polyaniline nanostructures are usually manufactured through a variety of procedures including chemical, solution, and spinning processing [130,131]. Graphene-filled polyaniline nanofibers have been fabricated and studied for electrical conductivity and other physical properties [132,133]. Zhou et al. [134] manufactured the pristine polyaniline and polyaniline-wrapped graphene nanofibers through wrapping and electrospinning techniques. After the electrospinning of polyaniline, graphene-wrapped polyaniline nanofibers were formed through the reduction and accumulation of graphene oxide nanosheets on the nanofiber surface (Figure 3) [40,135,136]. A transmission electron microscopy (TEM) image of graphene-wrapped polyaniline nanofiber depicted uniform graphene coating on the surface (Figure 4). Figure 5 demonstrates the specific capacitance of the polyaniline and graphene-wrapped polyaniline nanofibers. The specific capacitance of nanocomposite nanofibers was found to be considerably higher (250 Fg^{-1}) than the neat polyaniline nanofibers (~175 Fg^{-1}). The results indicated uniform nanofiller dispersion and network formation to promote the charge transfer and capacitance of the nanocomposite nanofibers. The capacitance was also found higher than the reported non-electrospun polymer/graphene nanocomposite electrodes [137].

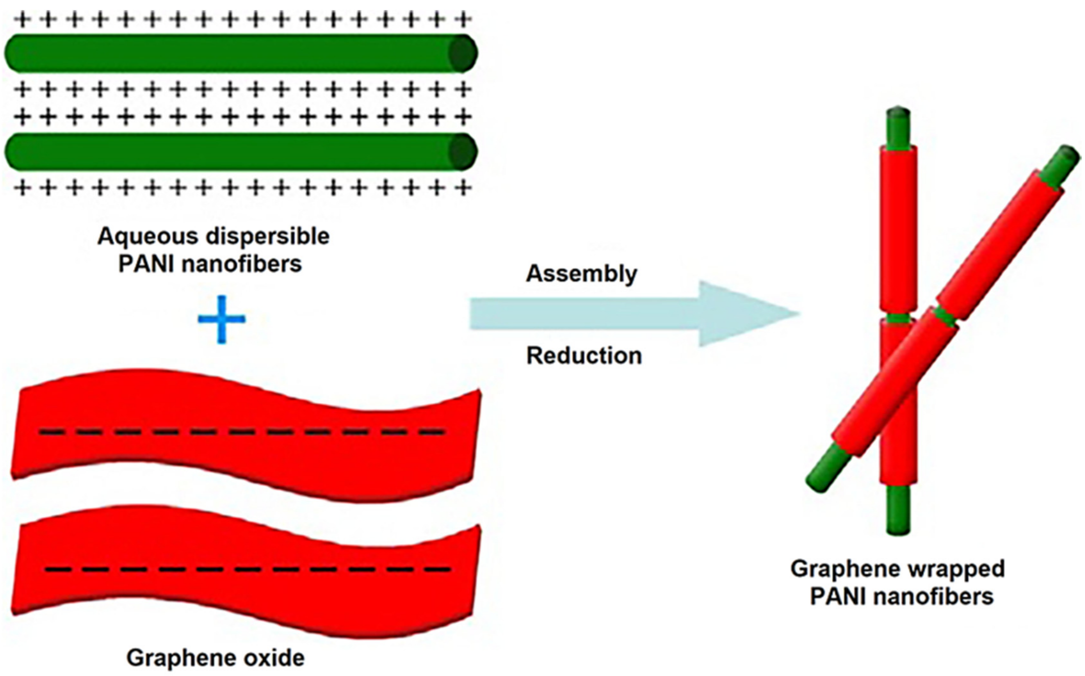

Figure 3. Formation of graphene-wrapped polyaniline (PANI) nanofiber [134]. Reproduced with permission from Elsevier.

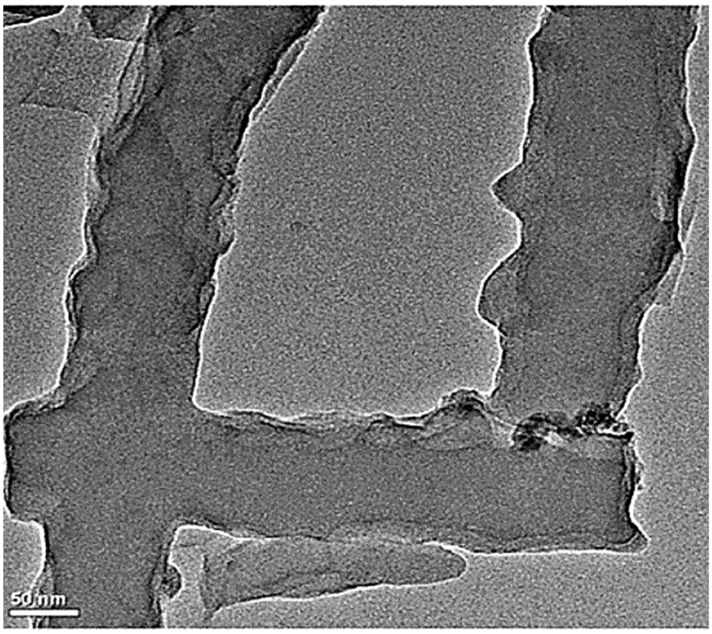

Figure 4. Transmission electron microscopy (TEM) image of graphene-wrapped polyaniline nanofibers [134]. Reproduced with permission from Elsevier.

