Review

Avant-Garde Polymer/Graphene Nanocomposites for Corrosion Protection: Design, Features, and Performance

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Abstract: Polymeric coatings have been widely selected for the corrosion resistance of metallic surfaces. Both the conducting and non-conducting polymers have been applied for corrosion confrontation. The conducting polymers usually possess high electrical conductivity and corrosion resistance features. On the other hand, non-conducting hydrophobic polymers have also been used to avert the metal erosion. To improve the corrosion inhibition performance of the polymer coatings, nanocarbon nanofillers have been used as reinforcement. Graphene, especially, has gained an important position in the research on the corrosion-protecting nanocomposite coatings. Here, graphene dispersion and matrix–nanofiller interactions may significantly improve the anti-corrosion performance to protect the underlying metals. The graphene nanofiller may form an interconnecting percolation network in the polymers to support their electrical conductivity and thus their corrosion confrontation characteristics. Further research on the polymer/graphene nanocomposite and its anti-corrosion mechanism may lead to great advancements in this field.

Keywords: anti-corrosion; coating; conducting polymer; nanocomposite; graphene; dispersion
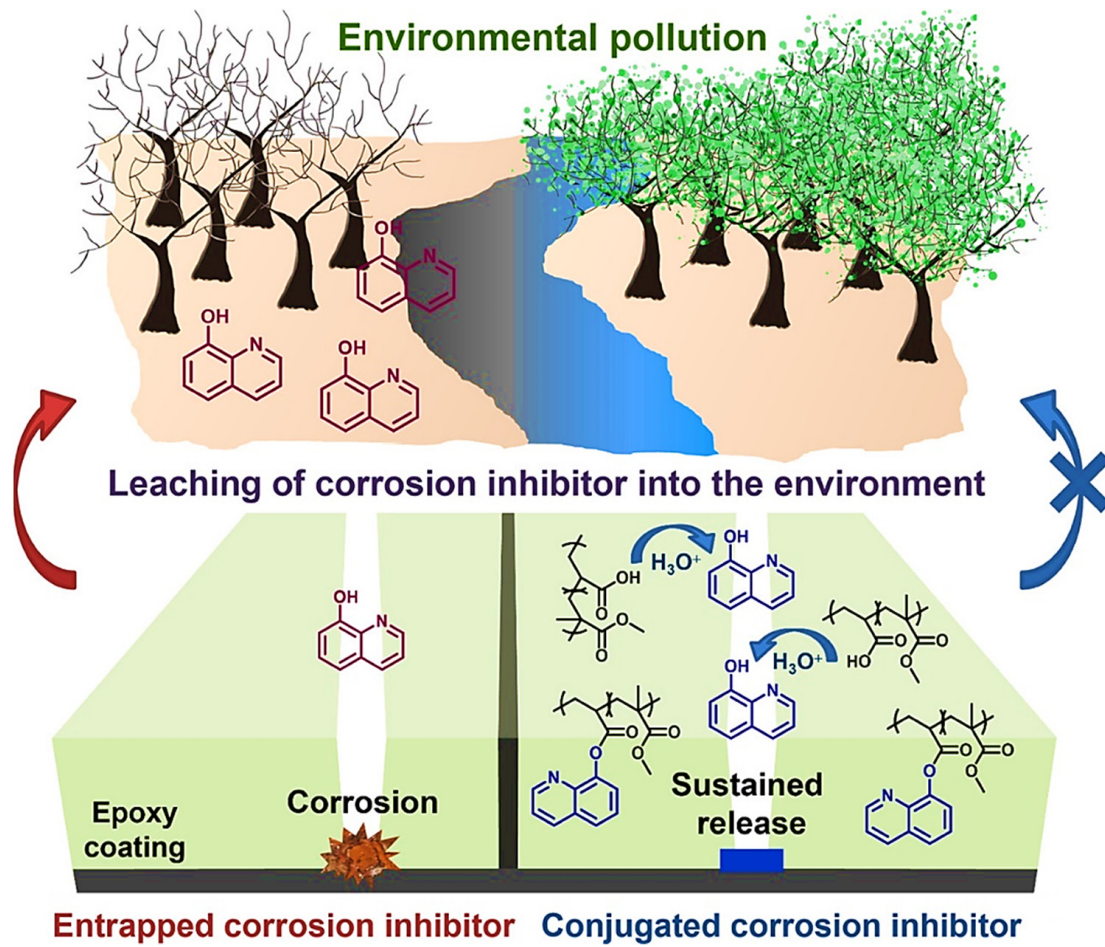
1. Introduction

