Review

Conjugated Polymer/Graphene Oxide Nanocomposites—State-of-the-Art

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Abstract: Graphene oxide is an imperative modified form of graphene. Similar to graphene, graphene oxide has gained vast interest for the myriad of industrial applications. Conjugated polymers or conducting polymers are well known organic materials having conducting backbone. These polymers have semiconducting nature due to π-conjugation along the main chain. Doping and modification have been used to enhance the electrical conductivity of the conjugated polymers. The nanocomposites of the conjugated polymers have been reported with the nanocarbon nanofillers including graphene oxide. This review essentially presents the structure, properties, and advancements in the field of conducting polymer/graphene oxide nanocomposites. The facile synthesis, processability, and physical properties of the polymer/graphene oxide nanocomposites have been discussed. The conjugated polymer/graphene oxide nanocomposites have essential significance for the supercapacitors, solar cells, and anti-corrosion materials. Nevertheless, the further advanced properties and technical applications of the conjugated polymer/graphene oxide nanocomposites need to be explored to overcome the challenges related to the high performance.

Keywords: graphene oxide; conjugated polymer; nanocomposite; conductivity; supercapacitor

1. Introduction

Conducting polymers or conjugated polymers form important group of polymers having semiconductivity and electronic materials [1]. The discovery of conjugated polymers is reported back to 1970s, which led to the development of wide range of conducting polymers polyacetylene, polycarbazole, polyaniline, polypyrrole polythiophene, etc. [2–4]. The conjugated polymers have beneficial electronic, optical, durability, heat stability, and other physical properties [5,6]. These polymers shown wide ranging applications in supercapacitors, sensors, photovoltaics, light emitting diodes, and electronic devices [7–9]. The conducting polymers have also been used in the biomedical field [10,11]. Graphene is a two dimensional one atom thick nanocarbon nanomaterial [12,13]. Graphene oxide, modified form of graphene, is remarkable candidate for the formation of polymeric nanocomposites [14,15]. Graphene oxide has been filled in the conjugated, thermoplastic, and thermosetting matrices. The polymer/graphene oxide nanocomposites have been explored for the thermal, mechanical, and electrical properties and advance applications [16,17]. Graphene oxide based nanomaterials have found applications in the photovoltaics, supercapacitors, sensors, batteries, fuel cells, radiation shielding, etc. [18–20]. In this review, essential prospects of the conducting polymer/graphene oxide nanocomposites have been presented. Particularly, the preparation, properties, and advanced performance of the conducting polymer based nanocomposites have been discussed. The conducting polymer/graphene oxide have found future potential for the efficient materials for energy devices and corrosion resistance materials. Conjugated polymer design and modification, and graphene oxide modification can be focused to improve the performance of the conducting polymer/graphene oxide in the technical applications.

2. Graphene Oxide

Graphene is a nanoallotropic form of carbon [21]. The word 'graphene' is derived from graphite with the suffix '-ene'. In graphite, the graphene nanosheets are stacked together through the weak dispersion forces. Graphene is a two dimensional monolayer of carbon atoms [22]. Graphene consists of a single layer of hexagonally arranges carbon atoms [23]. It is a thinnest known material in the word. The carbon atoms in one atom thick layer nanostructure are sp^2 hybridized and have delocalized π-electron clouds [24]. Graphite is an inexpensive source of graphene [25]. The graphene has been produced on large scale through the exfoliation and micro-mechanical cleavage of graphite. Moreover, the sophisticated techniques such as chemical vapor deposition, plasma enhanced chemical vapor deposition and thermal chemical vapor deposition techniques have been used [26]. Graphene oxide (GO) is an important modified form of graphene. GO simply consists of a graphene nanosheet with the surface groups [27,28]. GO possess the hydrophilic surface functionalities including the epoxide, carboxylic acid, hydroxyl, carbonyl, etc. [29]. Figure 1 compares the structure of graphene and GO. Various methods have been used to develop GO including the Brodie, Hummers and Offeman, and modified Hummers method [30]. Initially, Brodie method has been used i.e., the mixture of potassium chlorate and nitric acid converts graphite to GO [31]. Hummers and Offeman proposed the use of potassium permanganate, sodium nitrate, and sulfuric acid to transform graphite to GO [32]. An important use of GO has been found with the polymer matrices. GO has been used to enhance the electrical conductivity, thermal stability, thermal conductivity, chemical stability, and mechanical constancy. The polymer/GO nanocomposites have been used in the electronics [33], sensors [34], supercapacitors [35], energy devices [36], membranes [37,38], etc.
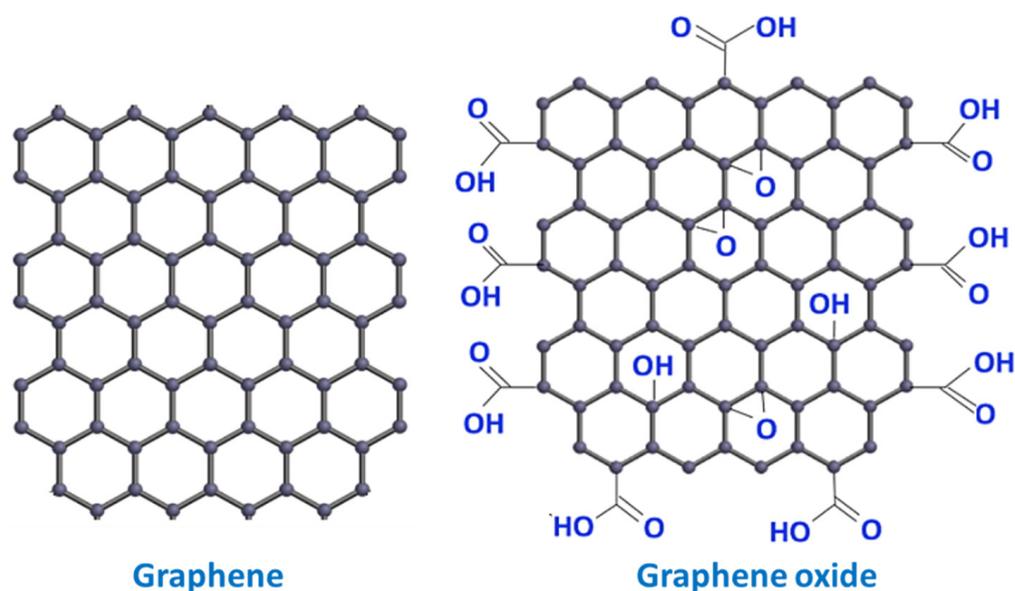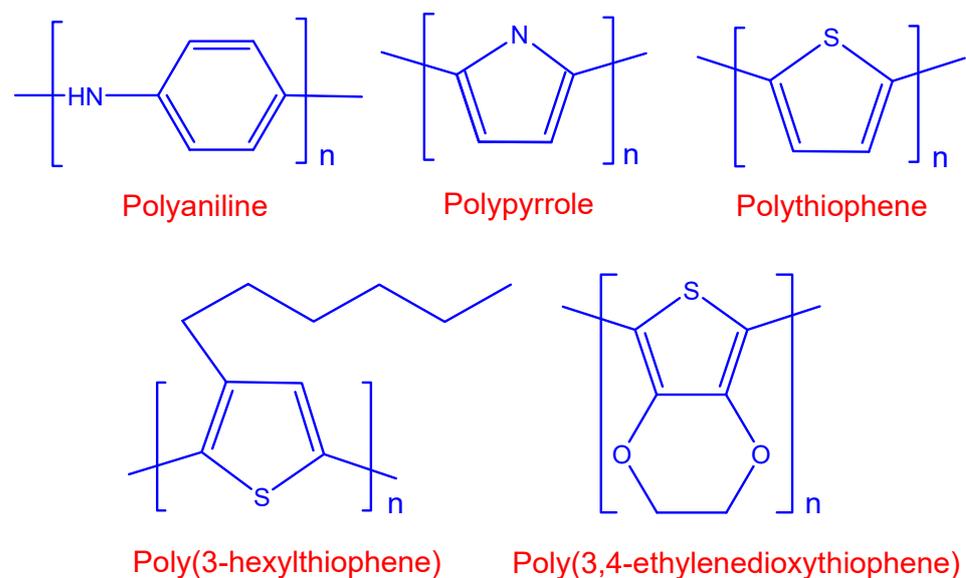