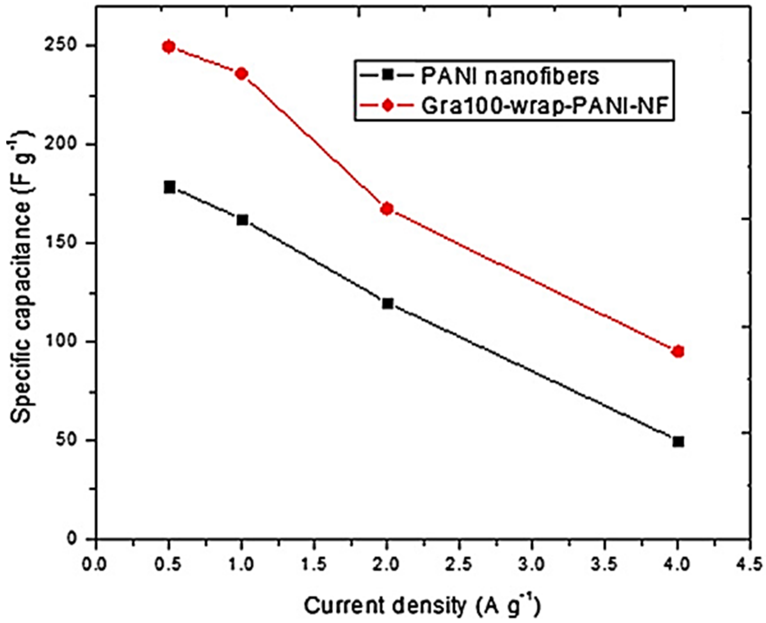

Figure 5. Specific capacitance of polyaniline (PANI) and graphene-wrapped polyaniline nanofibers (Gra100-wrap-PANI-NF) vs. varying current densities [134]. Reproduced with permission from Elsevier.

Additionally, the polyaniline/graphene nanocomposite nanofibers have been fabricated through the in situ polymerization of aniline on the graphene surface in acidic conditions [138,139]. In this method, the sodium dodecyl benzenesulfonate surfactant has been used for better graphene dispersion in the polyaniline nanofibers [140–142]. Then, the template method was also used to form the polyaniline/graphene nanocomposite nanofibers having high electrical conductivity and specific capacitance (>400 Fg^{-1}) [143].

High-performance nylon/graphene nanocomposites have been reported in the literature [144–146]. The nanofibers of nylon/graphene nanocomposites have been developed. Lee et al. [147] manufactured the solution-blown nylon 6 and graphene flakes-derived nanofibers. Maccaferri et al. [148] fabricated the nylon 6,6/graphene nanocomposite nanofibers by electrospinning technique. The effect of optimum electrospinning parameters and graphene suspension on the nanofiber morphology and physical properties were analyzed. The electrospinning processing parameters were optimized using the characteristics given in Table 3. Figure 6 shows TEM micrographs of nanocomposite nanofibers with 5 and 15 wt.% graphene contents. The nanofiber diameter was in the range of 200–300 nm. With increasing nanofiller loading, a protruded nanofiber surface was observed due to nanofiller aggregation in the matrix. The graphene disposition along the nanofiber surface was observed due to nanosheet stacking on the nanofiber surface. A classical approach was applied to analyze the macroscopic width and thickness of the material. The free volume among nanofibers was found to be filled by the bulk material (Figure 7). The porosity in the nanofibrous mat was observed due to the free volume of polymer (~80%), compared with the total volume of the nanomaterial.

Table 3. Electrospinning parameters used for nanofiber production [148]. Reproduced with permission from Elsevier.

Electrospun Solution	Graphene Content ppm (wt.%)	Flow Rate/mL/h	Electric Potential/kV	Distance/cm	Humidity (%)
NY-0G	0	0.32	20.0	17.0	38–40
NY-0.05G	500 (0.05)	0.23	20.8	18.0	43–45
NY-0.1G	1000 (0.1)	0.25	21.0	18.0	54–45
NY-1.5G	15,000 (1.5)	0.17	18.0	18.0	39–31
NY-2G	20,000 (2.0)	0.50	16.7	15.0	33–35
NY-5G	50,000 (5.0)	0.70	17.3	15.0	28–30
NY-8G	80,000 (8.0)	0.30	15.1	20.0	24–26
NY-15G	150,000 (15.0)	0.50	20.0	15.0	31–33

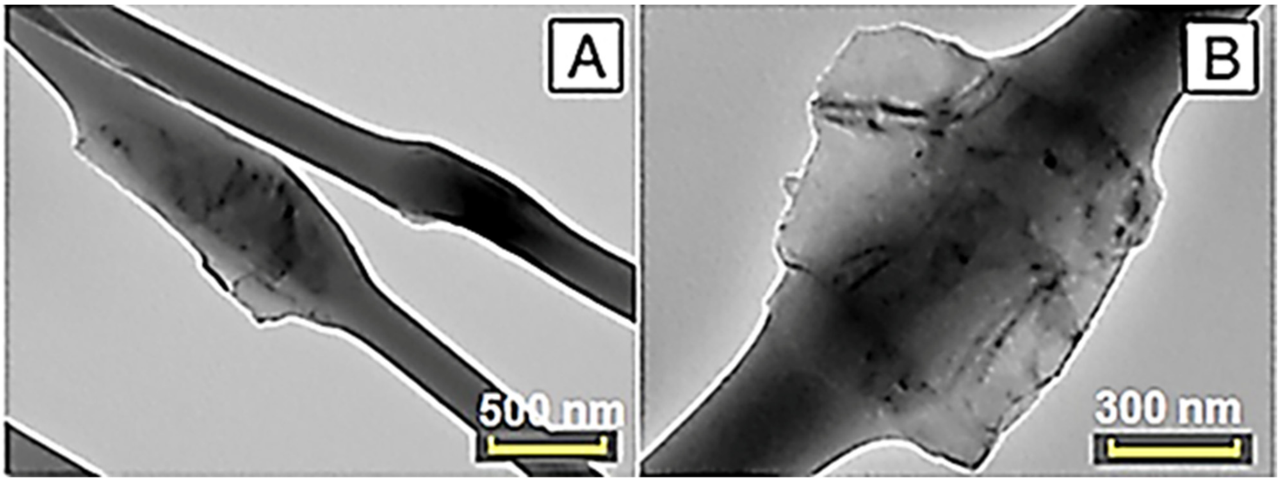

Figure 6. TEM micrographs of nanofibrous mats with (**A**) 5 wt.% graphene and (**B**) 15 wt.% graphene [148]. Reproduced with permission from Elsevier.