Corrosion is a serious technical issue affecting the progressive metal-based industries [1]. In metal industries, implementation of the anti-corrosion technology, protective coatings, and corrosion inhibitors have been the focused [2,3]. The durability and performance of metal-based materials can be enhanced through the use of corrosion protection coverings [4]. Among corrosion inhibition coatings are nanocomposite coatings, hybrid coatings, and healing coatings [5]. The nanocarbon nanofillers such as graphene, carbon nanotube, carbon nanoparticles, etc., have been filled in the polymeric coatings to enhance the corrosion performance [6]. The ensuing nanocomposite coatings have enhanced anti-corrosion, impermeability, and barrier properties, relative to the neat polymers [7]. Thin-layer and multi-layer graphene have been used as corrosion protective coatings on copper, nickel, or other metal surfaces [8]. The polymers reinforced with graphene have been used to form anti-rusting nanocomposite coatings [9]. Graphene nanocomposite with conducting and non-conducting polymers displayed fine anti-corrosion features. The polymer/graphene coatings have been develop using various techniques such as solvent casting, spray coating, dip coating, doctor blading, spin coating, etc. [10]. Graphene nanofiller may form tortuous diffusion paths in the polymeric matrices to prevent the percolation of corrosive molecules to the metal surface. Consequently, graphene dispersion influences the barrier properties of the polymer coatings [11]. In fact, the van der Waals or π–π

interactions in polymer/graphene may facilitate better interfacial interactions, dispersion, and enhanced barrier properties. The anti-corrosion performance of polymer/graphene nanocomposite coatings have been established through several analytical and physical techniques [12]. In this cutting-edge review, various noteworthy prospects of the corrosion resisting polymer/graphene nanocomposites have been deliberated. In particular, the design versatilities, essential features, and implication of anti-corrosion polymer/graphene nanocomposites have been stated.

2. Polymers in Corrosion Reticence

Corrosion is an environmental occurrence damaging the metal surface via chemical or electrochemical reactions [13]. To prevent corrosion, coatings have been applied on the metal surface to prevent the rusting process. For corrosion protecting coatings, adhesion and chemical inertness have been considered important to enhance the anti-corrosion properties for aerospace, automobile, construction, and engineering parts and industries [14–16]. In this regard, polymer coatings on iron, copper, and related alloy surfaces prone to rusting have been used for the corrosion inhibition [17]. The organic polymers are generally hydrophobic, so do not adsorb water molecules on the metal surface and avert the corrosive media [18]. Moreover, the organic polymers may prevent the chemical reactions occurring on the metal surface and at metal–corrosion media interfaces [19]. The conducting polymers have been commonly used as corrosion resistant coatings on the metal surfaces [20–22]. Conductive polymers or intrinsically conducting polymers are organic polymers with the ability to conduct electricity. These polymers have metallic conductivity or can be semiconductors. Among these polymers, polyaniline, polypyrrole, etc., can be galvanostatically deposited on carbon steel/stainless steel or other metallic surfaces. The corrosion rate of metal surfaces covered with coatings depends on the temperature, additives, pH, and different nanofillers addition. Neat polyaniline has been used as a corrosion resistant coating for mild steel [23]. The corrosion resistance of polyaniline coating has been explored using the electrochemical impedance spectroscopy (EIS), Tafel polarization, and degradation measurement techniques. On the other hand, non-conducting polymers are electrically insulating materials. These materials mainly comprise thermoplastics (polyethylene, polypropylene, polystyrenes, and countless others) and thermosetting polymers (epoxies, phenolics, polycarbonates, etc.). Non-conducting polymers such as epoxy has been effectively applied as anti-corrosion coatings on metal surfaces. To enhance the anti-corrosion efficiency of the epoxy coatings, various additives have also been applied (Figure 1) [24].

Thus, epoxy coatings with entrapped corrosion inhibitors have been used. Upon corrosion occurrence, the corrosion inhibitors may be released from the polymer-corrosion inhibitor conjugate to prevent the phenomenon. This approach has been found useful for the development of efficient coating materials with high corrosion protection efficiency. Table 1 shows some frequently used effective polymers adopted as anti-corrosion coatings.
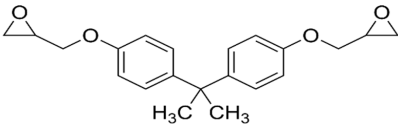
Figure 1. Anti-corrosion additives for commercial epoxy coatings [24]. Reproduced with permission from Elsevier.

Table 1. Corrosion protection polymers.

Polymer	Symbol	Structure	Ref.
Polyaniline (Conducting polymer)	PANI		[25]
Polypyrrole (Conducting polymer)	PPy		[21]
Poly(ethylene glycol) (Non-conducting polymer)	PEG		[26]