Figure 1. Graphene and graphene oxide.

3. Conjugated Polymer

Conjugated polymers or conducting polymers form an important separate class of polymers. Conjugated polymers are usually not characterized under the general groupings of the thermoplastic or thermosetting polymers. The intrinsically electrically conducting polymers or conjugated polymers have electrical and optical properties alike semiconductors [39]. However, the conjugated polymers have the advantages of light weight and easy processing compared with the metal based materials [40]. Such conjugated organic polymers are also known as *synthetic metals* [41]. The intrinsically conjugated polymers have integral conjugated electron system and high electron affinity [42]. There are al-

ternating single and double bonds in the structure causing delocalized electrons in the hybrid orbitals. The electrical conductivity of the conjugated polymers has been further enhanced using the dopants and oxidation-reduction reactions. The electron movement of the double bonds to the neighbouring atoms usually causes charge transfer through the system, which is often designated as *resonance*. The charge mobility may lead to the electrical conductivity of the polymers. The presence of charge in the conjugated polymers have caused increased doping capability of these materials. Initially, polyacetylene was discovered and considered as an intrinsically electrically conjugated polymer [43]. The electrical conductivity of polyacetylene has been studied and enhanced with the doping [44,45]. Other important electrically conducting polymers are polyaniline (PANI) [46], polypyrrole (PPy) [47], polythiophene (PTh) [48], poly (*p*-phenylene) [49], polycarbazole [50], and their derivative polymers (Figure 2). Among the conjugated polymers, PANI is an important and mostly used conjugated polymer. Polyaniline has fine processing, facile synthesis, employ inexpensive monomers, high conductivity, and potential applications [51]. Bhadra et al. [52] studied the electrical conductivity of PANI. The electron transport and percolation threshold phenomenon of the PANI matrix have been studied. All the intrinsically electrically conducting polymers have fine physical properties, the facile synthesis techniques, and the technical fields.


