



Review

Cutting-Edge Graphene Nanocomposites with Polythiophene—Design, Features and Forefront Potential

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Abstract: Among conducting polymers, polythiophene has gained an important stance due to its remarkable physical features. Graphene is a unique, two-dimensional, nanocarbon nanomaterial. As in other polymers, graphene has been reinforced in polythiophene to form advanced nanocomposites. This comprehensive review covers the design, essential features, and methodological potential of significant polythiophene and graphene-derived nanocomposites. In this context, various facile approaches, such as in situ processing, the solution method, and analogous simplistic means, have been applied. Consequently, polythiophene/graphene nanocomposites have been investigated for their notable electron conductivity, heat conduction, mechanical robustness, morphological profile, and other outstanding properties. Studies have revealed that graphene dispersion and interactions with the polythiophene matrix are responsible for enhancing the overall characteristics of nanocomposites. Fine graphene nanoparticle dispersal and linking with the matrix have led to several indispensable technical applications of these nanocomposites, such as supercapacitors, solar cells, sensors, and related devices. Further research on graphene nanocomposites with polythiophene may lead to remarkable achievements for advanced engineering and device-related materials.

Keywords: polythiophene; graphene; nanocomposite; dispersion; interaction; supercapacitor; solar cell; sensor



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1. Introduction

Among carbon nanofillers, graphene has attained an essential research spot by effectively enhancing polymer characteristics such as the microstructure, electron transference, mechanical robustness, heat stability, and several other features [1,2]. For conducting or conjugated polymers, graphene has been applied as an effective nanofiller [3]. Polythiophene belongs to the family of conductive polymers, with advanced conductivity, electrochemical, and physical properties [4]. Like other conducting polymers, polythiophene has been effectively applied to develop nanocomposites [5]. The resulting graphene-filled polythiophene nanocomposites demonstrate fine morphology, electron conduction, supercapacitance, sensing, strength, and heat-resistance properties for use in supercapacitors, solar cells, sensing devices, and related electronics and energy applications [6–8]. In this context, the dispersion and interactions of graphene nanoparticles in the polythiophene matrix has led to the development of a compatible interface and electron conduction pathways for fine charge mobility [9]. In addition, the interfacially interacting polythiophene/graphene

nanostructure is responsible for the overall improved characteristics and technical performance [10]. At this juncture, polythiophene derivatives have been found to be capable of developing fine connections with graphene nanosheets [11].

This innovative overview offers an outlook on the design, features, and potential of technically important polythiophene/graphene nanocomposites. The incorporation of graphene in polythiophene matrices has produced high-performance nanocomposites. The characteristics and performance of these nanocomposites depend upon the compatibility between matrix–nanofiller interactions and consistent nanoparticle dispersal. High-competence polythiophene/graphene nanocomposites have been observed to be functional in important technical fields. To the best of our knowledge, this review is ground-breaking by revealing the technical progression in the field of polythiophene and graphene-derived nanomaterials. Consequently, polythiophene and derivative matrices filled with graphene exhibit notable characteristics and provide high-efficiency nanomaterials. These innovative nanocomposites are anticipated for progressive applications such as supercapacitors, solar cells, and sensing devices.

2. Polythiophene and Derived Forms

Polythiophene is a conductive polymer formed by the polymerization of thiophene monomers and its backbone has repeating thiophene units [12]. Thiophene is actually a heterocyclic molecule with a sulfur atom. Polythiophene is a semiconducting polymer with a low band gap [13]. The remarkable properties of polythiophene include high electron transference, optical properties, thermal stability, and environmental stability [14]. Due to its π -conjugated structure, polythiophene has fine electrical conductivity behavior [15]. The electron transport mechanism has been found to depend upon the passage of polarons and bipolarons through the main chain [16]. Furthermore, electrical conduction through the polythiophene backbone can be enhanced through oxidation or reduction reactions. In this way, polythiophene may have high electric conduction around $>100 \text{ Scm}^{-1}$ [17]. Usually, organic solvents are used for the processing of polythiophene. A number of chemical and electrochemical methods have been used to form polythiophene [18]. Electrochemically polymerized polythiophene usually reveals better electrical conductivity relative to the polymers synthesized via other methods. Here, the oxidation process is usually supported due to the presence of an electron lone pair on the sulfur of the thiophene ring. Processes like doping or modification of thiophene moieties can also enhance the desired properties of this polymer [19,20]. The potential utility of polythiophene has been increased through the development of numerous derived forms [21–23]. Table 1 demonstrates some important polythiophene derivatives, such as poly(3-hexylthiophene), poly(3-octylthiophene), poly(3-octadecylthiophene), poly(3-dodecylthiophene), and more derived forms. Polythiophene along with its derived forms have been applied in solar cells, batteries, electronics, etc. [24]. In addition, polythiophene and its derivatives have been applied in nanocomposite formation [25]. Especially, using carbon nanofillers may enhance the π -stacking interactions with the polythiophene backbone and facilitate better dispersion, physical properties, and applications [26].

Table 1. Derivatives of polythiophene.

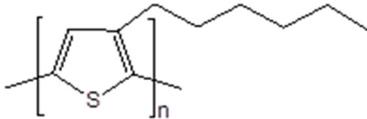
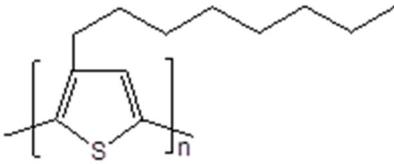
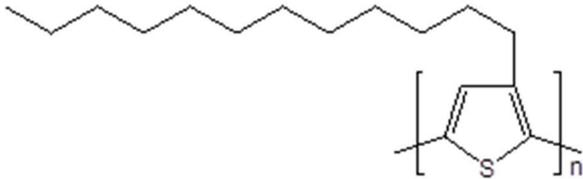
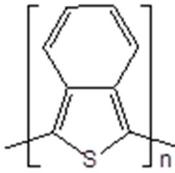
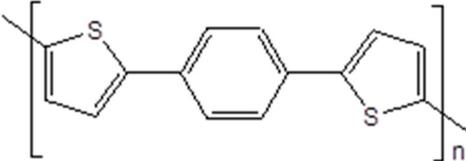
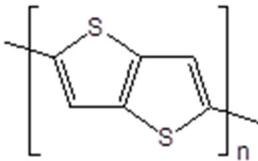
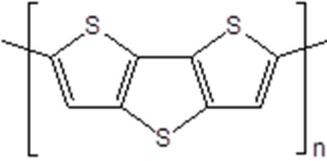
Derivative	Structure
Poly(3-hexylthiophene)	

Table 1. Cont.

