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REVIEW ARTICLE

High-tech graphene oxide reinforced conducting matrix nanocomposites—Current status and progress

Ayesha Kausar[1,2,*], Ishaq Ahmad[1,2], Tran Dai Lam[3]

[1] *NPU-NCP Joint International Research Center on Advanced Nanomaterials and Defects Engineering, Northwestern Polytechnical University, Xi'an 710072, Shanxi Province, China. E-mail: dr.ayeshakausar@yahoo.com*

[2] *UNESCO-UNISA Africa Chair in Nanosciences/Nanotechnology, iThemba LABS, Somerset West 7129, South Africa.*

[3] *Institute for Tropical Technology, Vietnam Academy of Science and Technology, Hanoi 100000, Viet Nam.*

ABSTRACT

Graphene oxide can be referred to as oxidized graphene. Similar to graphene, oxidized graphene possesses remarkable structural features, advantageous properties, and technical applications. Among polymeric matrices, conducting polymers have been categorized for π conjugated backbone and semiconducting features. In this context, doping, or nano-additive inclusion, has been found to enhance the electrical conduction features of conjugated polymers. Like other carbon nanostructures (fullerene, carbon nanotube, etc.), graphene has been used to reinforce the conjugated matrices. Graphene can be further modified into several derived forms, including graphene oxide, reduced graphene oxide, and functionalized graphene. Among these, graphene oxide has been identified as an important graphene derivative and nanofiller for conducting matrices. This overview covers essential aspects and progressions in the sector of conjugated polymers and graphene oxide derived nanomaterials. Since the importance of graphene oxide derived nanocomposites, this overview has been developed aiming at conductive polymer/graphene oxide nanocomposites. The novelty of this article relies on the originality and design of the outline, the review framework, and recent literature gathering compared with previous literature reviews. To the best of our knowledge, such an all-inclusive overview of conducting polymer/graphene oxide focusing on fundamentals and essential technical developments has not been seen in the literature before. Due to advantageous structural, morphological, conducting, and other specific properties, conductive polymer/graphene oxide nanomaterials have been applied for a range of technical applications such as supercapacitors, photovoltaics, corrosion resistance, etc. Future research on these high-performance nanocomposites may overcome the design and performance-related challenges facing industrial utilization.

Keywords: Graphene Oxide; Conductive Polymer; Nanocomposite; Conductivity; Supercapacitor

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1. Introduction

Conductive, conducting, or conjugated polymers constitute an essential category of polymers with semiconduction or electron conduction properties[1,2]. Important types of conjugated polymers include polyacetylene, polyaniline, polythiophene, polypyrrole, and derivatives. Conjugated polymers have been recognized for their optical, electrical, thermal, and physical characteristics[3]. Technological applications of conductive polymers have been observed for electronics, energy devices, biomedical fields, and so on[4]. Graphene is a one-atom-thick two-dimensional nanosheet material[5]. Graphene oxide is simply a graphene derivative having oxygen-containing surface functionalities on graphene nano-sheet. Graphene oxide has been utilized to form polymer nanocomposites with conducting polymers, thermosets, and thermoplastic matrices[6,7]. Particularly conducting polymers are generally well-known due to their remarkable properties, such as low price, high conductivity, lightweightness, simple fabrication, and reusability potential. Doping of conducting polymers on graphene oxide surfaces usually leads to interface formation, which improves the electrical properties of the resulting nanocomposite[8]. Due to interface development between matrix-nanofiller, charge transfer across the interface is greatly promoted. Consequently, these nanocomposites possess fine durability, conductivity, corrosion resistance, mechanical strength, wear resistance, and barrier properties[9]. Several approaches have been used for the fabrication of conductive polymer/graphene oxide nanocomposite, including in situ, solution, electropolymerization, electrodeposition, etc.[10]. Consequently, conducting polymer and graphene oxide derived nanocomposites revealed suitability for application in technical sectors such as photovoltaics, capacitors, sensing devices, radiation shielding, etc.[11–13]. Therefore, the major theme behind developing this review article is to gather and portray the field literature and efforts reported on graphene oxide reinforced conducting polymer nanocomposites so far, in a novel way by identifying the property/potential advantages of combining conducting polymers with graphene oxide nanofiller.