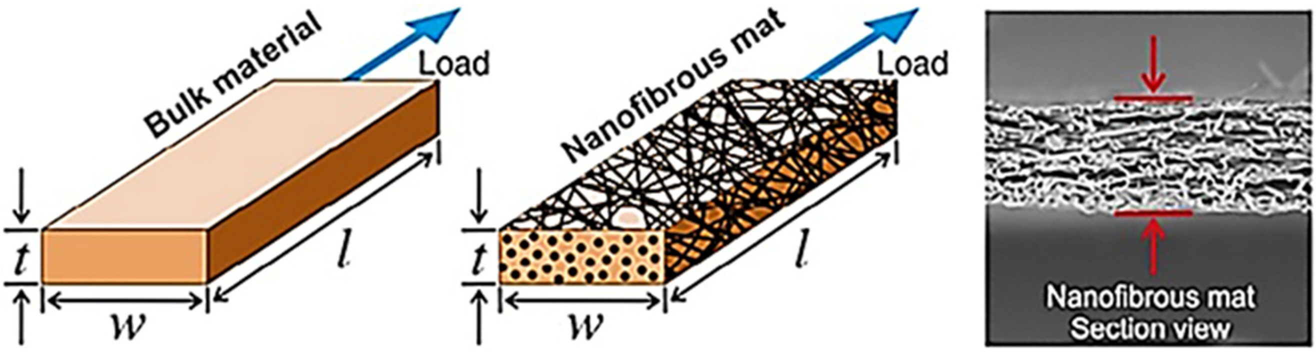

Figure 7. Schematic representation of bulk versus nanofibrous material and cross-sectional area of nanofibrous material [148]. Reproduced with permission from Elsevier.

The best quality nylon 6,6/graphene nanofibers (homogeneous surface and no bead formation) with a diameter of 260 nm were attained at a high flow rate of 0.70 mL/h. The effect of graphene on the mechanical properties of nanofibers such as stretch, slide, twist, and re-orientation was observed [149]. The nanofibers' quality was also analyzed after 20 months of aging. The nanocomposite nanofibers did not reveal any structural or texture alteration when compared with pristine nanofibers. Moreover, the mechanical properties of the nanofibers were also maintained after 20 months of aging, thus confirming the polymer/graphene nanofiber stability.

Leyva-Porras et al. [150] manufactured the electrospun nanofibers of pristine nylon 6 and nitroxide-functionalized graphene oxide reinforced nylon 6 nanocomposites. Figure 8 demonstrates the TEM image of neat Nylon 6 nanofiber. The average nanofiber diameter was ~200 nm. The nanofibers have a length of several microns. The Z-contrast image revealed two nanocomposite nanofibers situated perpendicular to one other. In both the nanofibers, fine graphene nanosheet dispersion was observed. TEM analysis revealed the deposition of functional 1–4 graphene oxide layers in nylon nanofibers. Few graphene oxide nanosheets were found fully embedded within the polymer fiber, whereas few nanosheets protruded from the fiber. Consequently, the simple reinforcement mechanism was found to affect the dispersion properties and nanofiber diameter.

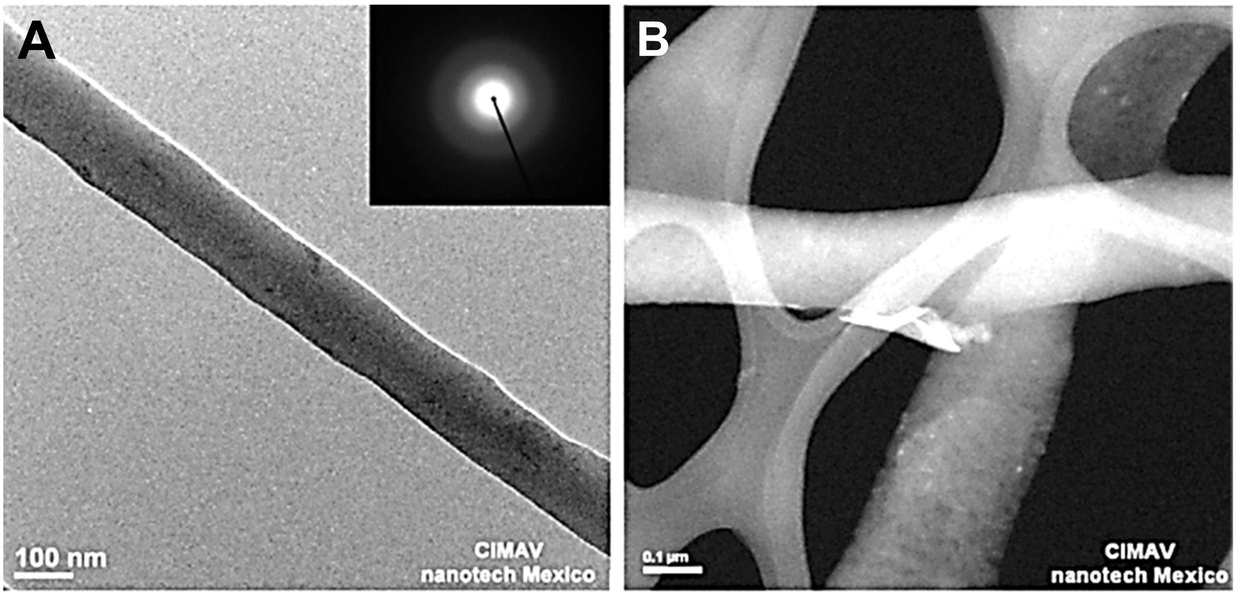

Figure 8. (**A**) TEM bright field image of nylon 6 nanofiber; (**B**) STEM Z-contrast image showing two nanocomposite nanofibers containing GOFT layers [150]. STEM = scanning transmission electron microscopy; GOFT = nitroxide-functionalized graphene oxide layers. Reproduced with permission from Elsevier.

Weise et al. [151] applied the melt spinning technique to fabricate polyamide 6 and polyamide 6/graphene nanocomposite nanofibers. The 3 and 5 wt.% graphene contents were filled in the polyamide 6 matrix using the melt compounding method. The effect of melt spinning process parameters was analyzed on the nanofiber properties. Here, the single filament was produced at a draw ratio of 2.5 and a winding speed of >100 m/min. The length-to-diameter ratio of nanofibers was ~2.0. Figure 9 displays the extrusion line and nanofiber winding system. According to differential scanning calorimetric analysis, neat nylon 6 had two crystalline peaks ~200 °C due to gamma and alpha phases. With the inclusion of graphene, the gamma peak disappeared due to a shift toward the alpha peak. The shift in peaks and merging were observed due to the alteration of the nylon 6 structure with increasing graphene contents. Moreover, the crystallization peak of pristine nylon 6 fibers appeared at ~189 °C, which was shifted to >195 °C with increasing nanofiller contents. Here, graphene was found to behave as a nucleating agent in the matrix to improve the crystallization effects. Additionally, the electrical conductivity of as-spun nanocomposite nanofibers was ~10 μ S m^{-1}, suggesting the anti-static textile application of the nylon 6/graphene nanocomposite [152].