Derivative	Structure
Poly(3-octylthiophene)	
Poly(3-dodecylthiophene)	
Poly(isothianaphthene)	
Poly(1,4-di-(2-thienyl)benzene)	
Poly(thieno [3,2-b]thiophene)	
Poly(dithieno [3,2-b:2',3'-d]thiophene)	

3. Graphene Dispersal Effects in Polythiophene

Graphene is a unique two-dimensional nanocarbon nanostructure made up of sp^2 hybridized carbon atoms arranged in a hexagonal lattice structure [27]. It comprises a one atom thick nanocarbon nanosheet. Due to its remarkable nanostructure, graphene has high electron conduction, thermal stability, and mechanical robustness properties [28]. Synthesis methods used for graphene include the mechanical exfoliation approach [29], chemical vapor deposition [30], thermal decomposition [31], and chemical or organic synthesis methods [32,33]. For large-scale production, exfoliation techniques have been preferred for the formation of graphene [34]. Applications of graphene have been observed in aerospace, automotive, electronics, biomedical, and countless other fields [35–37]. Graphene oxide is a well-known derivative of graphene [38]. Polythiophene solubility has been desirable for the high performance of the nanocomposite form [39,40]. Subsequently, including graphene or other nanofillers in polythiophene has resulted in improved conductivity, optical, heat or mechanical constancy, and overall physical profiles [41–43]. Almost all of the properties of polythiophene nanocomposites depend on nanoparticle dispersion in the matrix. Due to van der Waals interactions, graphene nanosheets possess an aggregation tendency upon

dispersion in solvents [44,45]. For polythiophene nanocomposite processing, dispersants (surfactants, ionic liquids) have been used for fine dispersion of graphene into the polymer matrix [46–48]. Iguchi and co-workers [49] developed a poly(3-hexylthiophene)- and graphene-derived nanocomposite. For in situ polymerization, graphene was dispersed in a mixture of toluene and *N*-methylpyrrolidone solvents. Consequently, the graphene dispersion behaviour was studied in the poly(3-hexylthiophene) matrix (Figure 1).

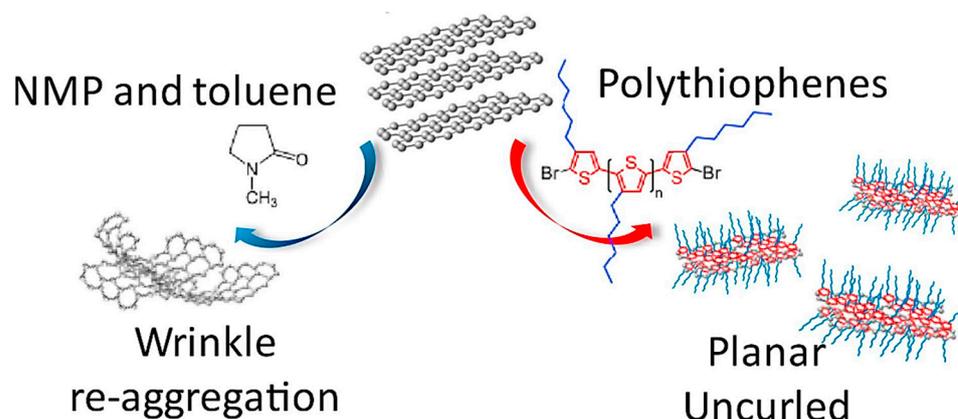


Figure 1. Graphene dispersion and interactions with poly(3-hexylthiophene) [49]. Reproduced with permission from Elsevier.

The wrinkling of graphene nanosheets and poor dispersion in the polymer result in poor physical properties and technical performance of polymer/graphene nanocomposites [50]. The formation of an interconnecting network by the graphene nanofiller facilitates electron transportation through nanocomposites [51]. Husain and co-workers [52] presented an important study on the π -conjugated system of polythiophene and graphene-derived nanocomposites. The combination of polythiophene and graphene may develop a co-ordinated conjugated system for electron conduction [53,54]. Figure 2 reveals a clear picture of π - π interactions between the polythiophene backbone and graphene nanostructure. Accordingly, the lone pair of electrons on the sulfur of thiophene can interact with the π electrons of the graphene nanosheet. Consequently, the conjugated system endorses rapid electron or charge transportation through the nanocomposite nanostructure [55,56]. Here, adding 15 wt.% graphene nanofiller led to superior electrical conductivity of $\sim 0.7 \text{ S cm}^{-1}$ due to matrix–nanofiller interactions in ethanol solvent [57]. Moreover, in this solvent–nanocomposite system, the lone pair of electrons on the ethanol oxygen interacted with the nanomaterial to develop better electron passage through the system [58].

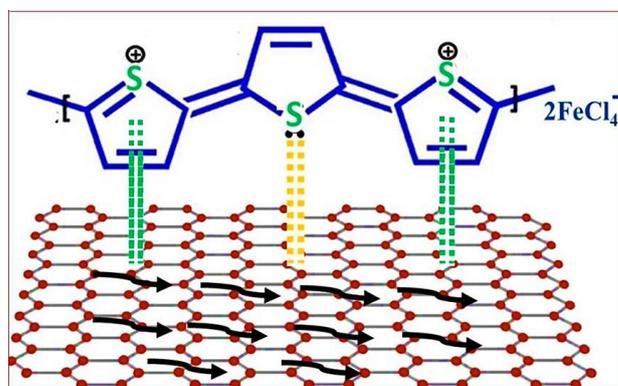


Figure 2. Interactions between polythiophene and graphene nanosheets in the nanocomposite leading to electron conducting pathways responsible for improved electrical conductivity [52]. Reproduced with permission from Elsevier.

To attain better dispersion properties, graphene has been modified with various surface functionalities [59–61]. In this way, the polar groups in organic solvents (dimethyl sulfoxide dimethylformamide, 1-methyl-2-pyrrolidinone, etc.) can better interact with the modified graphene nanostructures [62–64]. Consequently, the modified graphene forms may develop covalent or non-covalent interactions for better dispersion in solvents [65,66]. The poly(3-hexylthiophene) solubility and modified graphene dispersion in chloroform were analyzed [67]. Thus, the modified graphene revealed better dispersion properties in polar solvents than the pristine graphene [68,69]. Finely scattered graphene in polythiophene leads to high-performance nanocomposites.