Table 1. *Cont.*

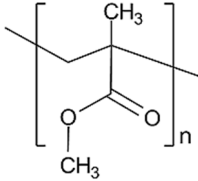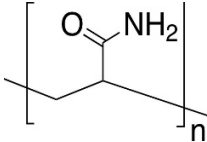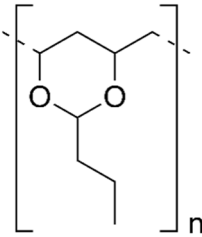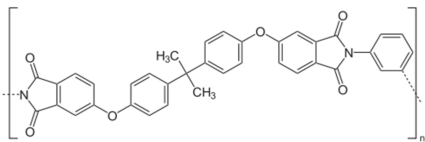| Polymer	Symbol	Structure	Ref.
Epoxy (Non-conducting polymer)	Bisphenol A diglycidyl ether (DGEBA)		[27]
Poly(methyl methacrylate) (Non-conducting polymer)	PMMA		[28]
Poly(acrylamide) (Non-conducting polymer)	PAM		[29]
Polyvinylbutyral (Non-conducting polymer)	PVB		[30]
Poly(ether imide) (Non-conducting polymer)	PEI		[31]

3. Corrosion Resistance by Polymeric Nanocomposites

The polymeric nanocomposites have been developed by using reinforcing nanoparticle nanofillers in the polymeric matrices [32]. The mixing of polymer with nanoparticles have improved the conducting, mechanical, thermal, barrier, anti-corrosion, and other physical properties [33]. Consequently, the polymeric nanocomposites have been concentrated for the aerospace, automotive, electronics, textiles, and biomedical fields [34]. The conducting polymers have been efficiently used for the corrosion protection [35]. The conducting polymers have fine adhesion to the metallic surfaces to prevent corrosion, wear, and scratch damage [36]. A recent study on polyaniline (PANI) as a modifier for functionalized carbon nanotube (CNT) in a silane matrix revealed corrosion and barrier protection of the mild steel samples [37]. Figure 2 shows the silane network linked polyaniline/carbon nanotube nanocomposite. The silane acts as an efficient corrosion inhibitor in the polyaniline/carbon nanotube nanocomposite. The use of silane linked nanocomposite coatings prevented the formation of defect sites on the coating surface and developed fine adhesion to the metallic surface. The electrochemical impedance, barrier properties, and scratch resistance of the nanocomposite coatings were investigated.
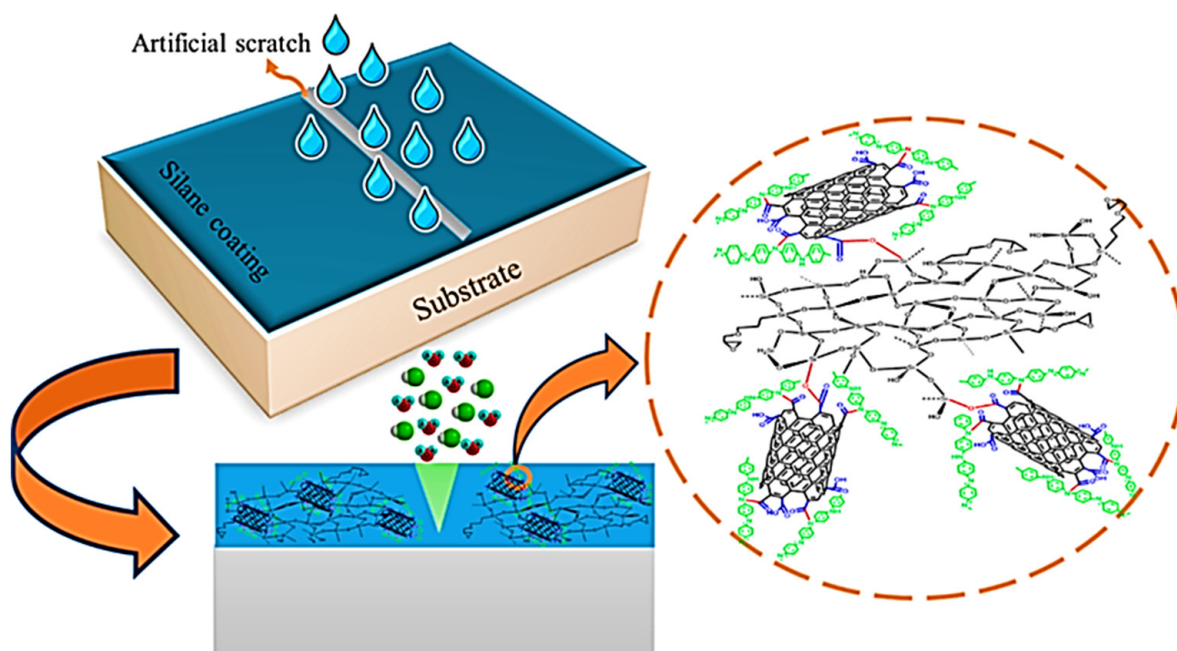
Figure 2. Schematic of interaction of polyaniline/carbon nanotube with silane matrix and metal substrate [37]. Reproduced with permission from ACS.