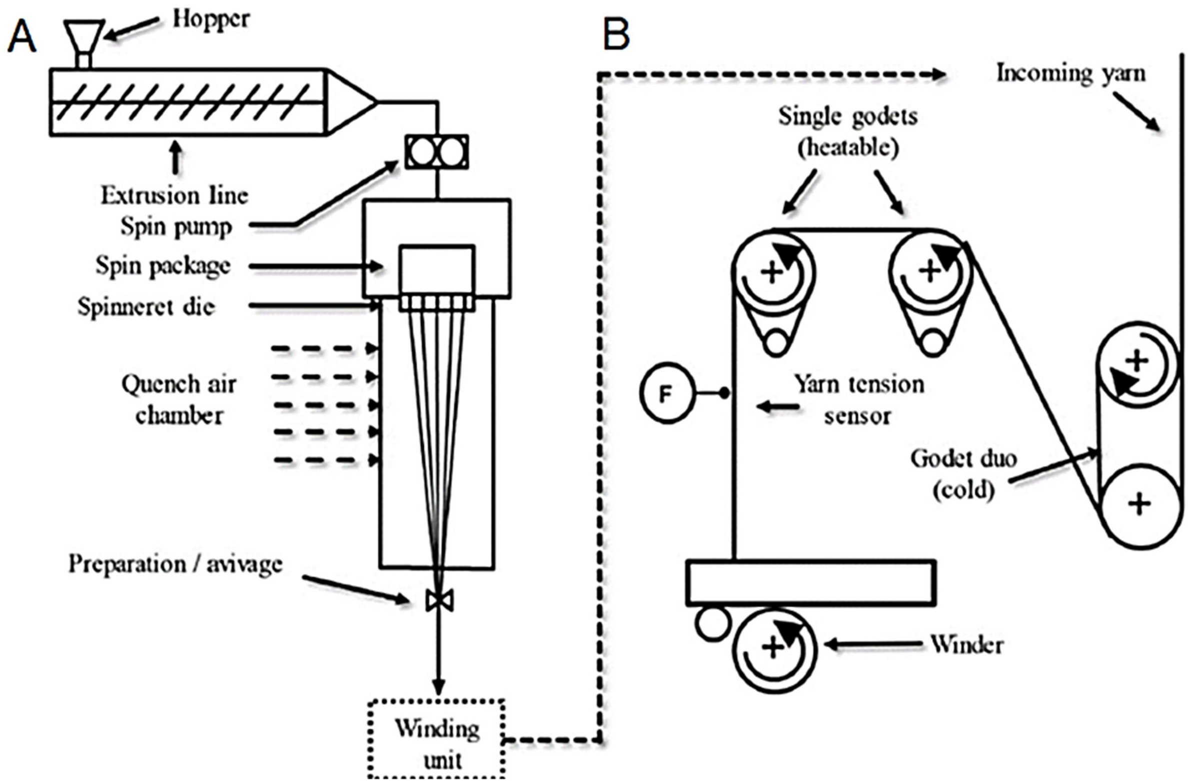

Figure 9. (**A**) The pilot-scale extrusion line and (**B**) winding system used to produce graphene-modified polyamide 6 yarns [151]. Reproduced with permission from Elsevier.

The polystyrene/graphene nanomaterials have been described in the literature [153–155]. Consequently, the polystyrene/graphene nanocomposite nanofibers have been processed using solution blending and electrospinning techniques [156]. Huang et al. [157] manufactured the electrospun polystyrene/graphene nanocomposite nanofibers. The hydrophobicity properties of the nanofibers were studied. Ponnamma et al. [158] developed the electrospun nanofibers of polystyrene, polystyrene/cobalt oxide, polystyrene/hexagonal boron nitride, and polystyrene/cobalt oxide/hexagonal boron nitride nanocomposites. The nanofibers were irradiated with gamma radiations to enhance the crosslinking density of the matrix nanofiller. This led to enhanced hydrophobicity and oleophilicity properties of the nanofibers. Figure 10 presents the contact angle studies of the nanofibers before and after gamma irradiation. Moreover, irradiation was found to enhance the surface roughness of the nanofibers. Neat polystyrene nanofibers had a contact angle of ~140°, which was enhanced to ~152° due to the superhydrophobicity of the nanocomposite. Due to these properties, the nanofibers have been applied as oil-water separators to hold oil molecules and filter the water molecules. Table 4 demonstrates the tensile strength and Young's modulus of the nanocomposite fibers. The mechanical properties of the nanofibers were enhanced with the nanofiller loading [159]. In both the irradiated and non-irradiated samples, the inclusion of two types of nanofillers enhanced the mechanical properties due to synergistic effects. Moreover, the gamma irradiation also improved the mechanical properties owing to matrix–nanofiller crosslinking and strengthening effects.

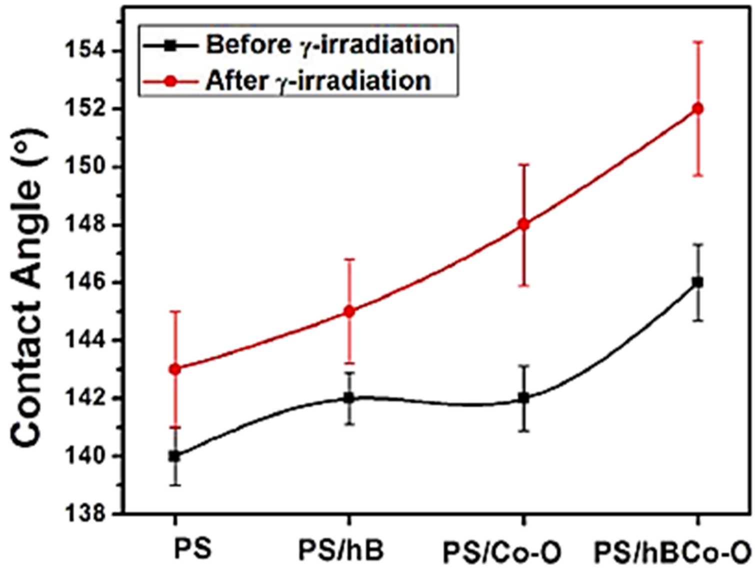

Figure 10. Contact angle of nanocomposite nanofibers before and after gamma irradiation [158]. PS = polystyrene; PS/Co-O = PS/Co_3O_4 at 1 wt.%; PS/hB = polystyrene/hexagonal boron nitride at 1 wt.%; PS/hBCo = PS/hexagonal boron nitride/Co_3O_4 at 1 wt.%. Reproduced with permission from MDPI.