Aspects such as graphene content, dispersion in the matrix, and matrix–nanofiller interactions have been found to be important for the physical features of polythiophene/graphene nanocomposites, including the microstructure, conductivity, heat conduction, mechanical properties, and other characteristics. Graphene-derived polythiophene nanocomposites face key challenges regarding nanoparticle dispersion. Particularly, graphene dispersion has been a problem during large-scale production of nanocomposite materials. The structure–property relationship in polythiophene/graphene nanomaterials is important for high-efficiency applications of nanocomposite nanofibers. Li et al. [70] explored the effect of dispersion of modified graphene on important properties. The modified graphene developed a heat transfer interface with the poly(3-hexylthiophene) matrix due to π - π interactions. The poly(3-hexylthiophene)/graphene nanocomposite revealed an in-plane thermal conductivity of $4.17 \text{ W m}^{-1} \text{ K}^{-1}$, which was higher than that of the pristine polymer matrix. The effect was observed due to better dispersal and interfacial properties of the matrix and nanofiller. Kausar et al. [71] formed polythiophene and graphene-derived nanomaterial. Owing to better graphene dispersion, a percolation threshold was attained at 1.5 wt.% nanofiller content, with a high electrical conductivity of $1.8 \times 10^{-3} \text{ S cm}^{-1}$. The resulting nanocomposite also showed a high thermal conductivity of 1.22 W/mK due to the development of a thermal interface. Sharif et al. [8] developed a polythiophene and graphene oxide-derived nanocomposite through in situ polymerization of the thiophene monomer in the presence of nanocarbon. The graphene oxide dispersion effect was observed in the development of conducting pathways with the polymer matrix. Consequently, the electrical conductivity of the polythiophene/graphene nanocomposite was found to be higher ($2.5 \times 10^{-8} \text{ S cm}^{-1}$) than that of neat polythiophene ($1.3 \times 10^{-7} \text{ S cm}^{-1}$). In addition to the electrical conductivity, the thermal stability of this nanocomposite was found to be enhanced due to graphene dispersion and interactions with the polymer. A weight retention of 15–22% was observed at $600 \text{ }^\circ\text{C}$ due to better thermal stability properties.

4. Polythiophene and Graphene-Derived Nanocomposites

Both graphene and graphene oxide have been used as nanofillers for polymers [72]. Graphene oxide is actually an oxidized form of graphene and it has several oxygen functional groups on the graphene surface. Graphene oxide has been frequently developed using a facile Hummer's method with a graphite precursor [73]. However, several other physical or chemical exfoliation techniques have also been reported [74]. However, due to the presence of functional groups, graphene oxide may have better dispersion and interactions with polymers, leading to enhanced compatibility properties [75–77].

Polythiophene is an essential conjugated polymer and its backbone has the fine capability of electron transfer and charge mobility [78]. Consequently, polythiophene has advantages of facile processing, high electron conduction, and the formation of π ring stacking, dipole-dipole interactions, and hydrogen bonding with nanofillers [79]. Here, graphene has been used as an efficient nanofiller for polythiophene and its derivatives. Accordingly, the inclusion of graphene has been found to improve the optical, electrical, electrochemical, and several other physical characteristics of polythiophene nanocomposites [80]. Nayebi and co-researchers [81] prepared a polythiophene and graphene-based nanocomposite. The structure and mechanical stability features of the polythiophene/graphene nanocomposite were investigated. Simulation studies were performed on the polythiophene/graphene

nanocomposite. Inherent defects were observed on the graphene surface, which were increased upon doping with the polymer. More atoms were removed from the graphene nanosheet due to the doping effect of the polythiophene chains. The stress–strain analysis of the nanocomposite revealed that the nanofiller loading increased the tensile stress and modulus of the nanocomposite up to a 20 wt.% graphene loading level. The enhanced mechanical features were attributed to the synergistic effects of the polythiophene and graphene in the nanocomposite form.

Hsieh and co-workers [82] produced a polythiophene and graphene-based nanocomposite. The nanomaterial revealed fine performance for field-effect transistors [83]. Liscio et al. [84] used poly(3-hexylthiophene), a polythiophene derivative, combined with reduced graphene oxide nanofiller to form a nanocomposite. The material was applied in field-effect transistors. Gemayel and co-workers [85] also applied a poly(3-hexylthiophene) derivative with graphene nanoribbon nanofiller. The poly(3-hexylthiophene)/graphene nanoribbon nanocomposite had semiconducting properties and fine potential for field-effect transistors [86]. Alvi et al. [87] reinforced the polythiophene and poly(3,4-ethylenedioxythiophene) matrices with the graphene nanofiller. The in situ polymerization method was used to form the polythiophene/graphene and poly(3,4-ethylenedioxythiophene)/graphene nanocomposite [88]. The nanocomposite was effectively used for a supercapacitor electrode. Moreover, poly(3-phenylthiophene) is a significant phenyl derivative of polythiophene [89]. Poly(3-phenylthiophene) can also establish secondary linkages with the graphene nanofiller. Furthermore, a poly(9-butyl-3,6-di(thien-2-yl)-9H-carbazole) and graphene oxide nanocomposite has been prepared using in situ polymerization [90]. The nanomaterial had a sufficiently high specific capacitance of 296 F g^{-1} for supercapacitor application. In addition, an important use of polythiophene is as a template for graphene synthesis [91]. Thus, graphene developed using the polythiophene template has been applied in electronics [92,93].

Shamsayei and co-researchers [94] developed a polythiophene and graphene oxide-derived nanocomposite. The electro-deposition method was used to deposit the nanocomposite material over the surface of the electrochemical electrode. The nanomodified electrode was used for solid-phase microextraction application. Molaei [95] reported the simultaneous copolymerization of pyrrole and thiophene monomers over the modified graphene oxide surface. In this context, SiO_2 nanoparticle-coated magnetic graphene oxide was prepared. The copolymerization of pyrrole and thiophene monomers on the nanofiller surface led to the formation of a polypyrrole/polythiophene and magnetic graphene oxide/ SiO_2 -derived nanocomposite. The unique nanomaterial structure was investigated for the solid-phase extraction of chromium, cadmium, lead, zinc, copper, and other toxic metals from water. Wang et al. [96] doped a reduced graphene oxide surface with thiophene monomer and sulfur. The polythiophene/sulfur/graphene oxide nanocomposite revealed enhanced electrical conductivity due to sulfur doping. Consequently, the nanocomposite was found to be efficient for dye-sensitized solar cells. In addition, the sulfur-doped nanomaterial was found to be more effective than the nitrogen-doped nanocomposite in enhancing conductivity and solar cell performance [97].