Hence, this overview is designed to cover indispensable properties and potential aspects of graphene oxide reinforced conductive polymer nanocomposites. Owing to the unique structural combination of conjugated polymer and graphene oxide, essential features of the conductive nanomaterials have been enhanced. Subsequently, conducting polymer and graphene oxide-based nanocomposites were investigated, aiming for energy storage and production devices and anticorrosion nanomaterials.

2. Graphene oxide

Graphene is a nanocarbon nanoallotrope with sp^2 hybridized carbon atoms in nanostructure[14]. It is a monolayer of carbon with hexagonal arrangements. Graphene is simply a single sheet of stacked graphite nanostructure[15]. Graphene is a one-atom-thick nanosheet of carbon atoms[16]. Due to hybridization and π electron conjugation, the graphene nanosheet reveals semiconductivity features[17]. Graphene has been prepared using a number of top-down and bottom-up techniques, such as graphite chemical or mechanical exfoliation, chemical vapor deposition, plasma-based chemical vapor deposition, thermally enhanced chemical vapor deposition, laser ablation, organic synthesis, and other important techniques[18]. Graphene oxide is a significant modification of the graphene nanosheet through the incorporation of various surface functionalities[19]. Mainly hydrophilic groups like carbonyl, acid, epoxide, hydroxyl, etc. have been observed on the graphene oxide nanosheet. **Figure 1** gives a comparison of the simple structures of graphene vs. graphene oxide. Hummer's approach and the Brodie method have been commonly used for the formation of graphene oxide[20,21]. Both of these methods involve using oxidizing agents for the formation of graphene oxide from graphene[22]. Graphene oxide possesses high conductivity, heat stability, mechanical strength, and chemical stability features. Graphene oxide has been essentially reinforced in polymers to form nanocomposites. Polymer and graphene oxide derived nanomaterials have been applied for electronics, energy devices, membranes, and biomedics[23–25].

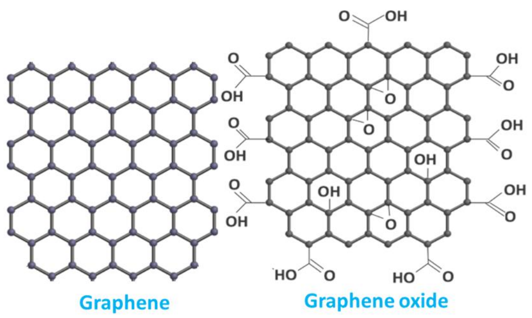

Figure 1. Graphene and graphene oxide.

3. Conducting polymers

Conductive polymers have been placed in a separate category of polymers and are not included in thermoplastics or thermosets classifications[26,27]. Inherently conducting polymers have conductivity properties similar to semiconductors[28]. Therefore, conducting polymers are often referred to as synthetic metals[29]. Conductive polymers have light weight and fine processability properties[30]. The π-conjugation in conducting polymer backbones formed an electron transportation system and electron affinity properties[31]. The presence of alternate single and double bonds in conducting polymers causes delocalization of electrons in sp^2 hybridized orbitals. Consequently, conductivity properties are actually due to electron transportation through double bonds and charge transfer by resonance. Doping agents and oxidation-reduction processes were applied to enhance the conducting characteristics of conductive polymers. Polyacetylene is an intrinsically conjugated polymer[32]. The doping process has been used to improve the electrical conductivity features of polyacetylene[33]. Other significant conductive polymers are polycarbazole[34], polythiophene[35], polypyrrole[36], polyaniline[37], and several derived forms (**Figure 2**).

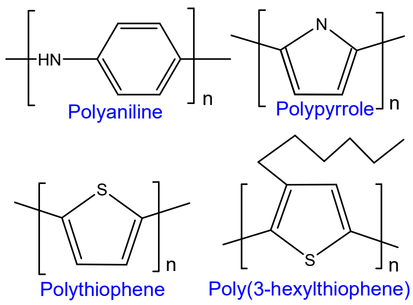

Figure 2. Some significant conjugated polymers.