Table 4. Mechanical properties of the nanocomposites. [158]. PS = polystyrene; PS/Co-O = PS/Co_3O_4 at 1 wt.%; PS/hB = polystyrene/hexagonal boron nitride at 1 wt.%; PS/hBCo = PS/hexagonal boron nitride/Co_3O_4 at 1 wt.%. Reproduced with permission from MDPI.

Sample		Tensile Strength (MPa)	Young's Modulus (MPa)
Non-irradiated	PS	28.54 ± 1.1	72.50 ± 6.44
	PS/Co-O	30.47 ± 2.3	76.33 ± 2.21
	PS/hBN	35.44 ± 2.5	77.65 ± 4.05
	PS/hBCo-O	48.24 ± 2.6	98.15 ± 4.79
Irradiated	PS	30.87 ± 1.3	73.30 ± 9.01
	PS/Co-O	31.05 ± 2.2	77.88 ± 8.75
	PS/hBN	38.25 ± 1.7	82.03 ± 2.45
	PS/hBCo-O	52.54 ± 2.1	110.35 ± 4.55

Poly(vinyl alcohol)/graphene nanocomposites have been developed [160–162]. Consequently, the poly(vinyl alcohol)/graphene nanocomposite nanofibers have been processed through electrospinning by Abdah et al. [163]. The inclusion of graphene in nanofibers reduced the diameter of nanofibers from 121 nm to 117 nm. The decrease in diameter was due to better interactions and homogeneous crosslinking in the matrix–nanofiller. The supercapacitor electrode based on poly(vinyl alcohol)/poly(3,4-ethylenedio xythiophene)/graphene nanocomposite nanofibers had a specific capacitance of 224.3 Fg^{-1} and power density of ~304.4 Wkg^{-1}. The specific capacitance was found higher than the non-filled poly(3,4-ethylenedioxythiophene) (167.92 Fg^{-1}) and poly(vinyl alcohol)/poly(3,4-ethylenedioxythiophene) nanofibers (182.73 Fg^{-1}). Enhancement in specific capacitance of nanocomposite nanofibers was observed due to an interconnecting network formation for electron/charge transportation. Barzegar et al. [164] also manufactured the electrospun poly(vinyl alcohol)/graphene nanocomposite nanofibers. The as-spun hollow nanofibers had uniform surfaces and fine graphene dispersion. Neat poly(vinyl alcohol) nanofibers have a diameter of 130–230 nm. The inclusion of graphene reduced the average diameter of nanofibers to ~238–302 nm. It appears that the graphene in polymer solution enhanced the electrical conductivity properties, so influencing the nanofiber diameters under the applied field effect. Moreover, the inclusion of graphene enhanced the thermal stability of the poly(vinyl alcohol) nanofibers. The maximum decomposition temperature of the nanocomposite nanofibers (393–542 °C) was found higher than the neat poly(vinyl alcohol) nanofibers (383–460 °C). The enhancement in the thermal conductivity of the nanocomposite nanofibers was attributed to the stable interlinked polymer–graphene nanostructure. Bao and co-workers [165] prepared the poly(vinyl acetate) and graphene-derived nanofibers via electrospinning. Nanocomposite nanofibers have chemically tunable optoelectronic properties along with enhanced mechanical and thermal properties, and processing advantages. The addition of graphene has enhanced the optical absorption properties of the poly(vinyl acetate)/graphene nanocomposite. The nanocomposite nanofibers were suggested to have applications in ultrafast photonics. For this application, the diameters of the nanofibers should be smaller than the optical wavelength. In this regard, the polymer/graphene nanocomposite nanofibers were attached on the faces of optical fibers to function as a passive mode-locker for ring laser cavity. The polymer-/graphene-based mode-locking performed better than the polymer absorbers in a wide wavelength range of around 10–30 nm.

Natural polymer- and graphene-based nanocomposite nanofibers have also been reported [166–168]. The cellulose, chitosan, and chitin-derived nanofibers have been designed and applied for biomedical applications such as tissue engineering, drug delivery, wound dressing, and other uses [169]. Neibolts et al. [170] designed biodegradable nanofibers of nanofibrillated cellulose, poly(butylene succinate), and graphene-based nanofibers. The electrospinning method was used to form the nanofibers with poly(ethylene glycol) as a compatibilizer. The nanocomposite nanofibers were used for the tissue engineering scaffolds. De Faria et al. [171] formed the electrospun chitosan/poly(lactide-co-glycolide) and graphene oxide-based nanocomposite nanofibers. The biodegradable nanofibers have fine antimicrobial properties towards Escherichia coli, Pseudomonas aeruginosa, and Staphylococcus aureus bacteria. Yoon et al. [172] prepared the electrospun poly(d,l-lactic-co-glycolic acid) and graphene oxide-derived nanocomposite nanofibers. The polymer and nanofiller were well-linked through the interfacial interactions. The hydrophilicity properties as well as the biocompatibility of the nanocomposite nanofibers were observed for the neuronal cells [173].