Non-conducting polymers have also been applied as the corrosion protection materials [38]. Among these, the polytetrafluoroethylene (PTFE) with Al_2O_3 nanoparticles has been applied as an anti-corrosion coating [39]. The scratch and wear resistance of the nanocomposite coating were analyzed [40]. The poly(ether imide)/hydroxyapatite nanocomposite coatings have also been designed for corrosion resistance [31]. EIS was used to study the coating performance. The polyvinyl butyral (PVB) and polyaniline (PANI) coatings with nano-TiO_2 filler have been pragmatic to form the anti-corrosion coatings [41,42]. Hence, polymeric nanocomposite coatings have pointed towards the corrosion resistance applications.

## 4. Polymer/Graphene Nanocomposites Coatings for Corrosion Protection

The corrosion resistance coatings have been used as a barrier layer between the corrosive species and the metal substrate [43,44]. In the nanocomposites based anti-corrosion coatings, the erosion protection efficiency relies on the type of nanoparticle and dispersion in the matrix [45,46]. Moreover, the coating thickness can be considered to enhance the corrosion inhibition [47]. The additives or nanoparticles used as nano-reinforcement can be inorganic, metallic, or organic in nature [48]. Among the carbon-based materials, nano scale graphene reinforcement stands out as an efficient corrosion resisting barrier layer [49]. The polymer/graphene nanocomposites possess the advantages of inexpensiveness, facile processing, and being lightweight [50]. The graphene reinforced polymeric nanocomposites have been applied as the anti-corrosion coating on the metal substrates [51]. These nanocomposites have excellent anti-corrosion performance owing to barrier effects on common corrosive mediums such as water and chemicals without affecting the properties of the base metal. For this reason, polymer and graphene derived coatings have been considered as the thinnest protective coatings. The nanocomposites have been emerged as promising anti-corrosive materials owing to unique properties such as impermeability, chemical inertness, high conductivity, and scalability. The mechanism of corrosion resistance of the polymer/graphene nanocomposite relies on graphene dispersion, adhesion to metal, coating thickness, and coating homogeneity [52,53]. Moreover, the nanocomposite layers may have minimum porosity and tortuous diffusion paths for corrosive species to delay the corrosion phenomenon [54]. Wide range of non-conducting polymers including ther-

mosets or thermoplastic matrices have been applied as the anti-corrosion coatings [55]. The electrical conductivity, thermal, mechanical, wear, barrier, permeability, and other related physical properties of the polymer/graphene nanocomposites have been investigated.