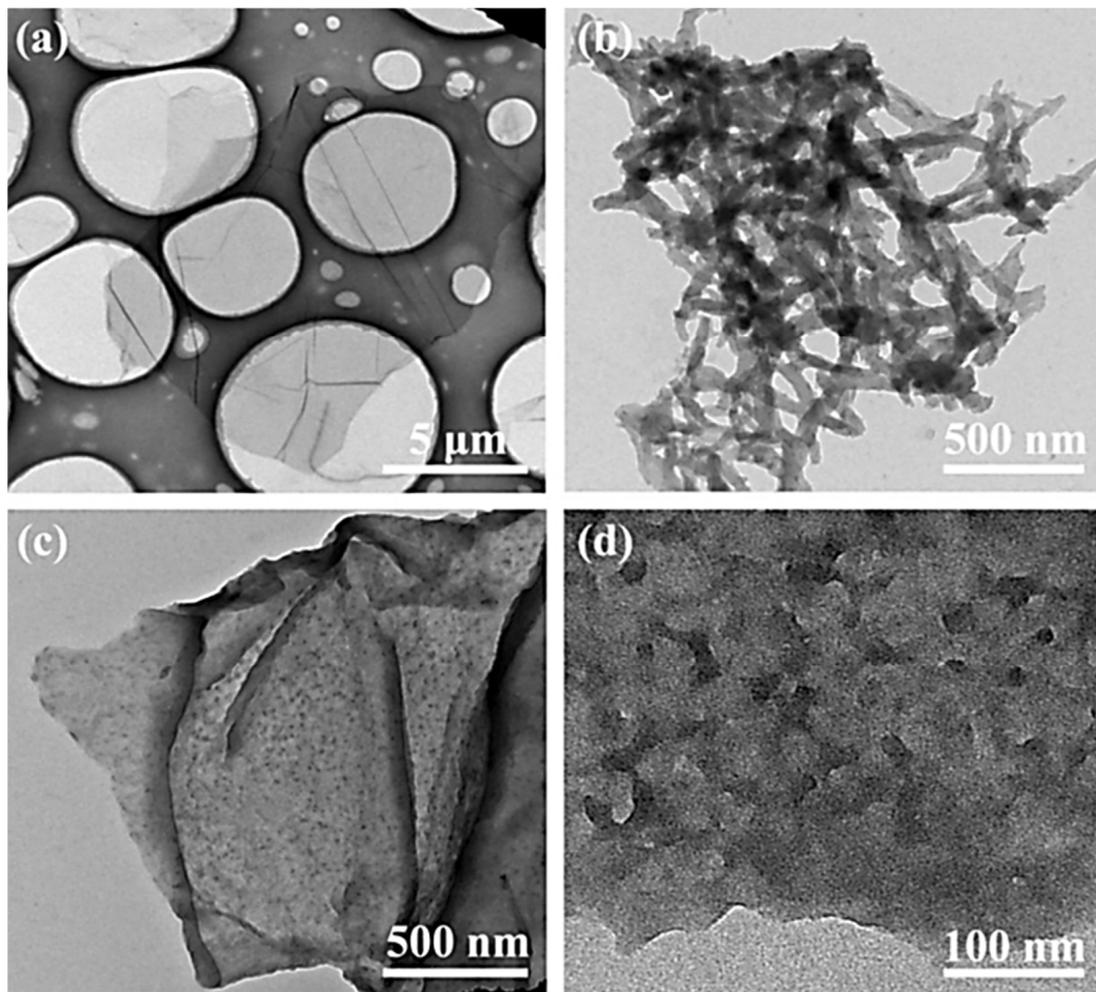**Figure 2.** Some important conducting polymers.

4. Conjugated Polymer/Graphene Oxide Nanocomposites

Graphene and GO have been considered as efficient nanofillers for the polymeric nanocomposites [53–55]. Graphene and GO have been used to improve the electrical, mechanical, thermal, and other physical properties of the polymer/graphene and polymer/graphene oxide nanocomposites [56,57]. Pristine graphene has dispersion problems in the polymer matrices due to the nanosheet wrinkling. As compared with graphene, GO possess surface functionalities for better dispersion in the polymer nanocomposites [58]. Consequently, the interfacial interactions between the polymers and GO nanosheets may resolve the dispersion problems. Appropriate processing techniques are also essential for better nanofillers dispersion [59]. The homogeneous GO dispersion may result in the high glass transition temperature, thermal stability, conductivity, and mechanical strength of the nanocomposites. Accordingly, the conjugated polymers such as PANI, PPy, and PTh have been filled with graphene and GO to form the nanocomposites [60–62].