Other than electrospun polymer/graphene nanofibers as reported above, few reports have been observed on the solution blow spinning formed nanofibers so far [174]. Mishra et al. [175] designed the metal-doped polymer/graphene nanofibers using the solution blow spinning technique. The nanofibers have been applied to construct the lithium-ion battery electrodes. Some authors in the literature noticed the polymer/graphene nanofibers produced through the centrifugal jet spinning method [176]. Amir et al. [177] developed the graphene nanoplatelets reinforced polyurethane/phenolic resin nanofibers using the centrifugal jet spinning technique. Here, the nanofiber diameter was greatly affected by the processing parameters such as the polymer concentration, rotation speed, and pressure. The graphene nanoplatelets were found to be finely dispersed and adhered to the nanofibers. Matharu et al. [178] also fabricated the poly(methyl methacrylate)/graphene nanofibers through a centrifugal jet spinning procedure. The graphene nanofiller was loaded around 2–8 wt.% contents. An increasing amount of nanofiller affected the morphology of the nanofibers. Scanning electron microscopy exposed the beaded and porous nature of the nanofibers with an increased amount of graphene contents. The nanofiber diameter was found in the range of 0.75–2.71 mm. Electrohydrodynamic direct writing has been rarely used for polymer graphene nanofibers [179]. For example, Wang et al. [180] designed the homogeneously aligned graphene nanostructures using the direct writing approach. The morphology of nanofibers was controlled by means of the optimized manufacturing parameters. Details of the centrifugal jet spinning, solution blow spinning, and direct writing techniques used for the nanofiber formation and comparison with electrospinning are already stated in Section 2.

5. Applications of Polymer/Graphene Nanocomposite Nanofiber

Natural polymer fibers of cotton, silk, linen, etc. have been studied [181]. A wide variety of synthetic polymer fibers have been produced using polyamide, polyester, polyethylene, polyacrylonitrile, etc. [182,183]. Synthetic polymer nanofibers have low density [184], durability [185], toughness [186], strength [187], abrasion resistance [188], chemical stability [189], and eco-friendliness [190]. The as-prepared polymer fibers have been used to form films, membranes, packings, textiles, and biomedical materials [191–193]. Further advancements in the properties of polymer nanofibers have been observed with the nanofiller addition [194]. Polymeric nanocomposite nanofibers have found applications in electronics, sensors, actuators, energy storage materials, and membranes [195]. Electronic devices have been considered a continuous source of environmental pollution [196]. Moreover, electromagnetic radiation may hinder the functioning of electronic systems [197]. Incidentally, nanocomposites have been used as shielding materials against electromagnetic radiation [198,199]. Conducting polymers such as polyaniline-based nanofibers have been developed for electromagnetic interference (EMI) shielding [200,201]. Furthermore, the polyaniline/nanocarbon nanocomposite nanofibers were applied for EMI shielding [202–204]. Lyu et al. [205] fabricated the polyaniline/aramid/graphene nanofibers using electrospinning. The nanofibers revealed the high strength of 179 MPa. In addition, the nanocomposite nanofibers had high EMI shielding effectiveness of 30 dB. However, limited attempts have been observed on the polymer/graphene nanofibers for EMI shielding.

Supercapacitors have been researched as promising energy storage devices [206]. Supercapacitors have the capability of efficient charge storage, avoiding any chemical reactions [207]. Nanocarbon-based materials have been successfully used in the supercapacitor electrodes and electrolytes, due to high electrical conductivity, optimum porosity, and chemical stability properties [208]. Consequently, the carbon nanoparticles like graphite, graphene, carbon nanotube, etc. have been focused [209–211]. Polymer/nanocarbon nanocomposites have also been used to design supercapacitor electrodes [212]. Rose et al. [213] manufactured the polyaniline/poly(vinyl alcohol)/graphene oxide nanofibers for supercapacitor electrodes. The nanocomposite nanofibers-based electrode had a high surface area and charge carrier mobility, leading to high capacitance performance. The manufacturing of polymer/graphene nanofiber-based electrodes also demands comprehensive studies for further advancements.

An important use of polymeric nanofibers has been observed for membrane fabrication [214,215]. The membranes based on the polymer/graphene and polymer/graphene oxide nanocomposite nanofibers have been reported [216–218]. The poly(D, L-lactic-co-glycolic acid)/graphene oxide nanocomposite nanofibrous membranes have superior mechanical properties due to fine graphene dispersion and interfacial interactions in the matrix–nanofiller [172,219,220]. The polyacrylonitrile/graphene and polyacrylonitrile/graphene oxide-derived nanofibrous membranes have been prepared [221–223]. The membranes were analyzed for physical properties and water purification performances [224]. The addition of graphene nanofillers facilitated the permeation and filtration properties of the membranes [225].

Essential uses of polymer/graphene nanofibers have been observed in the biomedical field. The nanocomposite nanofibers have great potential for biomedical applications due to biocompatibility and bioresorbability properties (Figure 11) [226]. Various patents have been reported for the application of graphene-based nanofibers in biomedical fields [227,228]. Graphene nanocomposites have been applied for tissue engineering, pharmaceutical, drug delivery, gene delivery, wound healing, and biomedical devices and systems [229,230]. For pharmaceutical applications such as drug transfusion, the nanofibers have fine biocompatibility and anionic-exchange properties.

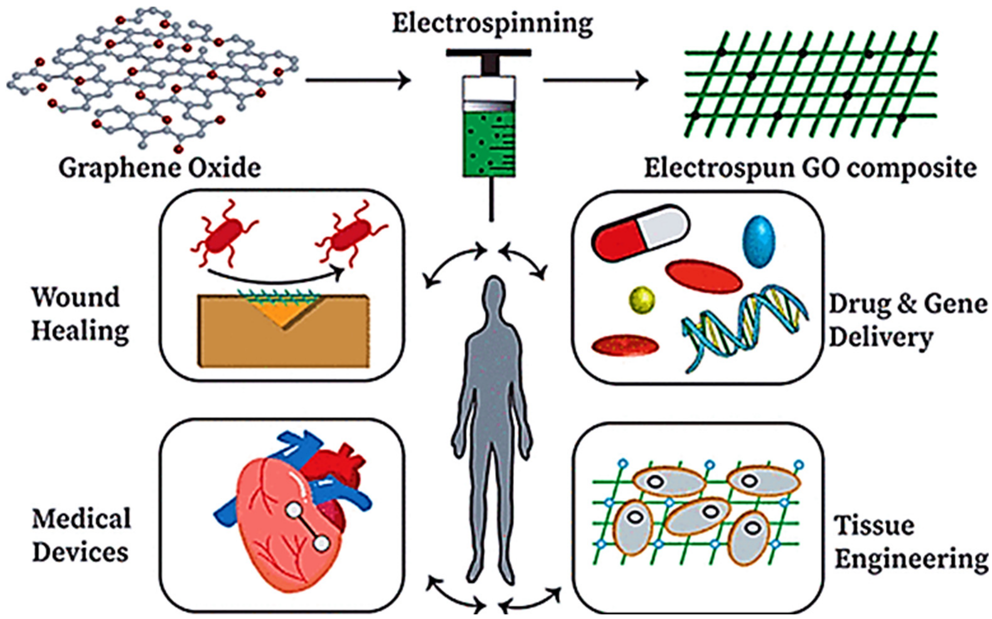

Figure 11. Biomedical applications of graphene-based nanofibers [226]. Reproduced with permission from ACS.