Epoxy resins have been commonly used for the production of corrosion resistance coatings [56]. Epoxy resins possess fine adhesion, heat stability, and chemical stability features [57,58]. Addition of graphene in epoxy based coatings have improved the anti-corrosion capabilities of the matrix [59]. The epoxy/graphene nanocomposites revealed high hydrophobicity and water contact angle and so attain high moisture resistance. Graphene oxide (GO) nanofiller has also been filled in the epoxy coatings [60]. Both the graphene and GO nanofillers have proficiency to hinder the permeation of corrosive molecules through the nanocomposite layer [61,62]. Zhang et al. [63] developed the neat epoxy resin and epoxy/graphene nanocomposite coatings (filled with varying graphene contents). The anti-wear and anti-corrosion performance of the coatings have been analyzed. Figure 3 presents the corrosion protective mechanism of the neat epoxy coating and epoxy/graphene nanocomposite coating. The difference between the Figure 3a,b can be observed in the form of the presence of graphene nanosheets in Figure 3b. The inclusion of graphene nanosheets created a hindrance to the percolation of corrosive species via homogeneous dispersion in polymer matrix [64]. In this way, graphene dispersion formed tortuous pathways for the percolation of corrosion molecules to prevent these species from reaching the metal surface. Thus, the probability of the corrosion phenomenon is delayed. Moreover, the sliding performance of the neat epoxy and epoxy/graphene nanocomposite coatings was analyzed. It has been observed that the epoxy coating with graphene additive prevented the formation of voids, pinholes, or microcracks after sliding. Figure 4 illustrates the potentiodynamic polarization curve of neat epoxy and epoxy/graphene nanocomposites studied in 3.5% NaCl solution. Table 2 depicts the electrochemical corrosion parameters via the Tafel extrapolation approach [65]. Inclusion of 0.4 wt.% graphene has caused positive shift in the corrosion potential of the nanocomposite relative to neat epoxy.
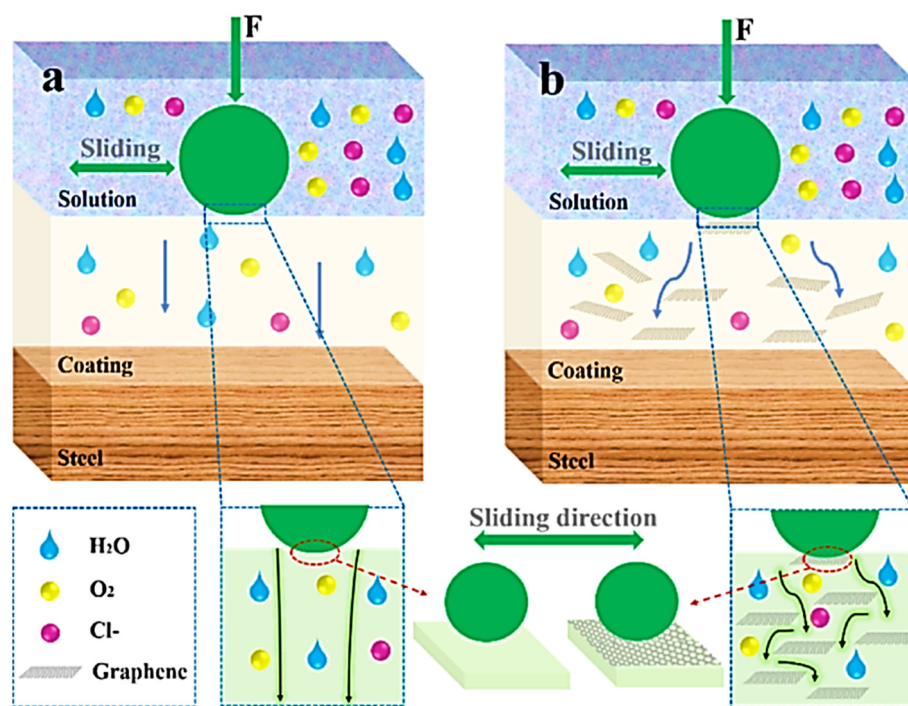
Figure 3. Schematic of mechanism of wear and corrosion resistance of the (**a**) epoxy coating and (**b**) epoxy/graphene nanocomposite coating [63]. Reproduced with permission from Elsevier.
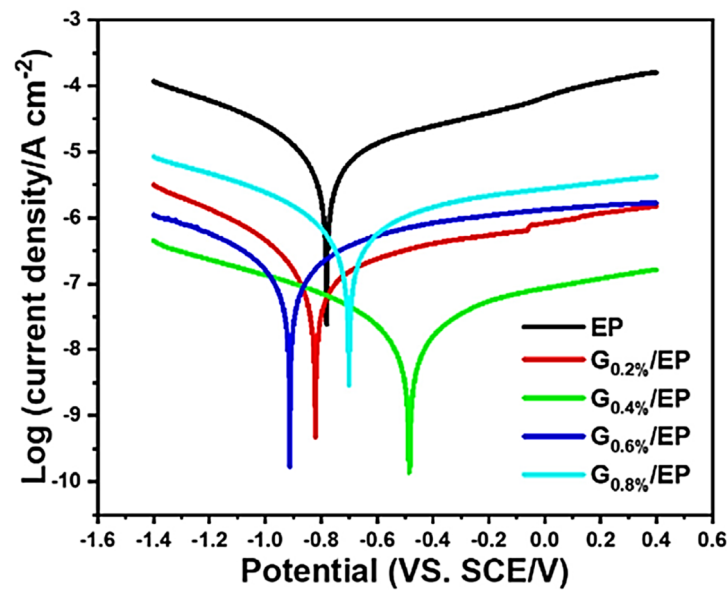
Figure 4. Potentiodynamic polarization curve of neat epoxy (EP) and epoxy/graphene (G/EP) nanocomposite coatings [63]. Reproduced with permission from Elsevier.

Table 2. Corrosion parameters of neat epoxy (EP) and epoxy/graphene (G/EP) nanocomposite coatings [63]. Reproduced with permission from Elsevier.

Sample	EP	$G_{0.2\%}$/EP	$G_{0.4\%}$/EP	$G_{0.6\%}$/EP	$G_{0.8\%}$/EP
I_{corr} (Acm^{-2})	4.11×10^{-6}	7.26×10^{-8}	8.08×10^{-9}	7.96×10^{-8}	2.75×10^{-7}
E_{corr} (V)	-0.78	-0.62	-0.49	-0.71	-0.70