Among these, polyaniline is the most widely studied conductive polymer. Polyaniline is a low-priced and easily possessable conjugated polymer[38]. Polyaniline has been studied for electron transportation and percolation threshold values. Consequently, conducting polymers have high electron conductivity and technical applications.

4. Graphene oxide nanofiller in conducting polymeric nanocomposites

Graphene and graphene oxide nano-reinforcements have been used to form polymeric nanocomposites[39]. Graphene derivative nanofillers have improved the electronic, strength, heat stability, and other features of the resulting nanocomposites[40]. However, graphene and derivatives can have poor dispersion in matrices because of the wrinkling effect. Graphene oxide possesses the advantage of better dispersion in polymers due to its surface functionalities[41]. Moreover, graphene oxide has been found to develop better interactions with the polymer matrix. Appropriate processing approaches need to be adopted for fine graphene oxide dispersion in polymer matrices[42]. Consistent graphene oxide dispersion in matrices has been found to enhance the electron conductivity, heat constancy, robustness, and physical features of the nanocomposites. In this context, conductive polymers have been filled with graphene oxide nanofiller to design efficient nanocomposites[43].

Polyaniline (an important conjugated polymer) and graphene oxide derived nanocomposites have been fabricated[44]. Li *et al.*[45] applied *in situ* polymerization for the development of polyaniline and reduced graphene oxide based nanocomposites. Gao *et al.*[46] also formed polyaniline and reduced graphene oxide based nanocomposites through *in situ* techniques. The reduced graphene oxide was produced by sodium borohydride. Chauhan *et al.*[47] fabricated polyaniline and reduced graphene oxide derived nanocomposites. Increasing reduced graphene oxide contents led to enhancements in electron conduction and specific capacitance properties. Graphene oxide was functionalized with sulfonic acid for modification. Consequently, the polyaniline and sulfonated graphene oxide revealed fine electron conduction features. Hawash *et al.*[48] fabricated polyaniline and graphene oxide nanosheet-immobilized granular tea waste-derived nanocomposites. The nanomaterial has been applied for effective removal of bromide (Br^-) from aqueous systems. The nanocomposite revealed bromide adsorption of 26.8 mg^{-1}. **Figure 3** shows an oxidation technique to form the polyaniline/graphene oxide nanosheet-immobilized granular tea waste nanocomposites. Graphene oxide functionalities have been found to interact with the functional groups on granular tea waste, such as carboxylic acid, hydroxyl, amine, amide, etc. Consequently, electrostatic as well as hydrogen binding interactions were developed to form the nanocomposite.

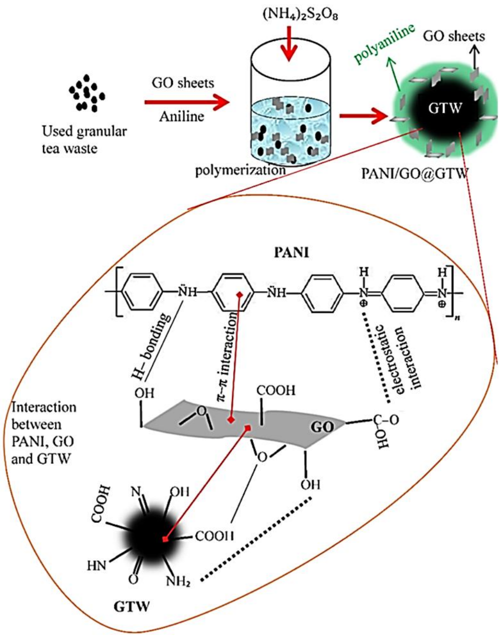

Figure 3. Schematic for the formation of PANI/GO@GTW nanocomposite[48].
PANI = polyaniline; GO = graphene oxide; GTW = granular tea waste; PANI/GO@GTW = polyaniline/graphene oxide nanosheet immobilized granular tea waste[48]. Reproduced with permission from MDPI.

