

REVIEW ARTICLE

Cutting-edge conjugated nanocomposites—Fundamentals and anti-corrosion significance

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ABSTRACT

This review is merely designed to throw light on the cutting-edge conjugated nanocomposites based on conjugated or conducting polymers and appropriate nanofillers. An important aspect of the conjugated nanocomposites has been observed in the anticorrosion of metals or metallic substrates. Particularly, including carbon nanoparticles (fullerene, graphene, and carbon nanotube) and inorganic nano-additives to the conjugated matrices have enhanced the physical features (morphology, electrical conductivity, mechanical stability, adhesion, and barrier properties) as well as corrosion resistance. To access the anti-corrosion potential, the conjugated nanocomposites have been coated on metal substrates using facile techniques of solution, spraying, dipping, and others. Accordingly, competent anti-corrosion conjugated nanocomposites have found potential for energy or electronic devices, engineering structures, and so on.

Keywords: conjugated; nanocomposites; matrix; conductivity; anti-corrosion

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1. Introduction

Nanocarbon nanoparticles have been considered important fillers for nanocomposites and applied for technical applications^[1,2]. Corrosion has been identified as a serious problem for metals employing industries^[3]. To thwart corrosion related challenges, polymer based materials and nanomaterials have been focused in literature^[4]. Here, conjugated or conductive materials have gained special emphasis to treat the corrosion issues of metals or other affected materials^[5]. A basic problem has been documented as the interaction of corrosion causing species to metal surfaces causing harmful effects^[6]. In this concern, the coating of conjugated materials on metal surfaces has been used to prevent corrosion^[7]. Mechanism of corrosion prevention has also been examined^[8]. Adding nanoparticles in conjugated nanocomposites has been found to further enhance the performance of these anti-corrosion coatings^[9]. The conjugated nanocomposite coatings have been explored using numerous chemical, electrochemical, and physical practices^[10,11]. According to the literature, the application of conjugated coatings on metal surfaces enhanced the adhesion strength and wear resistance, relative to neat steel metal^[12]. Fine nanoparticle dispersion has been found to enhance the nanocomposite coating performance to prevent the corrosion issue.

This article basically offers a cutting-edge review of conjugated nanocomposite coatings for anti-corrosion of metal surfaces. Fundamentals of corrosion, corrosion protection mechanisms, and coating processes have been stated. The importance of using nanocomposites and conjugated nanocomposites for corrosion protection utilizations has been analysed. Specifically, adding nanocarbon nanoparticles has been found to considerably enhance the anti-corrosion performance of these conjugated nanocomposite coatings. Morphology, physical features, and anti-corrosion performance of the conjugated nanocomposites have been deliberated. Resulting anti-corrosion coatings have been used in essential technical sectors. This is an all-embracing, revolutionary, and up-to-date review of corrosion resistance of conjugated polymers and derived nanocomposites covering design, properties, and technological relevance. To the best of our knowledge, the anticorrosion potential of conjugated polymer/nanocarbon nanocomposites has not been reviewed comprehensively in the literature before. Consequently, this article is undoubtedly a radical contribution in the field of corrosion resistance conjugated nanocomposites. Future growths in the field of anticorrosion conjugated polymer/nanocarbon nanocomposites are not possible for the field scientists or researchers before getting prior compiled literature knowledge on these materials.