The epoxy nanocomposite with 0.4 wt.% nanofiller presented lower corrosion current density (8.08×10^{-9} Acm^{-2}) and higher corrosion potential (-0.49) relative to neat epoxy coating, signifying superior barrier properties. Compared to neat epoxy coating, the corrosion current density of 0.4% nanofiller filled coating was dropped by two orders of magnitude, and corrosion potential increased by 37.2%. Thus, the results suggested better anti-corrosion efficiency of the epoxy/graphene nanocomposites, compared with the pristine epoxy resin [66]. Actually, the addition of 0.4 wt.% graphene nanosheets enhanced the tortuosity of penetration of aggressive media and water molecules in the coating, demonstrating a labyrinth effect [67]. Figure 5 depicts the salt spray test results of neat epoxy and epoxy/graphene nanocomposite coatings. The coatings with different GO contents were placed in salt spray box for 72 h. The results showed that neat epoxy coating was severely rusted and damaged at the scratched areas, so showing poor anti-corrosion behavior [68]. On the other hand, epoxy/graphene nanocomposite coatings have no obvious rust spots [69]. After 144 h of salt spray test, severe corrosion was observed in the form of dark yellow rust spots on scratched areas for neat epoxy coating. However, the nanocomposite samples were visibly less corroded. The rusting progress was further evident after 196 h of salt spray test. Remarkably, the rusting of nanocomposite coating with 0.4 wt.% nanofiller was held at a slight level, in each test duration. The authors [63] suggested that the 0.4 wt.% graphene loading was optimal in its enhancing of the anti-corrosion performance of the coating through better dispersion, matrix–nanofiller interaction, and better barrier effects.
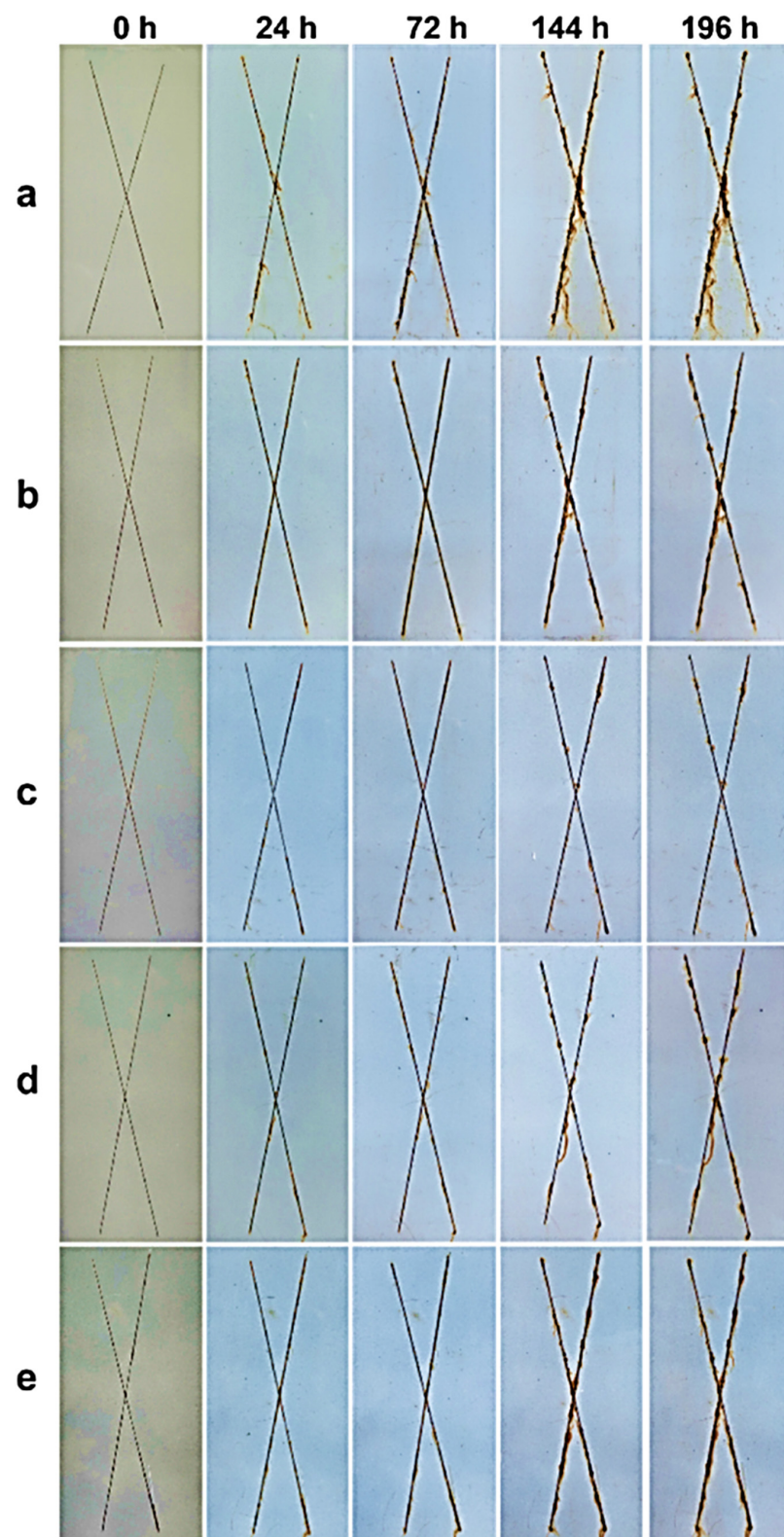
Figure 5. Digital images of the salt spray test for the (**a**) epoxy; (**b**) epoxy/graphene 0.2%; (**c**) epoxy/graphene 0.4%; (**d**) epoxy/graphene 0.6%; and (**e**) epoxy/graphene 0.8% coatings, respectively [63]. Reproduced with permission from Elsevier.