Polypyrrole is a significant conductive matrix with facile preparation and electrical conductivity features[49]. Several designs have been formed using polypyrrole and graphene nanofiller[50]. Similarly, polypyrrole and graphene oxide derived nanomaterials have been reported[51]. Graphene oxide dispersion has been used to enhance the physical properties of the polypyrrole nanocomposites, such as electron mobility and thermal transport[52]. These nanocomposites have been developed using *in situ*, electrochemical, emulsion, and solution polymerization techniques. Deng *et al.*[53] developed polypyrrole and graphene oxide based nanocomposites by the electrochemical synthesis method. The inclusion of 0.5 to 1 wt.% graphene oxide led to an impedance variation of 115 to 26 kΩ.

Wu *et al.*[54] prepared polypyrrole, reduced graphene oxide, gold nanoparticles, and glucose oxidase based multilayered films through the electrodeposition method. **Figure 4** illustrates the stepwise synthesis process for the formation of polypyrrole/reduced graphene oxide/gold nanoparticles/glucose oxidase nanocomposite. Initially, the biosensor was synthesized through the electrodeposition of polypyrrole/reduced graphene oxide on a neat glassy carbon electrode. Then, gold nanoparticles and glucose oxidase were immobilized on the electrode surface. The glucose oxidase formed self-assembly along with gold nanoparticles on the polypyrrole/reduced graphene oxide nanocomposite surface. Gold nanoparticles can efficiently bond to biomolecules such as enzymes or nucleic acids via covalent linking. **Figure 5** depicts current-time curves using a glucose biosensor based on polypyrrole/reduced graphene oxide/gold nanoparticle nanocomposite in the applied potential range of 0.35 V to 0.55 V. It has been observed that increasing applied potential increases the current values. For the biosensor, a working potential of up to 0.5 V was used to attain high selectivity or sensitivity. The nanocomposite electrode was found to be efficient in detecting glucose in the range of 0.2 to 8 mM. The detection limit was found to be 5.6 μM. In addition, biosensor was found to has eco-friendly properties.

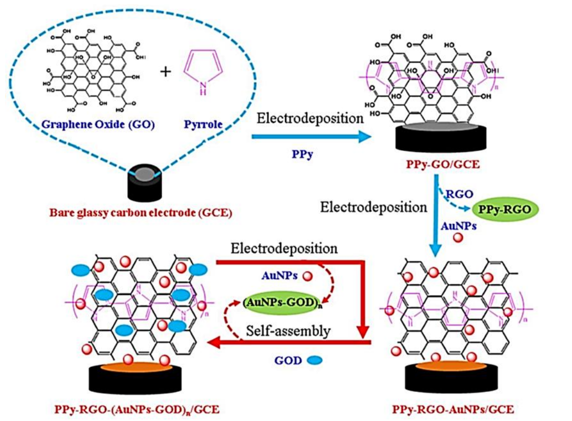

Figure 4. Schematic illustration of a glucose biosensor using the polypyrrole and reduced graphene oxide (PPy-RGO) and gold nanoparticles and glucose oxidase (AuNPs-GOD) multilayer films as the sensitive layer fabricated by the electrodeposition and self-assembly[54].
AuNPs = gold nanoparticles; GOD = glucose oxidase; PPy-RGO-AuNPs/GOD = polypyrrole/reduced graphene oxide/gold nanoparticles/glucose oxidase. Reproduced with permission from MDPI.

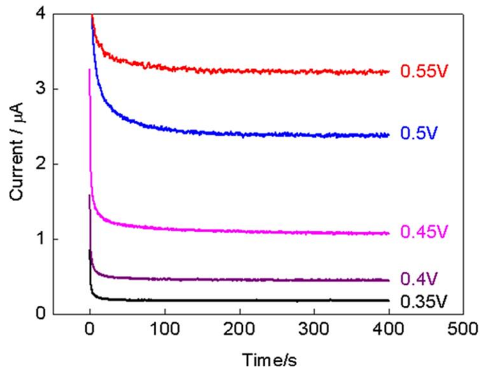

Figure 5. Current-time curves for PPy-RGO-AuNPs-GOD/GCE in 4 mM glucose at different applied potentials from 0.35 V to 0.55 V versus SCE[54].
PPy-RGO-AuNPs/GOD = polypyrrole/reduced graphene oxide/gold nanoparticles/glucose oxidase/electrode. Reproduced with permission from MDPI.