2. Conjugated materials for corrosion protection

Conjugated polymers or conducting polymers have been considered as important materials for corrosion protection of metals^[13-15]. Conjugated polymers like polyaniline, polypyrrole, and others have been frequently applied in anti-corrosion coatings. Corrosion effects can be perceived in these coatings due to the surface cracking leading to the approach of corrosive species to metal surfaces. Polyaniline coatings have been deposited on the steel or iron surface to prevent corrosion effects^[16]. A direct relation has been observed between the electrical conductivity and corrosion resistance of the materials. An increase in electrical conduction has improved the electrochemical and anti-corrosion performance of the conjugated polymers^[17]. The corrosion process is usually prevented by increasing the electrical conductivity of the anticorrosion coating applied on the metal surface. Therefore, a direct relationship exists between the rise in electrical conductivity and improving corrosion resistance. Metal corrosion usually occurs when metal atoms interact with atmospheric molecules to form oxides, hydroxides, and other molecules to cut the electron flow of metal atom's orbitals. In other words, when metal atoms are corroded, electrons cannot hop through the metal for electron conduction. Once the metal surface is corroded, electron flow pathways are restricted, so lowering the electrical conductivity. Conjugated polymers usually form a protection layer over the metal surface. Research revealed that the formation of a conductive polymer layer on the metal surface offers corrosion protection *via* the formation of passive oxide layer involving oxidation-reduction reactions and also the barrier effect. Consequently, the likelihood of the corrosion phenomenon is delayed. Nevertheless, complex corrosion protection mechanisms have been found to be involved in corrosion protection by conducting polymers. The mechanism of corrosion protection by conducting polymers is in turn affected by numerous parameters. The amount of coated material may also affect the corrosion rate. In addition, polypyrrole has been used to form anti-corrosion coatings on iron or aluminum metal surfaces^[18,19]. The corrosion rate greatly relies on factors like amounts, surfactants, pH, and other ecological factors. For conjugated polymers, anti-corrosion performance has been found to be upsurged using nanofillers as well as inhibitors^[20,21].

3. Nanocomposites of conjugated polymers for corrosion resistance

Undoubtedly, conjugated polymers have been found functional for the anti-corrosion of metals^[22]. Conjugated polymers have the efficiency to better interact with the metal surfaces to avert the harmful effects^[23]. Metal surfaces commonly act as anodes^[23] and conjugated polymers perform as cathodes to encounter corrosion issues through electrochemical reactions^[24]. Temporarily, anti-corrosion occurs due to metal-conjugated polymer interface reactions^[25]. Adding additives or nanoparticles has been preferred to supplement

the anti-corrosion performance by averting the corrosive media from the metal surfaces^[26]. Consequently, conjugated nanocomposites have been designed to have superior anti-corrosion and scratch/wear resistance features, than pristine conjugated polymers^[27]. High performance coatings of conjugated nanocomposites of polyaniline, polypyrrole, and others have been reported^[28,29]. These nanocomposite coatings have been frequently deposited using the chemical or electrochemical approaches and electron conduction and charge transference effects have been examined.

4. Conjugated nanocomposites with nanocarbon for anti-corrosion

Conjugated nanocomposites have been designed using the conducting polymer matrix and carbon nanoparticle reinforcements^[30–32]. Adding nanocarbons to conjugated nanocomposites has resulted in efficient coatings with facile synthesis, physical features, and technological uses. Among carbon nano-reinforcements, carbon nanotube has been adopted as a widely used and effective nanofiller to the conjugated polymers to augment the physical profile^[33–35]. Deshpande et al.^[36] specified nanocomposite coatings of polyaniline filled with multi-walled carbon nanotubes to protect carbon steel. Open circuit potential studies were used to assess the corrosion resistance performance. Anti-corrosion coatings of polypyrrole and carbon nanotube have also been developed^[37]. Han et al.^[38] formed high performance anti-corrosion polypyrrole/multi-walled carbon nanotube nanocomposite coatings. Richard Prabakar and Pyo^[39] designed multi-walled carbon nanotube reinforced poly(3,4-ethylenedioxythiophene) nanocomposite coatings for corrosion prevention. The nanomaterial coatings had electrostatic repulsions to avert the corrosion causing anions from the metal surface. Mariano et. al.^[40] reported on polyaniline and carbon nanotube based nanocomposites focusing on electron conduction and corrosion resistance profiles. Including 1 wt.% carbon nanotube revealed high electrical conductivity of 0.06 Scm^{-1} , i.e., five orders higher than neat polyaniline (**Figure 1**). On the other hand, lower carbon nanotube loading of 0.25 wt.% exposed minor electrical conductivity of about $\sim 10^{-7} \text{ Scm}^{-1}$. **Figure 2** presents a model for the conductive carbon nanotube filled polyaniline matrix. It was proposed that the carbon nanotube bundles were uniformly aligned in the matrix and separated by an insulating polyaniline layer. The aligned nanofillers developed conduction paths through the polymer for efficient electrical conductivity. It was suggested that the coupling may occur among adjacent carbon nanotube bundles for better passage of electrons, in the direction of applied electric field. The tunnelling effects between the carbon nanotube bundles may be hindered due to the polymer layer in between.