Jiang et al. [70] reported the epoxy (EP)/graphene oxide (GO) nanocomposite for the corrosion resistance application. EIS was used to explore the electrochemical behavior of the coatings. Figure 6 displays the Nyquist plots of the neat epoxy and nanocomposite coatings

tested in 3.5 wt.% NaCl solution. The decrease in the values of the impedance suggested the constant penetration of corrosive species through the coatings and also the barrier aptitude of the coatings. Consequently, the investigations on the epoxy/graphene and epoxy/graphene oxide nanomaterials offered an efficient strategy towards the corrosion resistance of the metallic materials.
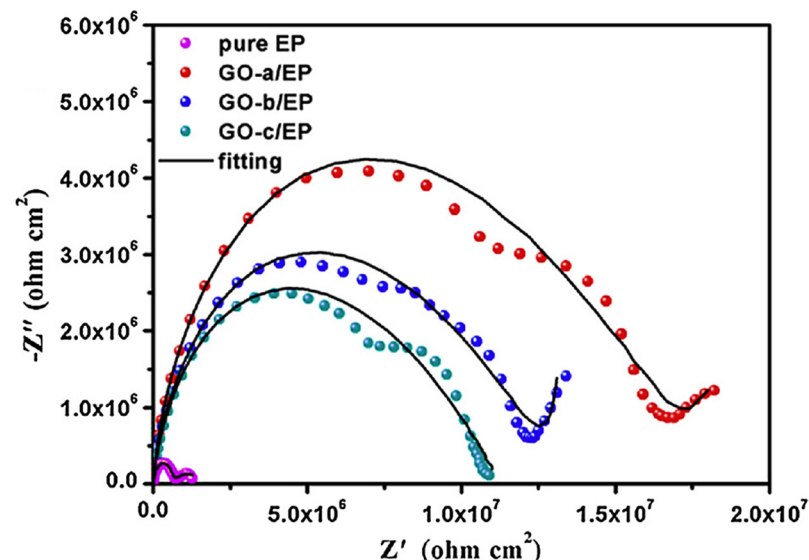
**Figure 6.** Nyquist plots of coatings immersed in 3.5 wt.% NaCl solution (7 days) [70]. GO-a/EP = Graphene oxide prepared using 3 g KMnO$_4$/Epoxy; GO-b/EP = Graphene oxide prepared using 5 g KMnO$_4$/Epoxy; and GO-c/EP = Graphene oxide prepared using 8 g KMnO$_4$/Epoxy. Reproduced with permission from Elsevier.

In that study [70], graphene oxide samples with different aspect ratio were prepared by using different amounts of oxidant, i.e., KMnO$_4$. The large capacitive loops of the nanocomposite sample GO-a/EP (graphene oxide prepared using 3 g KMnO$_4$/epoxy) and GO-b/EP (graphene oxide prepared using 5 g KMnO$_4$/epoxy) relative to neat epoxy coating were attributed to the higher aspect ratio of graphene oxide contributing to the better corrosion resistance of the nanocomposite coatings. The higher aspect ratio of graphene oxide nanosheets can better block the defects and pores in the coating. Moreover, the authors [70] also reported that the uniform nanofiller dispersion in the epoxy matrix donated to the corrosion resistance of the nanocomposite coatings.
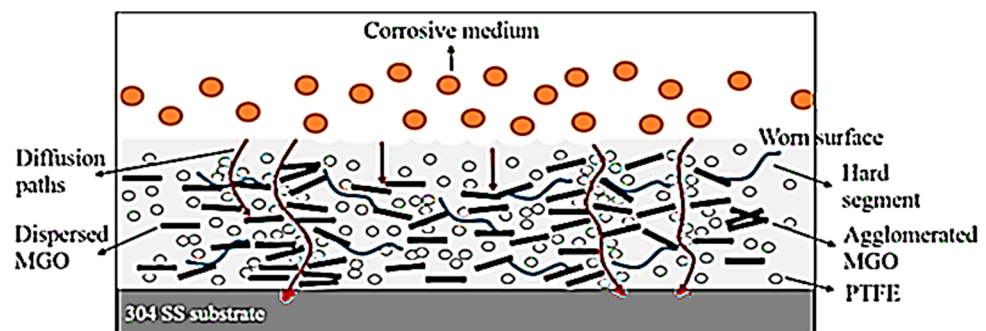
Polyurethane/graphene nanocomposites have been applied in the corrosion resisting applications [71]. Tsai et al. [72] developed polyurethane/graphene nanocomposite coatings and studied their corrosion resistance, adhesion, and anti-friction characteristics. Better anti-corrosion and adhesion performances were observed at the 4–8 wt.% graphene loading level. Bai et al. [73] used the modified graphene oxide to develop anti-corrosion coatings with the waterborne polyurethane and polytetrafluoroethylene. The synergistic effect of polymers and modified graphene oxide were found to improve the anti-corrosion performance. According to EIS studies, the corrosion resistance efficiency of the nanocomposite coatings was found superior relative to the neat waterborne polyurethane coating, as can be seen in Figure 7. Figure 8 presents a recent corrosion prevention mechanism of the waterborne polyurethane nanocomposite coating. After adding graphene oxide to waterborne polyurethane and polytetrafluoroethylene matrix, the uniform dispersion of nanofiller was attained causing better barrier effect. In turn, the graphene oxide dispersal blocked the diffusion pathways of the corrosive species. Use of polytetrafluoroethylene in the waterborne polyurethane also filled the gaps between the graphene oxide nanosheets and further enhanced the tortuosity of the diffusion paths of the corrosive medium. Therefore, the waterborne polyurethane/polytetrafluoroethylene/graphene oxide nanocomposite revealed fine corrosion resistance. Thus, the modified graphene oxide uniformly dispersed