Bora et al. [98] developed a polythiophene and graphene oxide-derived nanocomposite using the interfacial polymerization technique. The thermal stability of neat polythiophene and the polythiophene and graphene oxide-based nanocomposite was examined. The thermogravimetric studies showed maximum thermal decomposition temperatures of $240 \text{ }^\circ\text{C}$ for pristine polythiophene and $248\text{--}260 \text{ }^\circ\text{C}$ for the polythiophene/graphene oxide nanocomposite (1–3 wt.%). On the other hand, the initial decomposition temperatures for pristine polythiophene and the nanocomposite were observed around 200 and $300 \text{ }^\circ\text{C}$, respectively. A higher decomposition temperature for the polythiophene/graphene oxide nanocomposite was observed due to nanofiller reinforcement and the rigidity of the polythiophene chains towards heat exposure. Consequently, this effect was attributed to mutual matrix–nanofiller interactions in the nanocomposite. The electrical conductivity and resistivity behaviors of the unfilled polythiophene and polythiophene/graphene oxide nanocomposite were also analyzed (Table 2). Here, the resistivity value of neat

polythiophene was high (i.e., 4.9 Ωcm), with a corresponding electron conductivity of $5.8 \times 10^{-5} \text{ S cm}^{-1}$. The addition of graphene oxide nanoparticles considerably decreased the resistivity value to 1.97 Ωcm (3 wt.% nanoparticles). Similarly, the electrical conductivity value for the 3 wt.% nanocomposite was enhanced to $2.7 \times 10^{-4} \text{ S cm}^{-1}$, relative to neat polythiophene and 1 and 2 wt.% nanoparticle addition. Increased electrical conductivity and decreased resistivity were attributed to the formation of π - π stacking interactions between the polythiophene and graphene nanosheets. At 3 wt.% nanofiller inclusion, the graphene nanoparticles developed a percolation threshold and interconnecting networks to facilitate electron transportation through the nanocomposite system. Optimum electrical conductivity values also led to a reasonable specific capacitance of 99 F g^{-1} .

Table 2. Electrical conductivity and resistivity of pristine polythiophene and polythiophene nanocomposites filled with graphene oxide from 1 to 3 wt.% loading [98]. Reproduced with permission from Wiley.

Sample	Resistivity ($\Omega \text{ cm}$)	Conductivity (S cm^{-1})
Polythiophene	4.90	5.8×10^{-5}
Polythiophene with graphene oxide 1 wt.%	2.80	0.8×10^{-4}
Polythiophene with graphene oxide 2 wt.%	2.36	1.2×10^{-4}
Polythiophene with graphene oxide 3 wt.%	1.97	2.7×10^{-4}

Li et al. [99] formed a graphene oxide nanosheet-filled nanocomposite with an important derivative of polythiophene, i.e., poly [3-(2-(2-(2-(diethanolamino)ethoxy)ethoxy)ethoxy)ethoxy)thiophene]. Covalent grafting was observed between the polymer matrix and graphene oxide nanosheets. Grafting occurred due to the modification of graphene oxide to acyl chloride functional graphene oxide using thionyl chloride. Then, the esterification reaction was observed between the acyl chloride functionalities of the modified graphene oxide and the hydroxy groups of the polythiophene derivative. Figure 3 shows a schematic for the formation of the poly [3-(2-(2-(2-(2-(diethanolamino)ethoxy)ethoxy)ethoxy)ethoxy)ethoxy)thiophene]-graft-graphene oxide nanocomposite. Transmission electron microscopy (TEM) was applied to study the morphology of the nanocomposite. Figure 4A,B shows the wrinkled morphology of the nanocomposite in micrographs. Higher magnification micrographs depict the development of a well-defined interface in the polymer-grafted nanofiller (Figure 4C,D). At the interface, poly [3-(2-(2-(2-(2-(diethanolamino)ethoxy)ethoxy)ethoxy)ethoxy)ethoxy)thiophene] developed a layer that was a few nanometres thick. The polymer layer was clearly distinguished from the crystalline graphene nanosheet surface. Due to covalent grafting, better graphene dispersion and less nanosheet aggregation were observed.

In addition, polythiophene has been combined with functional graphene or functional graphene oxide nanosheets to form nanocomposites [100]. In this regard, iron oxide (Fe_3O_4) nanoparticles have been decorated on graphene nanosheets using the solvothermal technique [101]. Mehdinia et al. [102] filled modified graphene and Fe_3O_4 -based nanoparticles in a polythiophene matrix. Primarily, Hummer's method was applied for the development of graphene oxide. Here, the in situ polymerization technique was used for the formation of the polythiophene and graphene/ Fe_3O_4 nanocomposite. TEM studies were carried out for the morphological investigation of the graphene/ Fe_3O_4 and polythiophene/graphene/ Fe_3O_4 nanocomposite. TEM micrographs of the graphene/ Fe_3O_4 nanofiller showed graphene nanosheets with finely decorated Fe_3O_4 nanoparticles. In contrast, the polythiophene/graphene/ Fe_3O_4 nanocomposite revealed a polymer-coated nanocomposite nanoparticle morphology. Hence, simple synthesis routes were adopted for the development of the graphene/ Fe_3O_4 and polythiophene/graphene/ Fe_3O_4 nanocomposite. The graphene/ Fe_3O_4 and polythiophene/graphene/ Fe_3O_4 nanomaterial were tested for solid-phase extraction of polycyclic aromatic hydrocarbons. The nanomaterial

revealed a detection limit of $0.009\text{--}0.020\ \mu\text{g L}^{-1}$, showing fine extraction properties for polycyclic aromatic hydrocarbons.