Polythiophene and derived forms have been utilized to form nanocomposites[55]. Shamsayei *et al*.[56] fabricated polythiophene and graphene oxide based nanocomposites by the electrochemical method. The microstructure and electron conduction features of the nanomaterials have been explored. Bora and researchers[57] used the interfacial polymerization method for the formation of polythiophene and graphene oxide based nanomaterials. Due to nanofiller loading, dispersion, and the formation of a percolation network, the nanocomposite had a high electrical conductivity of 2.7×10^{-4} S cm^{-1}. Thermal stability analysis revealed higher degradation temperatures in the range of 248–260 °C for the nanocomposites relative to the neat matrix (200–300 °C). Yang *et al*.[58] fabricated poly(3-hexylthiophene) and reduced modified graphene oxide based nanocomposite. The morphology studies were performed to study the nanoparticle dispersion in the matrix. The poly(3-hexylthiophene) was found layered on the nanofiller surface. Pilo *et al*.[59] used a polythiophene derivative, poly(2,5-di(2-thienyl)thieno[3,2-b]thiophene, with graphene oxide to form the nanocomposites. The resulting nanomaterials have been utilized for the formation of enzyme-sensing electrodes. The biosensor had a detection limit and sensitivity of 0.036 mM and 9.4 µA mM^{-1} cm^{-2}, respectively. Zamani *et al*.[60] fabricated the poly(3,4-ethylenedioxythiophene)/graphene oxide nanocomposite. The material was electrodeposited on solid phase microextraction fiber to form the sensing electrode. The nanocomposite electrode had a detection limit of 0.005–0.025 µg L^{-1} for tricyclic antidepressants (nortriptyline, amitriptyline, desipramine, imipramine, etc.). The sensing electrode revealed drug extraction of up to 105%.

5. Significance of graphene oxide filled conducting nanocomposites

Supercapacitors have been categorized as effective energy storage devices[61]. Conductive polymers and derived nanocomposites have been applied to form supercapacitors[62]. Polythiophene has been effectively used in supercapacitors owing to efficient charge mobilization, eco and chemical stabilization[63]. Moreover, polythiophene and graphene oxide derived nanocomposites have been functional for supercapacitance application[64]. Mostly, *in situ* polymerization route was adopted to attain polythiophene/graphene oxide nanocomposite for supercapacitors. The poly(3,4-ethylenedioxythiophene) nanocomposites filled with graphene oxide possess a high specific capacitance of about 201–320 Fg^{-1}[65]. The poly(9-butyl-3,6-di(thien-2-yl)-9H-carbazole) and graphene oxide based nanocomposite also have a specific capacitance of up to ~320 Fg^{-1}[66]. Interactions among polythiophene or polythiophene derivatives and graphene oxide occur *via* non-covalent bonding and π-π stacking interactions. These interactions have improved electron transfer, specific capacitance, and charge-discharge performance[67,68]. Zhou *et al*.[69] formed polypyrrole and graphene oxide derived nanocomposite through an electrochemical approach. The nanocomposite was electrochemically co-deposited on the fluorine doped tin oxide substrate. Then, the layered supercapacitor was formed by sandwiching the nanocomposite layer between the fluorine-doped tin oxide substrates. Li *et al*.[70] developed cellulose and graphene oxide based nanocomposites. Then, polyaniline was layered on cellulose/graphene oxide through *in situ* polymerization of aniline monomers. Cellulose/graphene oxide/polyaniline nanomaterial possess a high electron conduction of about 1.15 S cm^{-1}. The supercapacitor electrode had a sufficiently elevated specific capacitance of

1,218 mF cm^{-2} at 1.0 mA/cm^2. The flexible supercapacitor electrodes have revealed constant capacitance with twisting, so they can be used for flexible electronics. **Figure 6** shows a schematic for the formation of nanocomposite. Moreover, the contact behavior of water droplets on nanocomposite surfaces was studied for super wettability behavior. Due to the porous nature of the nanocomposite, a water droplet was easily penetrated. **Figure 7** depicts cyclic voltametric curves for in series and parallel devices as compared to a single device. The supercapacitor was used to light a red-light emitting diode for 4 minutes. Consequently, the nanocomposites have been used to form flexible, weight-less electronics.