In addition to carbon nanotubes, graphene and graphene oxide nanofillers have been used to improve the electron conductivity, barrier, mechanical, and thermal features of the conjugated polymers^[41–43]. Owing to the large surface area and surface properties, graphene and related nanoparticles exposed fine barrier properties and anti-corrosion resistance^[44]. Chang et al.^[45] studied tortuous ways in polyaniline/graphene nanocomposite coatings, responsible for averting H_2O and O_2 molecules due to barrier effects, so producing anti-corrosion effects^[46]. Lin et al.^[47] fabricated epoxy nanocomposite coatings filled with polyaniline functional graphene

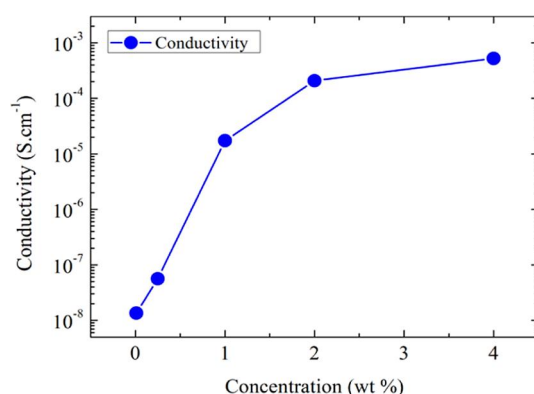


Figure 1. Conductivity as a function of carbon nanotube wt.% at 1 V^[40]. Reproduced with permission from ACS.

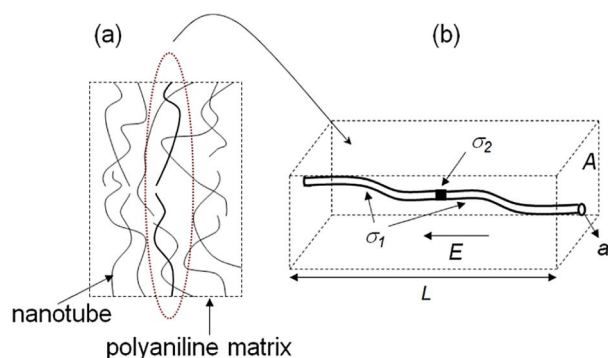


Figure 2. (a) Schematic of polyaniline/carbon nanotube nanocomposite; and (b) fibrillar conduction path through two carbon nanotube bundles separated by an insulating polyaniline layer. This junction is modelled by two domains (open and filled) with two conductivities in series^[40]. Reproduced with permission from ACS.