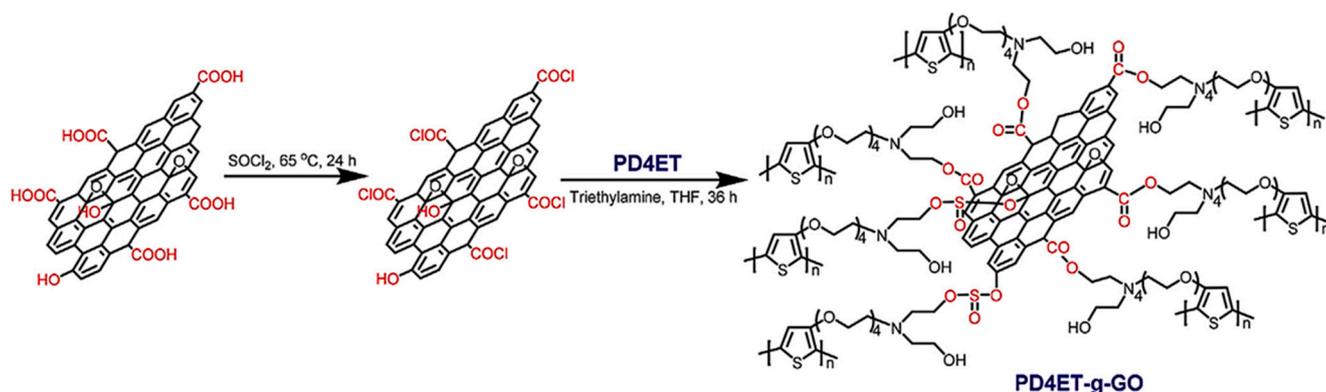


Figure 3. Schematic of covalent grafting of PD4ET side chains to GO nanosheets for the formation of PD4ET-g-GO nanocomposite [99]. PD4ET = poly [3-(2-(2-(2-(2-(diethanolamino)ethoxy)ethoxy)ethoxy)ethoxy)thiophene]; GO = graphene oxide; PD4ET-g-GO = poly [3-(2-(2-(2-(2-(diethanolamino)ethoxy)ethoxy)ethoxy)ethoxy)thiophene]-grafted-graphene oxide. Reproduced with permission from Elsevier.

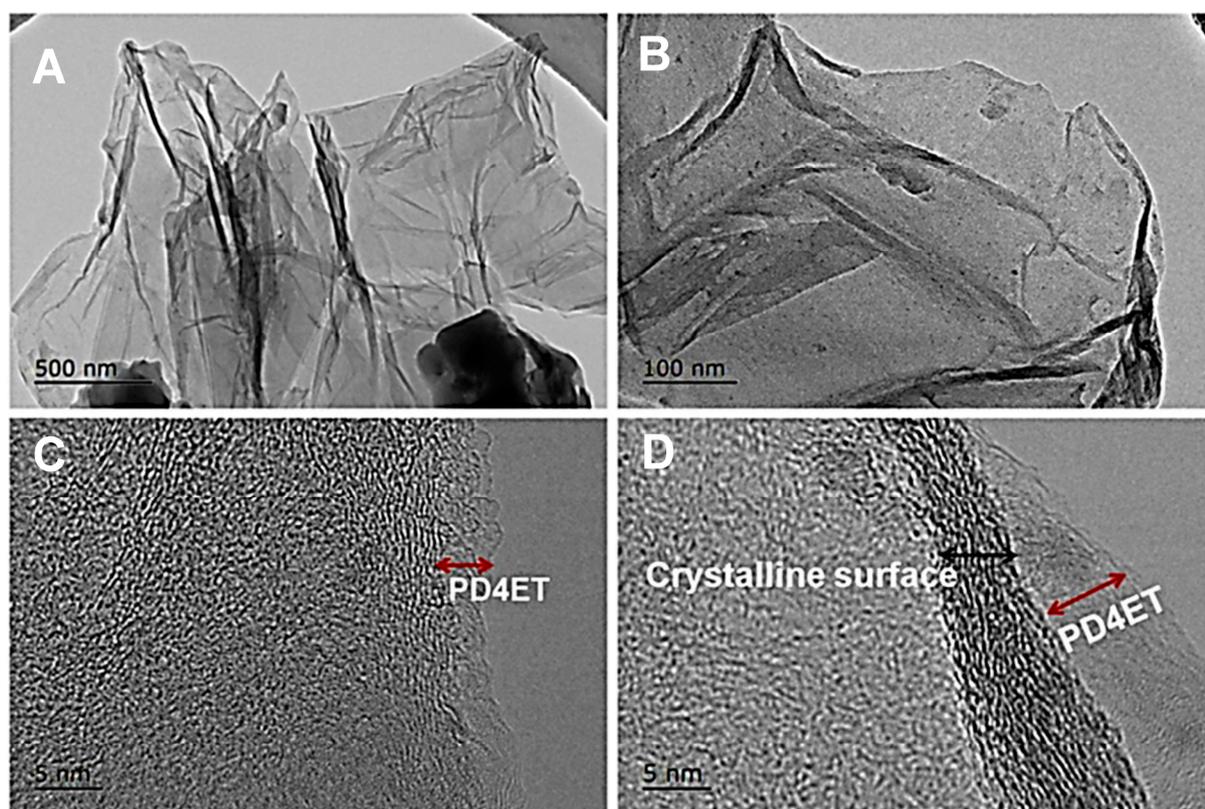


Figure 4. Characteristic TEM images of (A,B) single-layer structure of PD4ET-g-GO with lateral dimensions; and (C,D) showing an interface between PD4ET and GO in the PD4ET-g-GO nanocomposite at magnifications [99]. PD4ET = poly [3-(2-(2-(2-(2-(diethanolamino)ethoxy)ethoxy)ethoxy)ethoxy)thiophene]; GO = graphene oxide; PD4ET-g-GO = poly [3-(2-(2-(2-(2-(diethanolamino)ethoxy)ethoxy)ethoxy)ethoxy)thiophene]-graft-graphene oxide. Reproduced with permission from Elsevier.