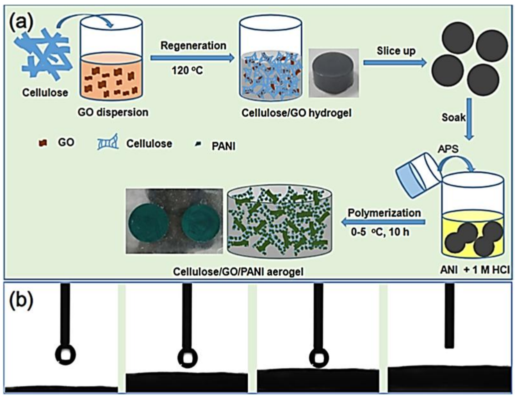

Figure 6. (a) Synthetic route to cellulose/graphene oxide/polyaniline nanocomposites; **(b)** the water droplet contact process on the nanocomposite surface[70].
GO = graphene oxide; PANI = polyaniline; ANI = aniline; APS = ammonium peroxydisulfate. Reproduced with permission from MDPI.

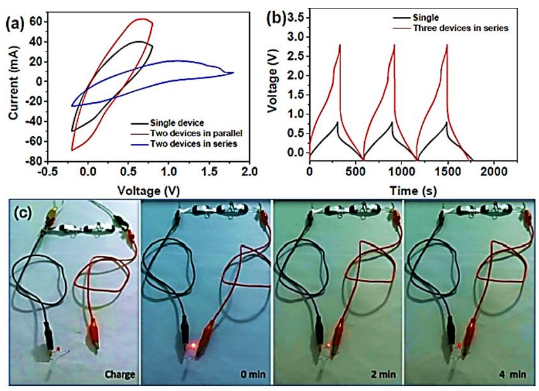

Figure 7. (a) CV profiles of single device, two devices in series, and two devices in parallel connection at a scan rate of 50 mVs^{-1}; **(b)** GCD profiles of single devices and three devices in series at a current density of 2 mA cm^{-2}; and **(c)** optical pictures of three devices in series connection to light a LED lamp for 4 minutes[70].
CV = cyclic voltammetry; LED = light emitting diode; GCD = galvanostatic charge/discharge. Reproduced with permission from MDPI.

Multipurpose energy production devices have also been focused on the use of conducting polymers[71]. In this context, conjugated polymers have been filled with efficient carbon nanoparticles such as fullerene, graphene, graphene oxide, and carbon nanotubes[72]. Ensuing nanocomposites have been used to improve solar cell efficiencies. Graphene oxide has been adopted as an efficient electron acceptor nanomaterial in solar cells[73]. Graphene oxide has a large surface area and electron-conducting pathways for electron passage through the material. Polythiophene and derivatives reinforced with graphene oxide have been integrated into solar cells[74]. Stylianakis *et al*.[75] fabricated and applied poly(3-hexylthiophene) and graphene oxide nanomaterials for bulk heterojunction solar cells. Poly(3-hexylthiophene) was used for electron donation, whereas graphene oxide worked for electron acceptance material[76,77]. Furthermore, graphene oxide formed a percolation network, allowing electron diffusion through the system[78]. Agbolaghi[79] produced the polyaniline and reduced graphene oxide nanomaterials through *in situ* techniques. The resulting nanocomposite has a solar cell efficiency of up to 7%.

Corrosion is a critical issue for metal-based industries[80]. In this context, different methods have been developed and applied for the corrosion protection of metals, such as the use of inhibitors and surface coatings[81]. Conductive polymers were reinforced with graphene and graphene oxide to develop anticorrosion coatings[82]. Moreover, pristine graphene oxide has been used as the a corrosion protective coatings[83]. Graphene oxide has been layered on nickel or copper metal for corrosion inhibition[84]. Similarly, graphene oxide reinforced polyaniline matrix has also been used as an anticorrosion coating[85]. Graphene oxide filled polythiophene nanocomposites have been applied for corrosion protection applications[86]. Electron conduction and corrosion defiance features of polythiophene/graphene oxide nanomaterials have been explored. However, few research efforts have been observed regarding polythiophene and graphene oxide derived anticorrosion nanomaterials so far.