oxide. The coatings have been reported for fine barrier and anti-corrosion features due to mutual interactions between the polyaniline, graphene oxide, and epoxy^[48]. Corrosion inhibition effects of graphene oxide have been explored in literature^[49]. Ramezanzadeh et al.^[50] designed polyaniline nanocomposite coatings filled with graphene oxide and cerium. The nanomaterial coatings were coated on the mild steel and corrosion effects were studied. Functionalization of graphene oxide with nanoparticles further boosted the anti-corrosion performance of these coatings by developing twisted diffusion paths throughout the nanocomposites. Sheng et al.^[51] fabricated polyaniline nanocomposites filled with graphene oxide and p-phenylenediamine functional graphene oxide nanofillers. The nanomaterials were prepared using the *in situ* redox polymerization. This study explored the synergistic effects between matrix and nanofiller on the morphology and anti-corrosion characteristics of the nanocomposites. Scanning electron microscopy and transmission electron microscopy micrographs of graphene oxide and p-phenylenediamine functional graphene oxide are given in **Figure 3**. It can be observed that the p-phenylenediamine functional graphene oxide had a more wrinkled nanosheet nanostructure owing to interactions between the p-phenylenediamine and graphene oxide. Comparatively, pristine graphene oxide has a lesser crumpled and smooth nanosheet nanostructure. Owing to the well-interacted phenylenediamine-graphene oxide nanostructure, the nanosheets avoid reaggregation tendency. **Figure 4** depicts the Nyquist plots of polyaniline/phenylenediamine/graphene oxide nanocomposites with various nanofiller loading. Adding nano additives enhanced the semicircle diameters due to improvement in the charge transfer resistance or polarization of the nanomaterials. Consequently, the addition of nanofiller averted the diffusion of corrosive electrolyte molecules through the nanocomposite coating. According to equivalent circuit studies, electrolyte resistance, polarization resistance, and charge transfer resistance revealed corrosion resistance of the nanocomposite coatings. **Figure 5** depicts the proposed model for the nanocomposites with corrosion diffusion pathways. Dispersion of functional nanofiller in polyaniline caused considerable hindrance to the penetrating corrosive molecules through nanocomposite coating towards metal. On the other hand, neat polymer coating had no resistance to the seeping corrosion species, so affecting the metal surface. Henceforth, these nanocomposite coatings had up to 99.9% anti-corrosion efficiency, as per potentiodynamic polarization results.

Catt et. al.^[52] filled graphene oxide in poly(3,4-ethylenedioxythiophene) matrix to develop the nanocomposite coatings using electro-polymerization. For anti-corrosion studies, the magnesium surface was focused. **Figure 6** depicts the cracked surface of poly(3,4-ethylenedioxythiophene)/graphene oxide coating after corrosion. In corrosion current vs. voltage scans, lower or more positive corrosion current was experiential for coated sample, related to the non-coated surface. The result depicted higher anti-corrosion efficiency of the nanocomposite material.

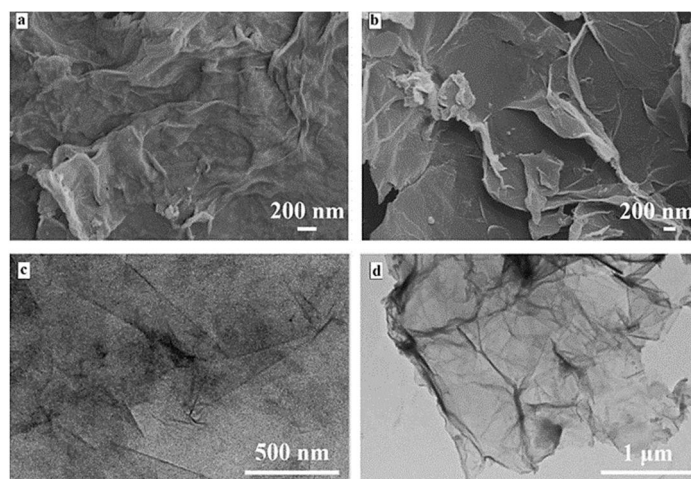


Figure 3. High magnification scanning electron microscopy images of (a) graphene oxide and (b) p-phenylenediamine functionalized graphene oxide; and transmission electron microscopy images of (c) graphene oxide and (d) p-phenylenediamine functionalized graphene oxide^[51]. Reproduced with permission from ACS.