Systematic studies have been carried out to assess the safe use of graphene nanofibers in pharmaceutical and drug delivery applications [231]. For these applications, the nanofibe rs must be biocompatible, cytocompatible, bioresorbable, and non-toxic. The potential pharmaceutical use of polymer/graphene nanofibers has been observed for cancer therapy [232]. The electrospun graphene nanocomposite nanofibers have the capability for proliferation, differentiation, cell adhesion, and viability for drug delivery systems. Consequently, nanofibers have found use in regenerative medicine [233]. Gupta et al. [234] prepared graphene nanofibers-based sorbent materials for managing pharmaceutical pollution. Inclusion of 20 wt.% graphene oxide in cellulose nanofibers enhanced the adsorption removal capacity up to 45.04 mg g^{-1} and 85.30 mg g^{-1} for ciprofloxacin and ofloxacin, respectively.

Graphene oxide nanofibers have found important uses in wound healing applications [235]. Recent patents revealed the wound-healing capability of graphene nanofibers in the medical sector [236]. The chitosan/graphene nanofibrous membranes were developed for anti-bacterial purposes [237]. The poly(vinyl alcohol))/graphene nanocomposite nanofibers have been manufactured for tissue engineering applications [238,239]. Moreover, poly(D, L-lactic-co-glycolic acid)/graphene oxide nanocomposite nanofibers have been studied as bioengineering scaffolds [240,241].

Applications of graphene-containing nanocomposite nanofibers have been used for medical devices including coatings, sensors, and triboelectric nanogenerators [242,243]. Progress has been observed in the areas of sensing and biosensing [244,245]. Moreover, the polymer/graphene nanofibers were researched for cardiac patches and other medical devices [246].

6. Future Prospects

In this article, the manufacturing and properties of several polymer/graphene nanoco mposite nanofibers systems have been reviewed. The factors such as graphene contents, dispersal, and matrix–nanofiller interactions have been found significant to determine the physical features of the polymer/graphene nanocomposite nanofibers such as microstructure, thermal, mechanical, conducting, and other characteristics. Graphene-based polymeric nanocomposites always face major challenges regarding dispersion. In particular, graphene dispersion may be a challenging problem during large-scale production of related nanocomposite materials. Here, the nanofiller aspect ratio, orientation, polymer type, and nanofiller contents directly affect the nanofiller dispersion in the matrix. Due to consistent nanofiller dispersion, there is a large interfacial area between the matrix and graphene reinforcement. The implementation of an efficient synthesis technique, optimum processing conditions, and the choice of the matrix have also been considered important for fine dispersion and enhanced physical properties. Due to fine dispersion, the microstructural properties of the nanocomposite nanofibers have been found to be enhanced. An important effect of graphene dispersion has been observed on the formation of percolation networks in the polymer matrices to enhance the electrical conductivity properties. Owing to fine nanoparticle dispersal and matrix–nanofiller bonding, stress transfer through the nanocomposite has been increased, leading to high mechanical properties. The thermal stability properties of the polymer/graphene nanocomposite nanofibers have also been found to depend upon the graphene dispersion and interface formation for enhancing the heat stability of the materials. Advanced structure–property relationships have been suggested to establish the high efficiency and applications of the nanocomposite nanofibers.

The graphene-derived nanofibers have been fabricated using the in situ solution/chemi cal approaches, template, method, melt processing, and electrospinning techniques. Among all techniques, electrospinning has been found to be the most efficient to enhance the morphological and physical properties (conducting, thermal, mechanical, etc.) of the polymer/graphene nanocomposite nanofibers. In electrospinning, a polymer solution or melt is initially prepared for better graphene dispersion. Graphene dispersion has been found as an important factor to define the texture and microstructure of the nanofibers. Significant application areas of the polymer/graphene nanocomposite nanofibers include EMI shielding, supercapacitors, membranes, and the biomedical field. In the case of EMI shielding materials, nanofibers need to be developed using a modified graphene nanofiller to enhance the electrical conductivity and EMI shielding effectiveness. Similarly, for the development of supercapacitor electrodes, functional graphene can form an interconnecting network to support electron and charge transportation. Consequently, the fields of EMI shielding and supercapacitors need further research efforts to attain high-performance materials. Few research endeavors have been observed for the polymer/graphene nanocomposite nanofibrous membranes aiming for biomedical applications. Focused future attempts have been required to reveal the true potential of these materials for pharmaceutical and biomedical systems. Additionally, the novel polymer/graphene nanocomposite nanofibers must be manufactured and researched for the aerospace, automobile, coatings, and textile fields. Here, challenges regarding graphene dispersion, modification, ultrahigh conductivity, heat stability, and strength properties need to be overcome, as discussed above.