Therefore, further investigations are desirable in this field to attain better designs and properties[87].

6. Prospects and conclusions

Continuous research efforts have been focused on conductive polymeric materials. Essential conjugated polymers such as polyaniline, polythiophene, polypyrrole, and derived polymers have been investigated with graphene oxide nanofiller. In this respect, various processing methods have been used to form the conductive polymer/graphene oxide nanomaterials. Consequently, structure, microstructure, and electron transportation properties have been explored. Interactions between conjugated polymers and graphene oxide have been found to enhance the features of the ensuing nanocomposites.

Inclusion of graphene oxide has been reported to enhance the electrical conductivity of the conducting polymer up to 50–90%. Enhancement in electrical conductivity depends upon the nanofiller dispersion and interaction with the matrix, which may ultimately lead to the formation of an electron-conducting network for percolation[88]. Cheng *et al.*[89] reported the fabrication and electrical conductivity properties of polyaniline and graphene oxide derived nanocomposites. Inclusion of 0.45 wt.% graphene oxide in polyaniline led to an electrical conductivity of up to 9.8 S cm^{-1}, which is 90% higher than that of the pristine polyaniline matrix. The synergistic effects between matrix-nanofiller were observed due to interactions between the oxygen functionalities of graphene oxide and amino groups on polyaniline and aromatic ring stackings, leading to fine dispersion and network formation. Good dispersion of graphene oxide in the conducting polymer matrix has enhanced surface-to-volume ratio and interfacial interactions, which contributed to overall enhanced electrical conductivity properties.

Supercapacitors have been developed using conjugated polymer/graphene oxide nanocomposites. An important use of polymer/graphene oxide nanocomposites was observed for photovoltaics. The corrosion resistant features of the conductive polymers and graphene oxide derived nanocomposites have also been studied. The anticorrosion features were enhanced using conducting polymers doped on graphene oxide surfaces. In the future, new design combinations and structure-property relationships of these nanomaterials need to be investigated for further developments in these fields. Moreover, research can be extended towards the development of efficient designs for microelectronics and digitally integrated circuits. The biomedical sector also needs to be explored for the application of conductive polymer/graphene oxide nanocomposites. Here, synthesis processes and mechanisms need to be investigated for the formation of high-performance conducting nanomaterials.

In this review, major problems regarding conducting polymer/graphene oxide nanocomposites have been identified, including the benefits of combining graphene oxide and conducting matrices, overall property/potential advantages, and challenges in this field. Graphene oxide has been identified as a low-cost, efficient nanocarbon[90]. In addition, lots of literature has been reported regarding the significant features and technical characteristics of conducting polymers and derived nanocomposites[91,92]. The main challenges discovered for conjugated polymers include poor processability and large-scale coating development. Consequently, hardly any conducting polymer/graphene oxide nanomaterials have been used for commercial level applications. However, reports have been observed for the future market of these nanomaterials[93]. Therefore, it is essential to investigate present research statistics and future forecasts on the upcoming industrial revolution of conducting polymer/graphene oxide nanocomposite in the form of this comprehensive review article[94]. As portrayed in this article, the detailed analysis of design, approaches, opportunities, and challenges has been found indispensable for the future development of conducting polymer/graphene oxide nanocomposites[95].

In short, this overview comprehensively covered the essential aspects of conducting polymer and graphene oxide derived nanocomposites. The morphology, electronic, thermal, and strength features of the nanomaterials were studied. In addition, the potential application areas for these nanomaterials have been stated, like energy storage, energy conversion, and anticorrosion. Hence, the remarkable structural, physical characteristics, and application areas of conducting polymer/graphene oxide nanocomposites have pointed towards efficient future nanomaterials for technical fields.

Conflict of interest

The authors declare no conflict of interest.

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