Polyaniline and GO based nanocomposites have been successfully designed [63]. Li et al. [64] formed the polyaniline/reduced graphene oxide nanocomposites using in situ polymerization. The aniline monomer and GO contents affected the morphology of the nanocomposites. Chauhan et al. [65] filled the PANI matrix with the reduced GO to develop the nanocomposites. The increase in the reduced GO contents improved the electrical conductivity and capacitance of the polyaniline/reduced graphene oxide nanocomposites [66]. The surface functional GO such as substituted with sulfonic groups has also been used as the nanofiller for conducting polymers [67,68]. The polyaniline/sulfonated GO nanocomposites possess high electron transport properties [69]. Fan et al. [70] converted GO to sulfonated graphene and filled in the PANI matrix to form the polyaniline/sulfonated graphene (PANI/SG) nanocomposites. The in situ polymerization was used. Figure 3 depicts the process of the conversion of GO into SG nanosheet. Here, the benzene sulfonic acid was used to convert the GO into SG. Transmission electron microscopy (TEM) was used to study the morphology of SG obtained from GO, PANI, and SG/PANI nanocomposite (Figure 4a–c). The SG had plate-like structure, whereas neat PANI form nanorods. The PANI/SG shown homogeneously dispersed sheet like morphology. The homogeneous morphology was due to the interactions between the polymer and sulfonated nanofillers. Gao et al. [71] prepared reduced GO nanofiller using $NaBH_4$. Then, the reduced GO was covalently grafted to the aniline monomer using *p*-phenylenediamine, aryldiazonium salt, and $NaNO_2$. In this way, several systems of the polyaniline/graphene oxide nanocomposites have been designed and studied for the important properties and applications.
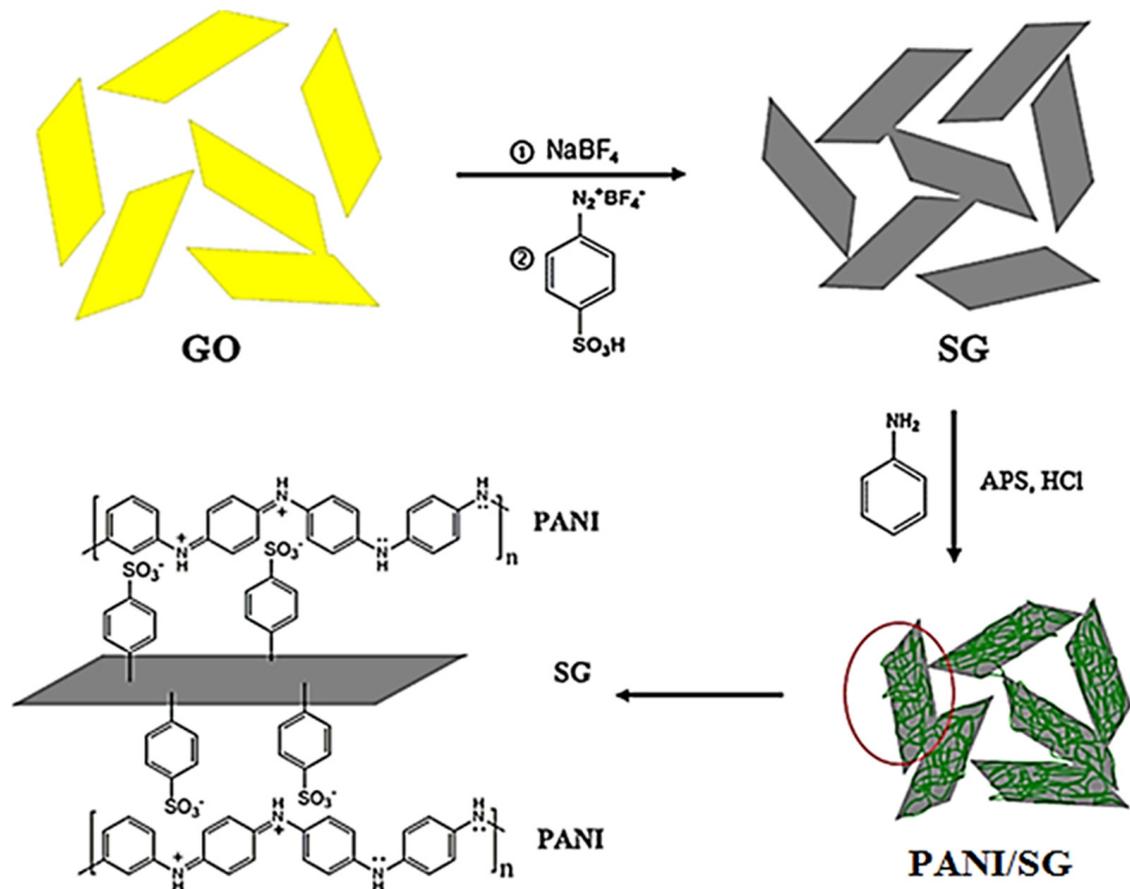
Figure 3. Schematic of SG, PANI/SG nanocomposite, and multilayer film assembled based on multiple interactions between positively charged PANI and negatively charged SG [70]. SG = sulfonated graphene.

Figure 4. (**a**) TEM images of SG, (**b**) PANI and (**c,d**) PANI/SG at 500 nm and 100 nm [70]. PANI = polyaniline; SG = sulfonated graphene.