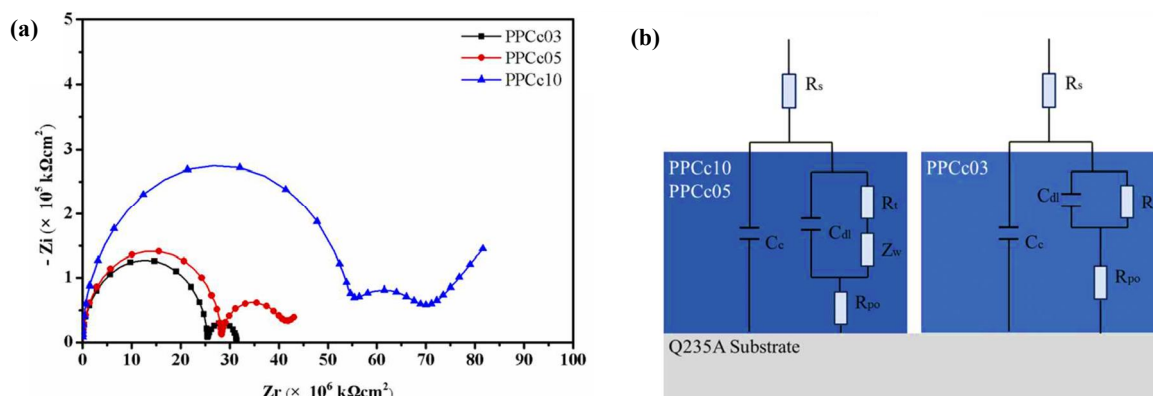


Figure 4. (a) Nyquist plots; and (b) equivalent circuit model of polyaniline/p-phenylenediamine functionalized graphene 3 wt.% (PPCc03), polyaniline/p-phenylenediamine functionalized graphene 5 wt.% (PPCc05), and polyaniline/p-phenylenediamine functionalized graphene 10 wt.% (PPCc10)^[51]. Reproduced with permission from ACS.

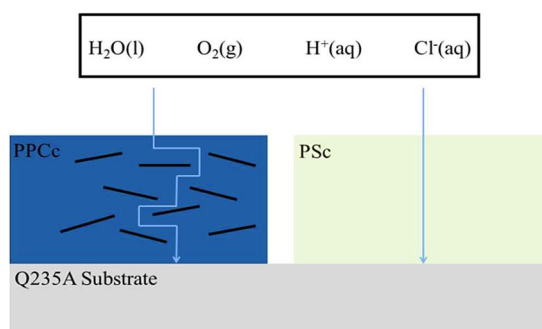


Figure 5. The model of corrosive diffusion pathways through the coatings^[51]. PPC = polyaniline/p-phenylenediamine functionalized graphene; PSc = pristine polyaniline. Reproduced with permission from ACS.

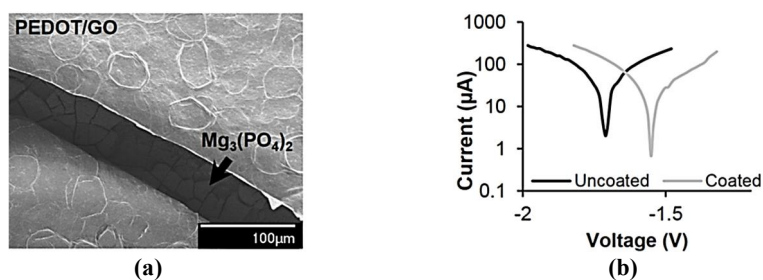


Figure 6. (a) Scanning electron microscopy image of a crack in PEDOT/GO film after corrosion with cracked scale like morphology indicated by an arrow; and (b) corrosion current vs. voltage scans of coated and uncoated surface^[52]. PEDOT/GO = poly(3,4-ethylenedioxythiophene)/graphene oxide. Reproduced with permission from Elsevier.

The inclusion of graphene and derivatives in conjugated polymers has promoted electron conductivity. This rise in electrical conductivity can be attributed to the fine electron conduction of graphene having high symmetry. Graphene has advantage of developing widespread π electron delocalization^[53]. The rise in electrical conduction of graphene coatings in turn supports oxidation-reduction reactions and barrier effects for corrosion resistance.