Poly(3,4-ethylenedioxythiophene) (PEDOT) is an important derivative of graphene [103]. Inclusion of graphene in PEDOT has led to enhanced mechanical, conducting, sensing, and other physical properties [104,105]. Mainly, PEDOT and graphene-derived nanocomposites are synthesized using in situ polymerization, the blending technique, vapor phase polymeriza-

tion, etc. [106,107]. Tung et al. [108] fabricated a PEDOT and reduced graphene oxide-based nanocomposite for a chemical sensor application. The sensor showed high selectivity, sensitivity, and rapid response towards trace analyte vapors. Tung et al. [109] developed a poly(3,4-ethylenedioxythiophene) and N-doped reduced graphene oxide-derived nanocomposite for a sensing application. The interfacial interactions and synergistic effects between the poly(3,4-ethylenedioxythiophene) and N-doped reduced graphene oxide led to superior properties. The nanocomposite had high conductivity and low sheet resistance ($0.56 \Omega/\text{square}$) properties. The mechanical resistance of the nanocomposite was also maintained even after 1000 bending cycles. Fine electrical, mechanical, and sensing performances have been suggested to be useful for advanced, non-metallic, flexible electronic devices.

In addition, the different morphologies of polythiophene and graphene have been applied in nanocomposite forms such as nanoparticles, nanofibrils, nanowires, etc. [110–112]. These morphologies have led to high surface areas, improved properties, and performance benefits of the resulting nanocomposites. Tran et al. [113] developed nanoparticles of poly(3-hexylthiophene-2,5-diyl) using the mini-emulsion technique and formed nanocomposites with graphene. The development of an interface between the poly(3-hexylthiophene-2,5-diyl) nanoparticles and graphene led to intermolecular charge transfer to accelerate the oxygen reduction reaction and the nanocomposite was employed as a low-cost electrocatalyst in sustainable fuel cells. Tran et al. [114] developed a poly(3,4-ethylenedioxythiophene) polystyrene sulfonate nanofibrils and graphene-based nanocomposite. The nanomaterial was formed using the direct ink writing technique and possessed high electrical conductivity of $\sim 630 \text{ S m}^{-1}$. The poly(3,4-ethylenedioxythiophene) polystyrene sulfonate nanofibrils/graphene-derived nanocomposite was used to form 3D-printed supercapacitor electrodes with fine durability and a power density of 11.3 kW kg^{-1} . Nanowire-like patterns of polythiophene and graphene-based nanomaterials have also been reported [115]. The self-assembled patterns have optical and electronic properties for optoelectronic devices.

5. Technological Potential of Polythiophene and Graphene-Derived Nanocomposites

Graphene has been found to be potentially applicable in supercapacitors [116–118]. In addition, polythiophene has sufficient charge mobility and pseudocapacitance for supercapacitor applications [119,120]. Consequently, polythiophene and graphene-derived nanocomposites have revealed applications in supercapacitors [121]. Owing to better polythiophene–graphene interactions and interface formation, superior interfacial charge transference and charge/discharge features of nanocomposites have been observed [122]. These properties resulted in fine capacitance performance for important technical utilizations [123,124]. Like polythiophene, polythiophene derivatives have also been applied as supercapacitive nanomaterials. Consequently, the resulting nanocomposites from poly(3,4-ethylenedioxythiophene) and graphene oxide have been found to be functional in high efficiency supercapacitors [125,126]. A specific capacitance of $\sim 300\text{--}600 \text{ Fg}^{-1}$ was observed for a poly(3,4-ethylenedioxythiophene)/graphene oxide nanocomposite-based supercapacitor. According to Li et al. [99], a poly [3-(2-(2-(2-(2-(diethanolamino)ethoxy)ethoxy)ethoxy)ethoxy)thiophene]-grafted graphene oxide nanocomposite-based supercapacitor device has been designed. The covalent linking between the poly [3-(2-(2-(2-(2-(diethanolamino)ethoxy)ethoxy)ethoxy)ethoxy)thiophene] and graphene oxide and interface development led to high electrical conductivity and a specific capacitance of 971 Fg^{-1} (1 Ag^{-1}). The single solid-state supercapacitor device and devices connected in series and parallel were analyzed by cyclic voltammetry to study the capacitive performance curves of the nanomaterial (Figure 5a). Moreover, the galvanostatic charge–discharge performance was studied for the single and series-connected devices (Figure 5b). The supercapacitor devices in series showed an output potential of about 0.9 V (Figure 5c). In addition, the designed supercapacitor was applied in lighting a red light-emitting diode with a working potential of 2.2 V (Figure 5d). Hence, modified graphene-filled polythiophene derivatives exhibit fine potential for the design of high-performance supercapacitor devices.