Polypyrrole is also an important conjugated polymer known for its easy synthesis, solubility, and high electrical conductivity [72]. Graphene nanofillers have been used to form the polypyrrole/graphene nanocomposites [73–75]. GO has been used with the PPy matrix to increase the physical properties of polypyrrole/graphene oxide (PPy/GO) nanocomposites [76,77]. GO dispersion in the PPy matrix has been used to incresae the physical properties [78]. The synergistic effects in PPy/GO have improved the electrochemical properties, electron mobility and thermal transport in the systems. Moreover, GO has been used to increase the mechanical properties and thermal stability of the PPy/GO nanocomposites [79]. The electrochemical polymerization, in situ route, and emulsion polymerization have been used as successful methods to form the PPy/GO nanocomposites [80,81]. Deng et al. [82] used the electrochemical technique to form the PPy/GO nanocomposites. The platinum electrode was used as references and coated with the PPy and PPy/GO nanocomposites. The electrochemical synthesis route for the formation of the nanocomposite is given in Figure 5. The nanocomposite was developed using the electrostatic interactions between the pyrrole cations and negatively charged GO nanosheets. Figure 6 shows the electrochemical impedance spectroscopy (EIS) results for the Pt electrode and the electrodes modified with PPy and PPy/GO with different GO contents.
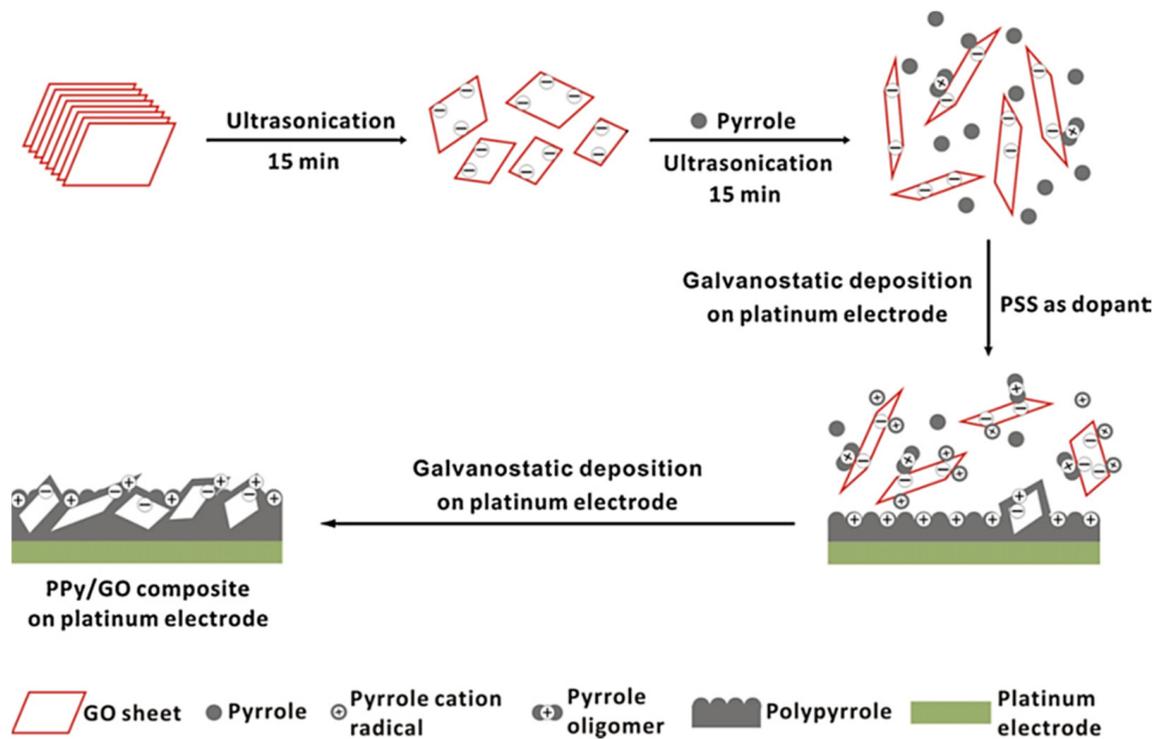
Figure 5. Electrochemical deposition of PPy/GO coating on the Pt electrode site [82]. PPy/GO = polypyrrole/graphene oxide; Pt = platinum.
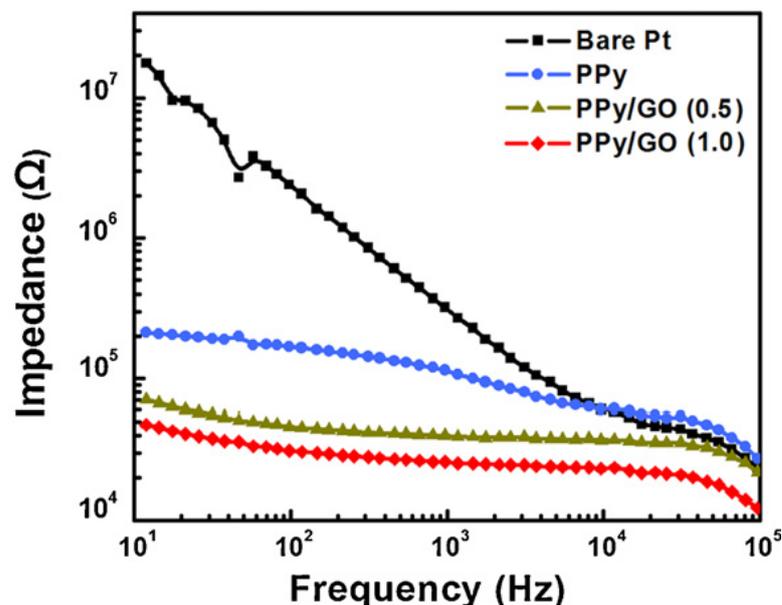
Figure 6. Electrochemical properties of Pt electrode modified with electrochemically deposited PPy, PPy/GO (0.5), and PPy/GO (1.0) coatings [82]. PPy/GO = polypyrrole/graphene oxide; Pt = platinum.

　　The neat Pt electrode had higher impedance than the PPy and nanocomposites in the frequency range of 10–10^5 Hz. The nanofiller loading of 1 wt.% further reduced the impedance, compared with the 0.5 wt.% PPy/GO. The impedance values of the PPy, PPy/GO (0.5 wt.%), and PPy/GO (1.0 wt.%) were 115 kΩ, 40 kΩ, and 26 kΩ, respectively. The difference in the impedance values were attributed to the morphological difference and effect of GO in the nanocomposites.