Fullerene is a significant zero dimensional carbon nanofiller used in corrosion resisting polymeric nanocomposites^[54–56]. Here, fullerene has capability to develop van der Waals or π - π interactions with the polymers^[57]. The mutual interactions industrialised better diffusion trails for corrosion species and supported electron conduction through the coatings^[58]. Consequently, the corrosion mechanism can be explained on the basis of matrix-nanofiller associations in charge of transporting conjugated polymer/fullerene systems^[59]. Polyaniline and fullerene based nanomaterials have been designed^[60–62]. Polyaniline has been reported to form a charge transfer complex with fullerene nanoparticles^[63]. Cheng et al.^[64] fabricated fullerene C₆₀ filled polyaniline nanocomposites. The electrical conductivity was found to increase from 9×10^{-10} to 63.7 Sm^{-1} , leading to efficient anti-corrosion performance. Gizdavic-Nikolaidis et al.^[65] also prepared a nanocomposite coating of C₆₀ nanoparticles reinforced polyaniline. Dispersion in the nanocomposite was premeditated using transmission electron microscopy. The nanoparticle sizes of 50–100 nm were observed in the matrix. Fine dispersion produced better electron transference routes to support charge transfer and anti-corrosion performance. Wang et al.^[66] developed polyaniline and fullerene C₆₀ nanowhisker derivative nanocomposites. **Figure 7** illustrates the microstructure of polyaniline doped fullerene C₆₀ nanowhiskers according to scanning electron microscopy. Adhesion of a thin polymer layer can be experiential on a fullerene nanorod surface due to the formation of a charge transfer complex. Such nanostructures have improved the electrical conductivity, leading to better corrosion confrontation^[67]. Polypyrrole has also been filled with fullerene nanoparticles to form high-tech nanocomposites^[68–70]. Wysocka-Zolopa et al.^[71] reported on the polypyrrole and fullerene C₆₀ based nanocomposites using chemical oxidative polymerization. **Figure 8** depicts the *in situ* synthesis of polypyrrole and fullerene C₆₀ resulting in nanocomposite. Here, the *in situ* route seemed effective in developing the well-interlinked polypyrrole-fullerene nanomaterial.

Polythiophene is another conjugated matrix polymerized *via* electrochemical polymerization^[72]. A doping process has been used to upsurge the electron conduction of polythiophene even higher than 100 Scm^{-1} ^[73]. Polythiophene derived forms like poly(3-octylthiophene), poly(3-hexylthiophene), poly(3-octadecylthiophene), etc., have also been focused^[74,75]. Zabihi et al.^[76] developed fullerene C₆₀ filled poly(3-hexylthiophene) nanocomposite coatings. The nanomaterial had superior charge transfer and anti-corrosion due to matrix-

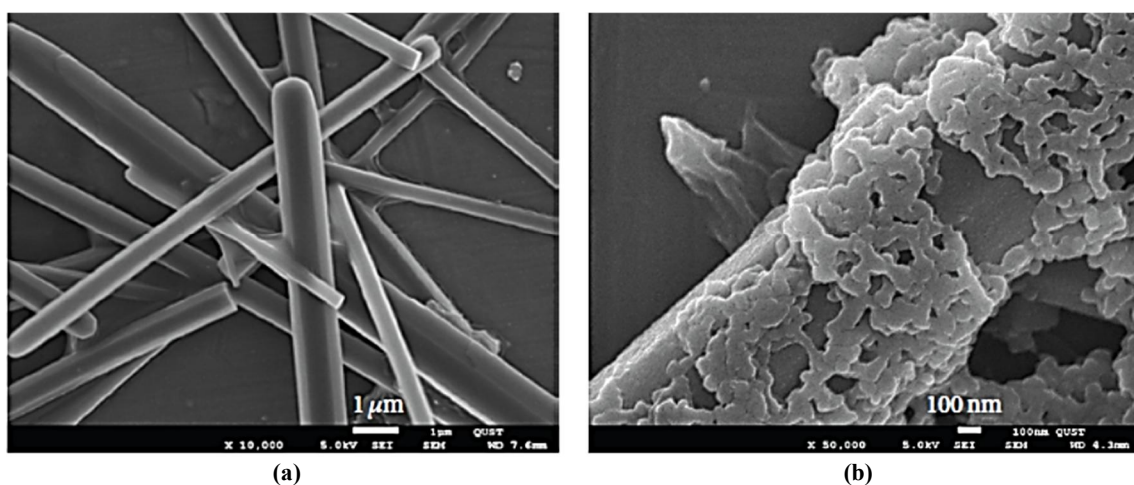


Figure 7. Scanning electron microscopy images of (a) fullerene nanowhiskers; and (b) polyaniline doped nanowhiskers^[66]. Reproduced from Hindawi (Published Open Access).