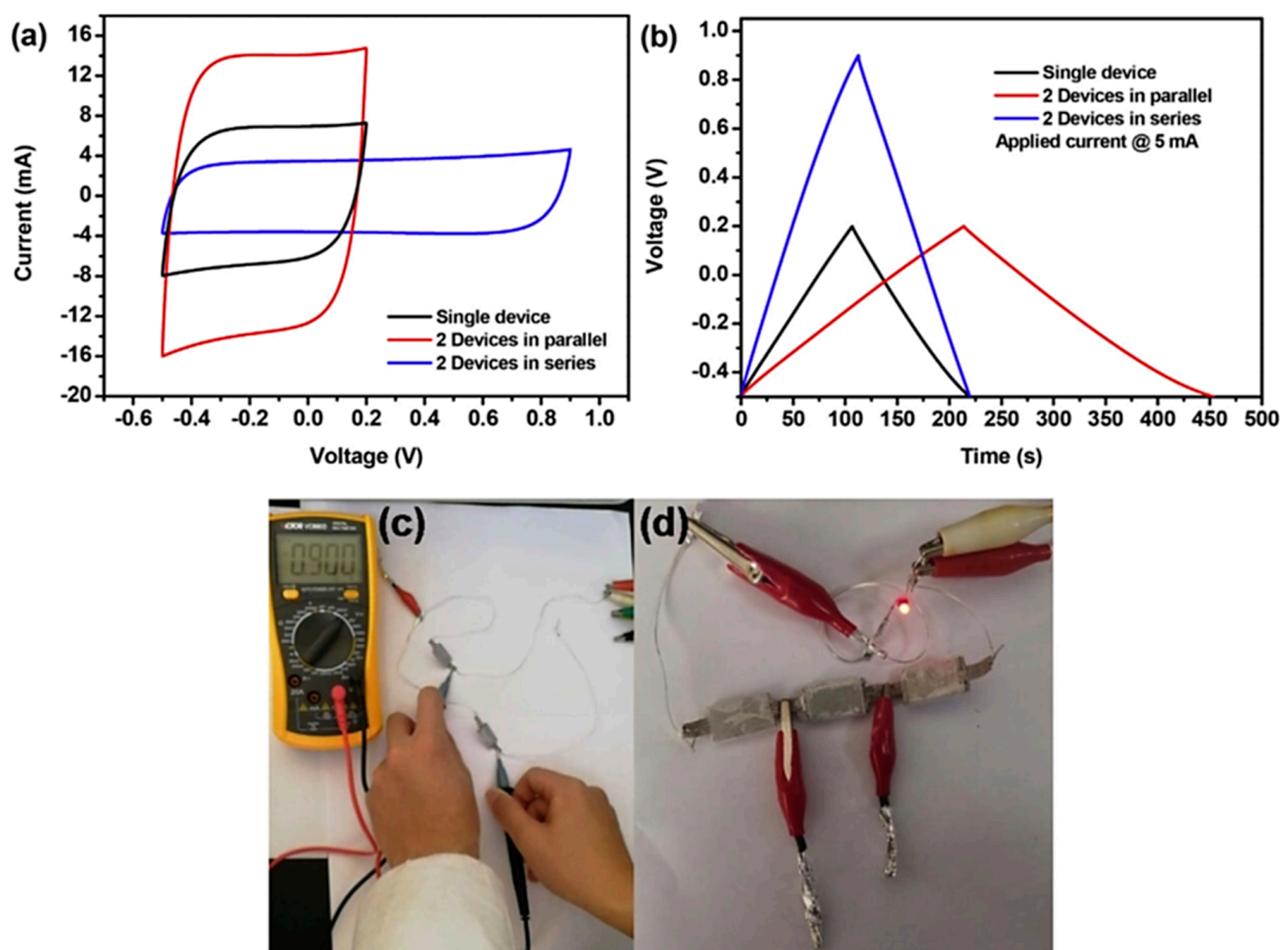


Figure 5. (a) Cyclic voltammetric curves of a single solid-state supercapacitor device, two in series, and two in parallel form (100 mVs^{-1}); (b) galvanostatic charge–discharge curves of solid-state supercapacitor devices for a single device, two device in parallel, and in series connections; (c) digital image of two devices in series having an output open potential of 0.9 V after charging of 1.5 V battery for 8 s; and (d) photograph of a red light-emitting diode (LED) powered with a three supercapacitor tandem device [99]. Reproduced with permission from Elsevier.

An important application of graphene has been observed in solar cells [127]. The nanocomposite of polythiophene and graphene revealed fine potential for high efficiency solar cells [128,129]. In nanocomposite form, polythiophene and graphene have donor–acceptor interfaces for charge transport, which are beneficial for application in solar cells [130,131]. The polythiophene derivatives behave as active electron donors, while graphene behaves as an electron acceptor for solar cell applications [132]. For example, poly-(3-hexylthiophene) and graphene-based nanocomposites have been designed for bulk heterojunction solar cells [133–135]. Stylianakis and co-workers [136] developed phenyl isothiocyanate modified graphene oxide and filled in poly(3-hexylthiophene) for bulk heterojunction solar cell application. The nanocomposite was processed using the solution method and ultrasonication technique. The resulting bulk heterojunction photovoltaic device is presented in Figure 6A. In this device, poly(3-hexylthiophene) behaves as an electron donor while graphene oxide–phenyl isothiocyanate functions as an electron acceptor. The development of a donor–acceptor complex is responsible for superior solar cell performance.

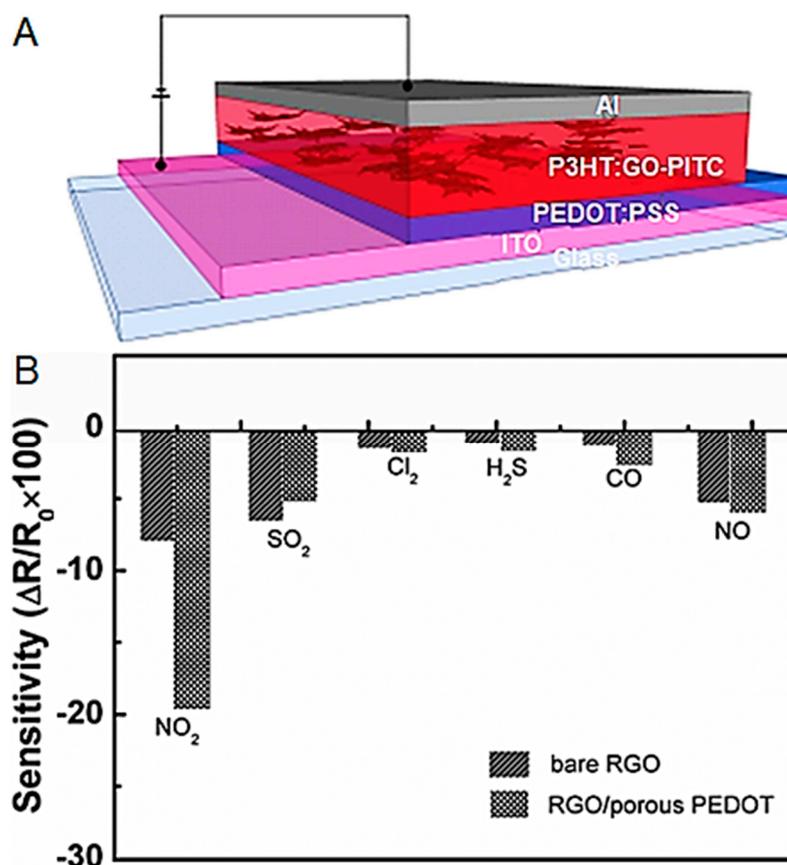


Figure 6. (A) Schematic of photovoltaic device with P3HT:GO-PITC thin film as active layer and structure ITO/PEDOT:PSS/P3HT:GO-PITC/Al [136]; and (B) plots of sensitivity of neat RGO and PEDOT/RGO nanocomposites toward different gases [137]. Al = aluminum; ITO = indium tin oxide; PEDOT:PSS = polystyrene sulfonate/poly(3-hexylthiophene); P3HT:GO-PITC = poly(3-hexylthiophene):graphene oxide with phenyl isothiocyanate; ITO/PEDOT:PSS/P3HT:GO-PITC/Al = indium tin oxide/poly(3,4-ethylenedioxythiophene):polystyrene sulfonate/poly(3-hexylthiophene):graphene oxide with phenyl isothiocyanate/aluminum; RGO = reduced graphene oxide; RGO/PEDOT = reduced graphene oxide/poly(3,4-ethylenedioxythiophene). Reproduced with permission from ACS.