Polythiophene and its derivatives are important matrices for the nanocomposites [83]. Shamsayei et al. [84] prepared the polythiophene/graphene oxide (PTh/GO) nanocomposites. The electrochemical technique was used for the PTh/GO nanocomposites. The conductivity and morphology properties were studied. Yang et al. [85] proposed the poly(3-hexylthiophene)/reduced modified graphite oxide (P3HT/re-mGO) nanocomposites. In solvent, the neat re-mGO was aggregated, whereas P3HT/re-mGO nanocomposite has fine dispersion [86]. The morphology study by Atomic force microscopy (AFM) reveals the adhering of polymer chains with the GO nanosheets. The polymer was coated on the nanofiller surface (Figure 7).
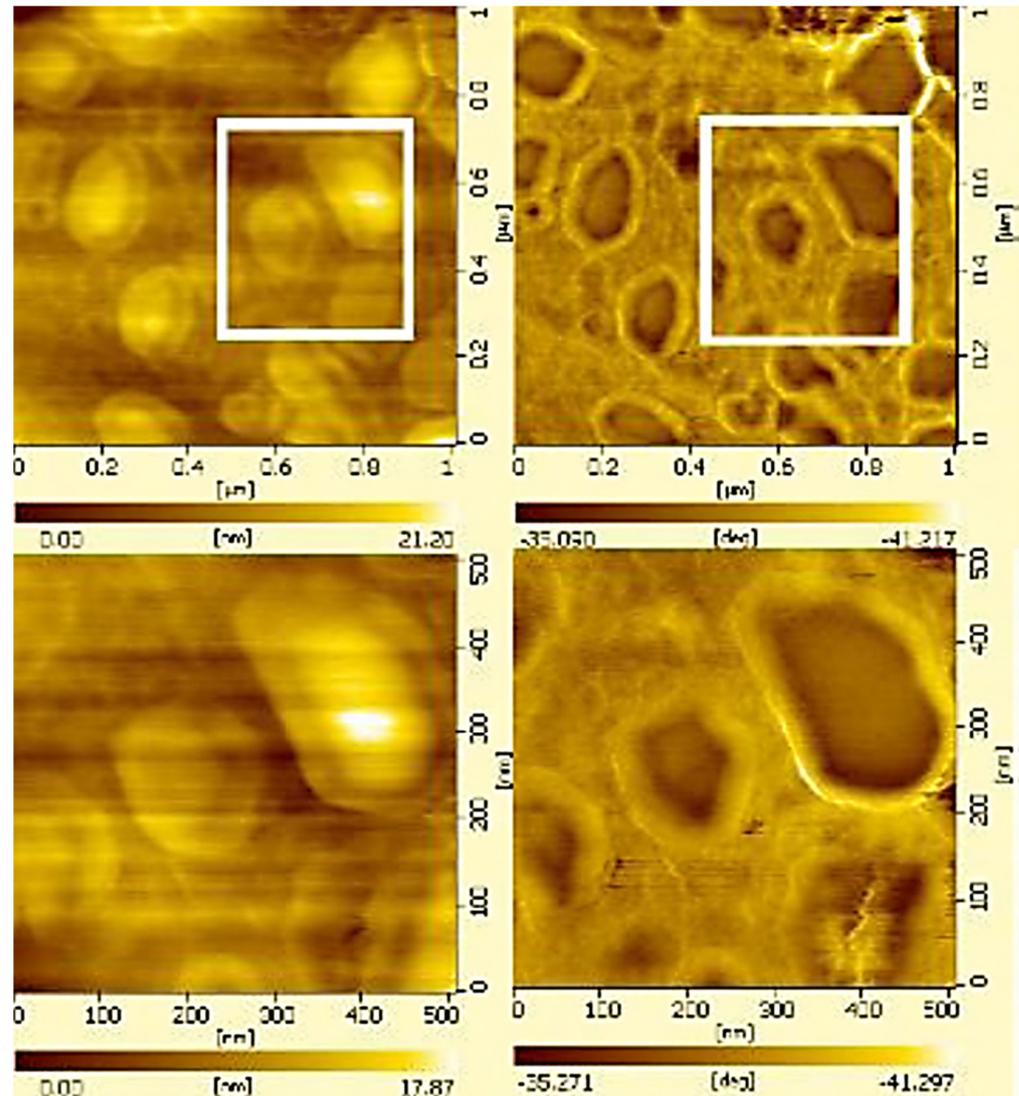
Figure 7. AFM images of P3HT/re-mGO nanocomposites (5 mg/mL) with plan images (**left**) and phase images (**right**) [85]. P3HT/re-mGO = poly(3-hexylthiophene)/reduced modified graphite oxide.

Bora et al. [87] prepared the PTh/GO nanocomposites through the interfacial polymerization. The nanofiller loading improved the electrical conductivity of the nanocomposites to 2.7×10^{-4} S cm^{-1}. The thermogravimetric analysis (TGA) was used to study the thermal stability of the nanocomposites (Figure 8 and Table 1). Incorporating the GO nanofiller in the PTh matrix improved the thermal stability of the polymer matrix. The PTh/GO nanocomposite (3 wt.% nanofiller) revealed increase in the weight retention to 19%, compared with the neat PTh (4%). The weight loss of the PTh matrix occur in the

range 200–300 °C. For the nanocomposites, the degradation starts at higher temperatures of 248–260 °C.
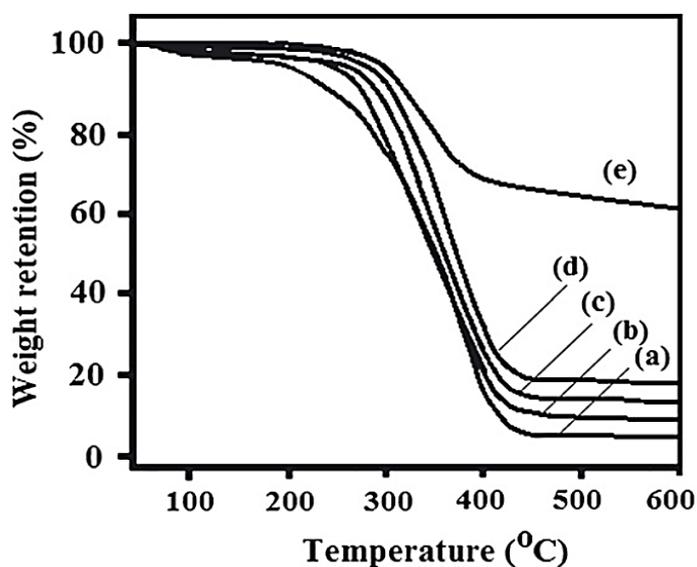
Figure 8. TGA curves: (**a**) PTh; (**b**) PTh/GO 1, (**c**) PTh/GO 2 and (**d**) PTh/GO 3 nanocomposites; (**e**) GO [87].

Table 1. TGA data for PTh, GO and PTh/GO composites [87].