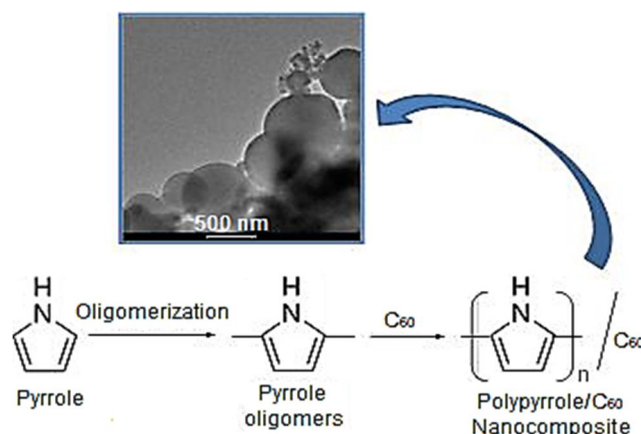


Figure 8. Development of polypyrrole and fullerene C_{60} derived nanocomposites^[71]. Reproduced with permission from ACS.

Table 1. Design, synthesis, and corrosion efficiency of conjugated polymer and nanocomposites.

Material	Synthesis	Corrosion efficiency	Ref
Polyaniline	In situ or electrochemical polymerization	$I_{\text{corr}} 0.112 \mu\text{A cm}^{-2}$; $E_{\text{corr}} -0.596 \text{ V}$ 3.5% NaCl; 8 h	[77]
Polyaniline	<i>In situ</i> polymerization ammonium persulfate; sulfuric acid	$\sim 0.06 \text{ Scm}^{-1}$	[40]
Polyaniline/carbon nanotube	<i>In situ</i> polymerization ammonium persulfate; sulfuric acid	$\sim 10^{-7} \text{ Scm}^{-1}$; Tunneling 11-15 Å vs. carbon nanotube contents	[40]
Polyaniline	<i>In situ</i> chemical oxidative polymerization; ammonium persulfate	$5.30 \times 10^{-3} \text{ Scm}^{-1}$	[78]
Polyaniline/carbon nanotube 20 wt.%	<i>In situ</i> chemical oxidative polymerization; ammonium persulfate	$2.72 \times 10^{-1} \text{ Scm}^{-1}$; Visibly decreased corrosion relative to pristine sample	[78]
Polyaniline	<i>In situ</i> chemical oxidative polymerization	$I_{\text{corr}} -610 \mu\text{A cm}^{-2}$; $E_{\text{corr}} 0.718 \text{ mV}$	[79]
Polyaniline/carbon nanotube 2 wt.%	<i>In situ</i> chemical oxidative polymerization; ammonium persulfate	$I_{\text{corr}} -467 \mu\text{A cm}^{-2}$; $E_{\text{corr}} 0.31 \text{ mV}$	[79]
Polypyrrole	<i>In situ</i> polymerization	$I_{\text{corr}} -0.943 \mu\text{A cm}^{-2}$; $E_{\text{corr}} 17.5 \text{ mV}$	[80]
Polypyrrole/carbon nanotube 1 wt.%	<i>In situ</i> polymerization	$I_{\text{corr}} -0.294 \mu\text{A cm}^{-2}$; $E_{\text{corr}} 21.5 \text{ mV}$	[80]
Polythiophene	<i>In situ</i> method	Electrical conductivity $\sigma 9.9 \times 10^{-12} \text{ Scm}^{-1}$	[81]
Polythiophene/carbon nanotube	<i>In situ</i> method	Electrical conductivity $\sigma 1.2 \times 10^{-10} \text{ Scm}^{-1}$	[81]
Polyaniline	Electrodeposition	$I_{\text{corr}} 2.46 \times 10^{-8} \text{ A cm}^{-2}$; $E_{\text{corr}} 0.086 \text{ V}$	[82]
Polyaniline/graphene oxide	Electrodeposition	$I_{\text{corr}} 5.04 \times 10^{-9} \text{ A cm}^{-2}$; $E_{\text{corr}} 0.247 \text{ V}$	[82]
Polythiophene	Solution method	$I_{\text{corr}} 3.47 \times 10^{-5} \text{ A cm}^{-2}$; $E_{\text{corr}} -0.62 \text{ V}$	[83]
Polythiophene/graphene oxide	Solution method	$I_{\text{corr}} 5.70 \times 10^{-5} \text{ A cm}^{-2}$; $E_{\text{corr}} -0.578 \text{ V}$	[83]
Poly(3-hexyl-thiophene): phenyl-C61-butyric acid methyl ester	Solution method	Increase in corrosion efficiency to 100%; 100 h	[84]