For effective future utilization of polymer/graphene nanocomposite nanofibers in EMI shielding, supercapacitors, membranes, biomedical, and other fields, certain challenges need to be overcome. Most importantly, the electrospun polymer/graphene nanocomposites have attained progress for EMI shielding. For the future application of polymer/graphe ne-based nanocomposite nanofibers in the EMI shielding materials, in addition to the fine nanofiller dispersion, the thickness and weight of these shields must be decreased to enhance the radiation shielding impact. In this concern, the optimization of polymer nanocomposite nanofiber properties during fabrication has been required for better EMI shielding. The nanofiller functionalization for the nanocomposite nanofibers definitely enhances the dispersion, interactions, conductive, and EMI shielding properties. For practical applications, the new hybrid systems have been designed for EMI shielding to efficiently function in the range of 8.2–12.4 GHz with an effectiveness of >30–50 dB [247]. Moreover, the design of graphene-derived layered nanocomposite shields with 8 to 16 layers can better reduce the impedance mismatch between the adjacent layers to enhance the EMI shielding performance. Hence, future high-performance shields can be developed by adjusting the number of layers to meet the commercial criterion. Additionally, there exist challenges to the commercialization of these nanofibrous EMI shields [248]. Presently, EMI shielding application demands the use of nanocomposite materials in harsh environments of high or low temperatures, and strong acids or strong bases medium. The surface of electrospun nanofibers must be modified with inorganic dopants to enhance their resistance properties. Moreover, the polymer/graphene nanofibers must have strong interfacial interactions to overcome the loss of mechanical performance during EMI shielding. Furthermore, the advanced EMI shields for radar technology must be developed with a wide-band frequency range of 8–26.5 GHz [249]. Undoubtedly, the electrospun polymer/graphene nanofibers have been advantageously used for supercapacitors; however, key issues have been faced during their use [250]. Generally, polymer/nanofibers have a large specific surface area, high electrical conductivity, and high electrochemical activity toward high-performance supercapacitor electrodes. In addition, the microporous nanofibrous structure may enhance the specific surface area to facilitate the ion transport and capacitance properties at low current densities. On the other hand, strong mechanical robustness has been desirable for the mesoporous electrodes while keeping flexibility. Here, it has been a challenge to integrate the porous functionalities with strength and flexibility together in the polymer/graphene nanofiber electrodes. Moreover, the mass production of these electrospun electrodes at low cost for large-scale applications has been the key issue. Graphene derived micro-supercapacitors designed on paper substrates can be a good addition to the future energy storage devices. The micro-supercapacitors have been focused according to the future demands of simple, flexible, and reasonable energy storage devices. To improve the overall device performance, the redox-active species may lead to the volumetric capacitance of 29.6 mF cm^{-3} [251]. The micro-supercapacitor devices can also be produced using printing techniques. The application of electrospun polymer/graphene nanofibers in membrane application demands the large-scale formation of these materials, which has been found to be difficult while maintaining a low cost [252]. Here, the foremost research analyzed for overcoming the challenges in transforming electrospinning from a lab-scale technology to an industrial-scale for talented applications. In addition, limited designs have been developed and studied for the separation membranes so far. Therefore, more research has been demanded in this area to overcome the related challenges. Additionally, the potential formation of new polymer/graphene nanofibers along with the post-treatment strategies have been desirable and challenging to control the pore formation on the nanofiber surface because, in membrane separation applications, surface area and porosity of nanofibers have been considered as key factors for efficient molecular transportation. Hence, the challenges related to the better separation applications can be overcome through the post-treatment techniques to endorse the transportation mechanism to the microporous nanofibers. The pore clogging issue of polymer/graphene membranes has been observed and solved by varying the membrane designs [253]. The antifouling polymer/graphene membranes have been developed using the functional graphene reinforced polymeric membranes. Here, polymer/graphene oxide-derived nanocomposite nanofiber membranes revealed the least microfouling phenomenon. Another advantage of polymer/graphene-derived nanofibrous membranes have been observed for the formation of freestanding, thin, and strengthened membranes for the water treatment systems. However, the gathering of pollutants (organic/inorganic contaminants like dyes, metal particles, bacteria/viruses, etc.) on the membrane surface can still prevent the rapid passage of clean water [254]. Additionally, the main challenges to using polymer/graphene nanofibers in tissue engineering and drug delivery applications involve attaining better biocompatibility and non-toxicity properties [255]. Challenges in using polymer/nanofiber nanomaterials in tissue engineering involve improving the function of damaged organs by forming precise biological substitutes [256]. In this regard, sufficient related research attempts have been desirable on the advanced polymer/graphene nanofibers. Moreover, the engineered polymer/graphene nanofiber tissues must have the capability to mimic the physiological environment perfectly by upholding their structural, topographical, and mechanical properties. Biocompatible graphene-based 3D scaffolds have been developed having mechanical and organ-biomimicking properties [257]. Owing to the large specific surface area and surface chemistry, the graphene-based 3D scaffolds can better interact with proteins/peptides to form strong interactions. Such nanocomposites have strong capability to direct stem cell differentiation to specific tissues including the cardiac cells, nerve, bone, etc. The porous graphene nanostructures also allow nutrient diffusion/waste discharge during tissue regeneration. Due to high electrical conductivity, graphene oxide-derived scaffolds have been applied for the cardiac and nerve tissue cultures. Further studies have been desirable to explicate these interactions and mechanisms towards organ-specific scaffolds for tissue engineering. Additionally, the challenges in drug delivery of polymer/graphene nanofiber can be overcome by sufficient research attempts in this field [237]. Current graphene research has focuses on the graphene nanomaterials with large surface area for loading biomolecules [258]. Toxicity of graphene-based drug carriers have also been researched for better future designs. Here, major encounters have been found related to the controlled loading and release of drugs in various mediums. New design combinations of polymer and graphene or graphene oxide-derived nanofibers may enhance the effective drug transport capacity for facile uptake by cells, so efficient drug delivery carriers can be designed.

7. Conclusions

This comprehensive review article presents an overview of polymer/graphene-based nanocomposite nanofibers. The design, manufacturing, structural, morphological, and physical characteristics of polymer/graphene nanocomposite nanofibers have been the focus. The polymer and nanocomposite nanofibers have been processed using various manufacturing techniques such as freeze-drying synthesis, interfacial polymerization, phase separation, template synthesis, drawing method, STEP method, electrospinning, and other spinning techniques. For graphene-based nanocomposite nanofibers, electrospinning has been found as a widely used and effective method. For graphene-based nanofibers, the choice of manufacturing technique, type of polymer, graphene form, solvent used, concentration, speed, flow rate, applied field, and other process parameters, and physico-chemical properties, influence the quality, morphology, diameter, and applications of the as-prepared nanofibers. The effect of manufacturing techniques and processing parameters on varying the nanofiber diameter and properties have been surveyed. Consequently, the manufactured graphene-based nanofiber properties including flexibility, modulus, tensile strength, toughness, electrical properties, thermal properties, capacitance, hydrophobicity, optical properties, chemical features, and other physical properties have been investigated. High-performance nanocomposite nanofibers have revealed potential for EMI shielding materials, supercapacitors, membranes, pharmaceutical, and biomedical-related devices and systems. Future advancements in the field of polymer/graphene nanocomposite nanofibers and related application areas depend upon the new design possibilities and invention of innovative manufacturing methodologies for these nanofibers, to overcome the related challenges.

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