Sample	Weight Loss (%)			Weight Retention at 600 °C (%)
	At 200 °C	At 300 °C	At 500 °C	
PTh	10	15	80	4
PTh/GO 1	8	13	77	11
PTh/GO 2	7	12	73	17
PTh/GO 3	6	10	70	19
GO	1	25	35	60

5. Significance of Conjugated Polymer/Graphene Oxide Nanocomposites

Supercapacitors are efficient energy storage devices. Supercapacitor generally stores energy on the conducting material surface. High performance supercapacitors have been developed using the conducting polymers and nanocomposites [88–90]. Polythiophene based supercapacitors possess high electrical conductivity, charge mobility, chemical stability, and environmental friendliness [91]. The PTh/GO nanocomposites have been applied in the supercapacitors [92,93]. For this purpose, the PTh/GO nanocomposites have been developed through the in situ oxidative polymerization technique [94–96]. Various derivatives of PTh have also been used in supercapacitors. The poly(3,4-ethylenedioxythiophene) (PEDOT)/GO based supercapacitors had high specific capacitance of 201 Fg^{-1} [97–99]. The PEDOT/GO nanocomposite revealed high specific capacitance of 320 Fg^{-1}. The poly(9-butyl-3,6-di (thien-2-yl)-9H-carbazole)/GO showed specific capacitance of 296 Fg^{-1} [100,101]. The interactions between the polythiophene and GO through π-π stacking or non-covalent linking have enhanced the electron mobility, capacitance values, and charge/discharge performance [102–104]. Zhou et al. [105] produced the PPy/GO nanocomposite using the electrochemical method. The electrochemical co-deposition of PPy/GO nanocomposite on the fluorine-doped tin oxide (FTO) substrate is given in Figure 9a,b. Figure 9c depicts the formation of layered supercapacitor device with PPy/GO sandwiched between the FTO substrates. Fan et al. [70] reported the supercapacitor based on the PANI/SG derived from GO. Figure 10 shows the change in the specific capacitance with current density of the

nanocomposites. The specific capacitance of the SG, PANI, and PANI/SG was decreased with the current density from 0.5–4 Ag^{-1}. The highest specific capacitance was found for the PANI/SG nanocomposites in the range of 410–478 Fg^{-1}. The high capacitance of the PANI/SG nanocomposites was due to the improved interactions between the conjugated polymer and nanofillers and electron transportation.
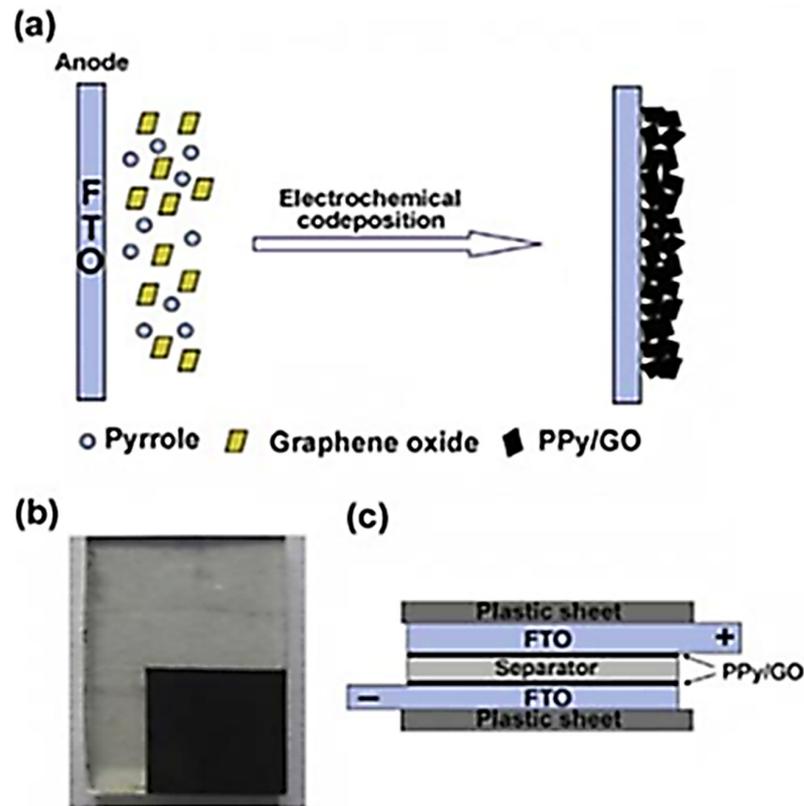
Figure 9. (**a**) Schematic for electrochemical co-deposition of PPy/GO nanocomposites; (**b**) real photo of PPy/GO-deposited FTO conducting glass; (**c**) construction of supercapacitor cell [105]. PPy/GO = polypyrrole/graphene oxide; FTO = Fluorine-doped tin oxide.
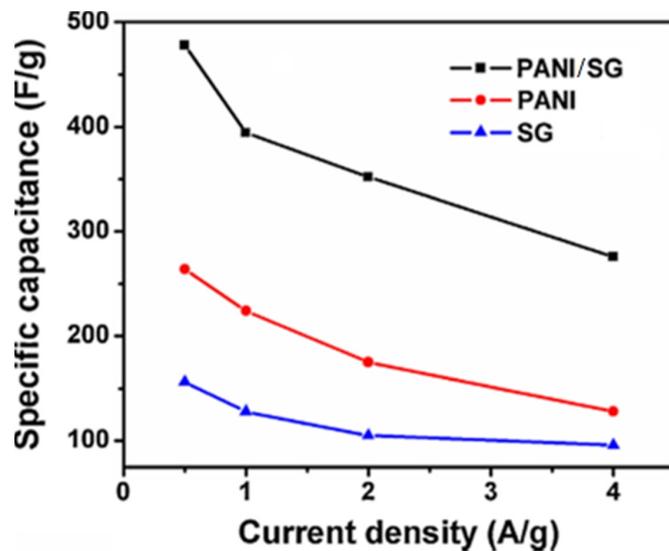

Figure 10. Variation of the specific capacitance with current density for SG paper, PANI powder, and PANI/SG paper [70]. PANI = polyaniline; SG = sulfonated graphene.