Table 3 illustrates the photovoltaic features of the poly(3-hexylthiophene)/graphene oxide-phenyl isothiocyanate-derived bulk heterojunction device. Adding 20 wt.% of nanofiller was found to improve photovoltaic device parameters such as power conversion efficiency (1.02%), fill factor (0.46%), open-circuit voltage (0.51 V), and short-circuit current density (4.34 mA cm⁻²). Thus, the combination of polythiophene derivatives and graphene has revealed fine potential for photovoltaic devices.

Table 3. Device performance of photovoltaic devices based on P3HT:GO-PITC nanocomposites with different GO-PITC contents [136]. GO-PITC = graphene oxide with phenyl isothiocyanate; P3HT:GO-PITC = poly(3-hexylthiophene):graphene oxide with phenyl isothiocyanate. Voc = open-circuit voltage; Jsc = short-circuit current density (Jsc); Ff = fill factor (FF); η = power conversion efficiency. Reproduced with permission from ACS.

GO-PITC (wt.%)	Jsc (mA cm ⁻²)	Voc (mV)	Ff	η (%)
0	0.04	0.40	0.28	0.004
10	3.96	0.57	0.39	0.88
20	4.34	0.51	0.46	1.02

Graphene has its own remarkable applications for gas sensors, electrical sensors, electrochemical sensors, and strain sensors [138]. Similar to graphene, polythiophene and graphene-derived nanocomposites have been applied in sensing applications [139,140]. The polythiophene/graphene nanocomposites possessed high surface area and electrical conductivity features, facilitating sensing performance [141,142]. These nanocomposites have been effectively applied as gas sensors [143]. Bai and colleagues [144] formed polythiophene and ethylenediamine functionally reduced graphene oxide-derived nanomaterial. Including 5 wt.% of functional nanofiller in the nanocomposite was found to be effective for sensing 10 ppm of NO₂ gas. Yang and co-researchers [137] intended the poly(3,4-ethylenedioxythiophene) and reduced graphene oxide-derived nanocomposite for sensing various gases. Figure 6B shows the performance of the poly(3,4-ethylenedioxythiophene)/reduced graphene oxide nanocomposite in the presence of numerous gases. Here, NO₂ gas was applied as a standard for analyzing the sensor performance at the same concentration as the other gases. NO₂ gas at 2 ppm revealed higher sensitivity relative to the other analyte gases when analyzed with the poly(3,4-ethylenedioxythiophene)/reduced graphene oxide nanocomposite-based sensor. For comparative analysis, a neat reduced graphene oxide-based sensor was also tested for sensing different gases. The performance of the poly(3,4-ethylenedioxythiophene)/reduced graphene oxide nanocomposite sensor was found to be much higher than that of the neat carbon nanostructure owing to synergistic effects and compatible interface formation [145]. Additionally, understanding the sensing mechanism was found to be essential in analyzing the performance of the poly(3,4-ethylenedioxythiophene)/reduced graphene oxide-based nanocomposite sensor.

In addition to supercapacitors and photovoltaics, polythiophene/graphene nanocomposites have been studied for photocatalytic degradation, antibacterial, and other biomedical applications. Noreen et al. [146] formed a polythiophene and graphene nanoplatelet-derived nanocomposite using oxidative chemical polymerization. Here, various graphene nanoplatelet contents (10–50 wt.%) were reinforced in the polymer matrix. The nanocomposite was found to be useful for the photocatalytic degradation of bromo phenol blue. The nanocomposite with 50 wt.% nanofiller content led to a higher photocatalytic degradation efficiency of 94.1%, relative to pristine polythiophene (31.3%). Moreover, the polythiophene/graphene nanoplatelet nanocomposite with 50 wt.% nanofiller had fine antimicrobial activity towards *E. coli* and *S. aureus* bacterial strains. The nanocomposite had a larger inhibition zone of 18 mm for bacterial strains, relative to the neat polythiophene (5–9 mm inhibition zone). Hence, the nanocomposite was not only effective for the photocatalytic degradation of dye but was also a potential candidate for bacterial growth inhibition. Mousavi et al. [147] applied a polythiophene/graphene nanocomposite for DNA detection. These nanocomposites have appropriate binding sites to link with different biomaterials for sensing purposes. Further advancements in this field may lead to a new generation of biomedical devices for DNA or gene sensing.

6. Challenges and Conclusions

Polythiophene and graphene have been produced via non-covalent or covalent means using facile techniques. Here, graphene dispersion, polythiophene solubility, and matrix–nanofiller interactions have been found to significantly improve the physical properties of the resulting nanocomposites. Especially, the effect of graphene nanofiller was found to be essential to enhancing the microstructure, conductivity, heat conduction, strength, and other related properties. Accordingly, these nanomaterials have been found to be effective in the photovoltaic, supercapacitor, and gas sensing arenas. Important challenges in using pristine graphene nanofiller are aggregation, poor dispersion, and poor miscibility with matrices. Here, using advanced processing methods has been found to be significant for developing compatible polythiophene nanocomposites. Moreover, using modified graphene forms, such as graphene oxide, reduced graphene oxide, etc., along with polythiophene derivatives with substituted moieties has been found to enhance physical as well as chemical interactions and homogeneous dispersion. Functional graphene and

modified polythiophene may be used to develop well-interconnected, high-performance, advanced nanomaterials. Until now, polythiophene/graphene nanocomposites have been used for solar cells, sensors, and supercapacitive devices. Nonetheless, several important technical fields are still unexplored for these nanomaterials, such as the space and automotive industries, radiation defense, tissue engineering, drug delivery, bio-imaging, and many more.

In brief, this overview explains vital aspects of polythiophene and graphene-derived nanomaterials. The consequences of graphene scattering, processing, and physical characteristics on the performance of polythiophene/graphene have been thoroughly discussed. Pristine polythiophene and graphene-based nanomaterials as well as substituted polythiophene and functional graphene-derived nanocomposites have been investigated. Several exceptional physical properties have been observed for these nanocomposites. Non-modified polythiophene and non-functional graphene have disadvantages of poor dispersion, low miscibility, and interactions. Promising applications of these nanomaterials have been observed so far in solar cells, supercapacitors, sensors, etc. However, future research in this field may discover numerous additional design varieties and technical areas for polythiophene/graphene nanomaterials.

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