nanofiller associations. **Table 1** demonstrates important literature examples on the design, synthesis, and corrosion efficiency of pristine conjugated polymers as well as nanocarbon nanocomposites for comparison. Including nanocarbons have found to enhance the electrical conductivity as well as anticorrosion potential of the nanocomposites.

5. Prospects and conclusions

Conducting nanocomposite coatings has gained an immense stance in the methodological and engineering industries^[85,86]. Conjugated polymers may have optimum electron conductivity, however, there is a lack of structural resilience, limiting major applications. In this context, nanoparticles have been filled in the conjugated polymers to improve the adhesion, durability, and wear properties, when applied on the metal surfaces^[87]. Especially, carbonaceous nanoparticles have been used to improve the barrier effects and corrosion inhibition features of the conjugated polymers. The main factors contributing to the anti-corrosion performance include the matrix-nanofiller associations, interface formation, development of charge transportation paths, and formation of resistive paths for the seeping of corrosive species^[88]. All these aspects can contribute to the prevention of corrosive species approaching the metal surface for corrosion purposes. Corrosion prevention mechanisms have been mostly investigated in terms of barrier effects as well as the electrochemical defense of metals against corroding species^[89]. For anti-corrosion coating fabrication, mostly *in situ* polymerization, electro-polymerization, and solution practices have been applied. Recently, sophisticated techniques like spin coating, spray coating, and dipping routes have been considered for the formation of evenly layered nanocomposites on metal surfaces.

The fate of conjugated nanocomposite coatings depends upon the design and fabrication of efficient innovative self healing materials^[90]. Consequently, the self-healing phenomenon can better promote the corrosion resistance of the nanocomposite coatings^[91]. In effect, the self healing coatings have the potential to release self healing particles to recuperate the corrosion damage in the materials^[92]. In such coatings, self healing nanocapsules can be loaded which can act in specific corrosion conditions^[93]. Future labors are demanded to generate the self healing coatings of the conducting polymers and conjugated nanocomposites having outstanding anti-corrosion efficiency^[94]. In this way, self healing anticorrosion coatings need to be designed for future electronics, automobile, and engineering industries^[95].

This article is planned to schematically portray the impact of conjugated polymer and conjugated nanocomposites in the corrosion protection of metals. For the formation of conjugated nanocomposite coatings, conducting polymers like polyaniline, polypyrrole, polythiophene, and related derivatives have been used as matrices. These polymers were filled with nanocarbon nanofillers to enhance the anti-corrosion features. Facile routes have been adopted to fill the conjugated matrices with carbon nanoparticles. Subsequently, anti-corrosion properties and involved mechanisms were examined. Carbon nanoparticles have resulted in improved anti-corrosion performance due to fine nano-additive dispersion, mutual interactions, and the formation of charge or electron diffusion pathways throughout the nanocomposite. All these synergistic effects led to a fine anti-corrosion performance. Hence, the conjugated nanocomposites have been efficiently used to protect the metals and metallic components from harmful corrosion effects.

Author contributions

Conceptualization, AK; methodology, AK; data curation, AK; writing—original draft preparation, AK; writing—review and editing, AK, and IA; visualization, AK; supervision, AK; project administration, AK. All authors have read and agreed to the published version of the manuscript.

Conflict of interest

The authors declare no conflict of interest.

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