The increasing demand of multi-purpose energy devices have developed the advanced solar cells based on the conjugated polymers [106]. Blending of the conducting polymers with nanocarbons (graphene, GO, carbon nanotube) have improved the power conversion efficiencies of the solar cells [107]. The nanocarbons such as GO may act as electron accepting materials in the solar cell nanocomposites [108]. GO own large surface area and electron transport pathways for high charge/electron conductivity [109]. Consequently, the solar cells have been integrated with the PTh/GO based nanomaterials [110–112]. Agbolaghi et al. [113] formed the polyaniline-*grafted* reduced graphene oxide (PANI-*g*-rGO) nanocomposite using the in situ oxidative polymerization method. The PANI-*g*-rGO improved the solar cell efficiency to 7%. Stylianakis et al. [114] established the poly(3-hexylthiophene)/GO nanocomposites for the bulk heterojunction solar cell. The GO act as electron acceptor and poly(3-hexylthiophene) function as electron donor [115–117]. Moreover, GO was finely dispersed in the polymeric matrix and developed percolation network for the efficient electron transportation [118,119].

The electromagnetic radiation emission can daunt the performance of electronic devices and systems [120]. Consequently, the electromagnetic radiation emission has been deliberated as the environmental pollution. Incidentally, the polymeric nanocomposites have been applied to block the electromagnetic waves [121]. Graphene oxide has been used as an significant polymeric nanofiller in conducting polymers for EMI shielding [122]. Research attempts have been performed on the polyaniline and graphene oxide based nanocomposites for EMI shielding [123]. The EMI shielding efficiency of the polymer/graphene oxide has been found up to 40 dB. The polyaniline and graphene decorated with silver nanoparticles had EMI shielding efficiency of 29.33 dB [124]. In addition to polyaniline, poly(3,4-ethylenedioxythiophene) and GO based nanocomposites have also been reported [125–127]. It has been observed that the fine electrical conductivity is needed for the high EMI shielding efficiency. Subsequently, the conducting nanocomposites have ability to overcome the problem of electromagnetic radiation and low absorption [128]. The high EMI shielding performance of the polymer/graphene oxide nanocomposites can be useful for the electronic and optoelectronic devices [129].

Corrosion is a severe technical issue for the metal industries [130,131]. Various methods have been developed to prevent the erosion of metal based materials such as protective coatings, corrosion inhibitors, and better-quality anti-corrosion methods [132–134]. The anti-corrosion performance of polymers have been enhanced using the nanocarbon nanofillers such as graphene and GO [135–137]. The thin layers of GO have been studied as anti-rusting coatings [138]. The GO coatings have been used to inhibit the erosion of copper and nickel [139,140]. The PANI/GO nanofiber based coatings have been used for the corrosion resistance [141]. The PTh/GO nanocomposites have been used for the anti-rusting coatings [142]. Further investigations on the anti-corrosion and electrical conductivity properties of the conjugated polymers and graphene oxide based nanocomposites may expand this field for future advancements in this field [143].

6. Future and Summary

Graphene oxide-based conjugated polymer nanocomposites are still at the initial stages of development and there are growing interests in overcoming the challenges and applications of these materials. The attainment of ideal dispersion state of the GO nanoparticles and property enhancement are the challenging factors. The homogeneous nanofiller dispersion may offer large interfacial area between the matrix and the GO reinforcement. Consequently, the reinforcing effect of the nanofiller involve several features such as aspect ratio of GO, nanofiller orientation, polymer matrix type, polymer/nanofiller ratio, and nanofiller dispersion in the matrix. The challenging problem which need to be addressed is the large scale production of the uniformly dispersed conjugated polymer/GO nanocomposites. Primarily, fine GO dispersion and interaction in matrix/nanofiller is a major challenge.

According to above literature, conjugated polymer base materials have been discovered as a broad research field. Polyaniline, polypyrrole, polythiophene, and PTh derivatives have been polymerized using various polymerization techniques and filled with the graphene based nanofillers. It has been observed that the structure and morphology of the conducting polymer/GO depend on the fabrication technique used and the synthesis conditions. Here, the challenging factors are the identification of efficient synthesis method, optimum reaction conditions, and the choice of matrix and functional GO. In situ method has been perceived as the most widely used method in this regard. Moreover, the interactions in the conducting polymer/GO nanostructures may enhance the electrical conductivity and essential features of these materials. The structure-property relationships are predicted to establish the advanced applications of the conjugated polymer/GO nanocomposites. Supercapacitor performance can be enhanced by using the conjugated polymer/GO and doped polymer matrices. The photovoltaic effect of the conducting polymer/GO can also be improved using the doping process. The anti-corrosion performance of the conjugated polymer/GO coatings can also be upgraded using design improvement and doping techniques. In future, the conducting polymer/GO can be significantly used for the microelectronics applications. Progress in the conjugated polymer/GO nanocomposites may also lead to fine digital integrated circuits. The biomedical field is also awaiting the use of conjugated polymer/GO nanocomposites. For future advancements, various features of conjugated polymer/GO need to be considered including the modification of the conjugated main chain, functionalities of GO, altered synthesis strategies, mechanism of conjugated polymer/GO interaction, and structure-property relationship.

Briefly speaking, this review states essentials aspects of the conjugated polymer/GO nanocomposites. Conjugated polymer/GO nanomaterials have enhanced the morphological, electrical, thermal, and mechanical features. The physical properties and applications of the conjugated polymer/GO nanocomposites have been reviewed. Progress in the conjugated polymer/GO nanocomposites revealed solicitations in supercapacitors, solar cells, and corrosion protective coatings.

Conflicts of Interest: The author declares no conflict of interest.

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