in the polymer matrix developed tortuous diffusion pathways for the corrosion species causing the barrier effect.
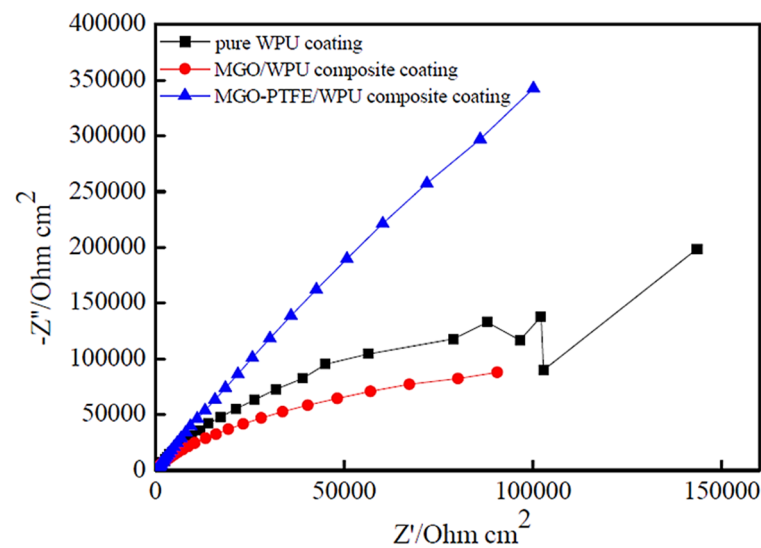

Figure 7. EIS diagrams of pure WPU; 0.5 wt.% MGO/WPU nanocomposite; and 0.5 wt.% MGO-PTFE/WPU nanocomposite coatings [73]. WPU = waterborne polyurethane; MGO/WPU = modified graphene oxide/waterborne polyurethane; MGO-PTFE/WPU = modified graphene oxide/ polytetrafluoroethylene/waterborne polyurethane. Reproduced with permission from MDPI.


Figure 8. The anticorrosion mechanism of waterborne polyurethane nanocomposite coating [73]. PTFE = polytetrafluoroethylene; MGO = modified graphene oxide. Reproduced with permission from MDPI.

Singh et al. [74] studied polymeric isocyanate crosslinked with hydroxy functional acrylic adhesive matrix and hydrophobic graphene oxide-based nanocomposite. The corrosion resistant nanocomposite coatings were fabricated on the copper by electrophoretic deposition (EPD). Potentiodynamic polarization and EIS techniques were used to study the electrochemical degradation of the nanocomposite coating. The nanocomposite coatings seemed impermeable to ion diffusion of oxidizing gas and corrosive liquid solution. Figure 9 shows the EPD set-up. Here, two parallel copper plates (cathode and counter electrode) were separated by 10 mm gap. Cathode was deposited with the nanocomposite. EPD was performed at constant DC voltage mode 10–30 V. Figure 10 presents the deposition current as a function of deposition time. It can be observed that, initially, the current decreased sharply for a short span of time when the voltage was applied and then reached a stable state at protracted deposition time. The initial rapid fall in current was attributed by the authors [74] to the increase in graphene oxide concentrations on cathode due to the DC electric field. Potentiodynamic polarization and EIS were carried out in 3.5 wt.% NaCl solution using three electrode electrochemical cell to study the corrosion

behavior of nanocomposite coating. The EIS was carried out between 10 mHz and 100 kHz frequency range using an electrochemical analyzer. Figure 11 shows the Tafel plot for the nanocomposite coating. The nanocomposite coating had small corrosion current density (3.49 $\mu A/cm^2$) relative to bare copper (38.25 $\mu A/cm^2$), showing enhanced corrosion protection of the nanomaterial. Figure 12 shows the field emission scanning electron microscopy images of the copper substrate before and after the corrosion test and the nanocomposite coating after the corrosion test. The copper substrate was found entirely damaged (cracked surface) after exposure to corrosion in 3.5% sodium chloride solution. On the other hand, the micrograph of the nanocomposite coated copper revealed that the metal remained undamaged after corrosion testing in 3.5% sodium chloride solution. Thus, the surface analysis of nanocomposite coating also revealed superior anti-corrosion capability.
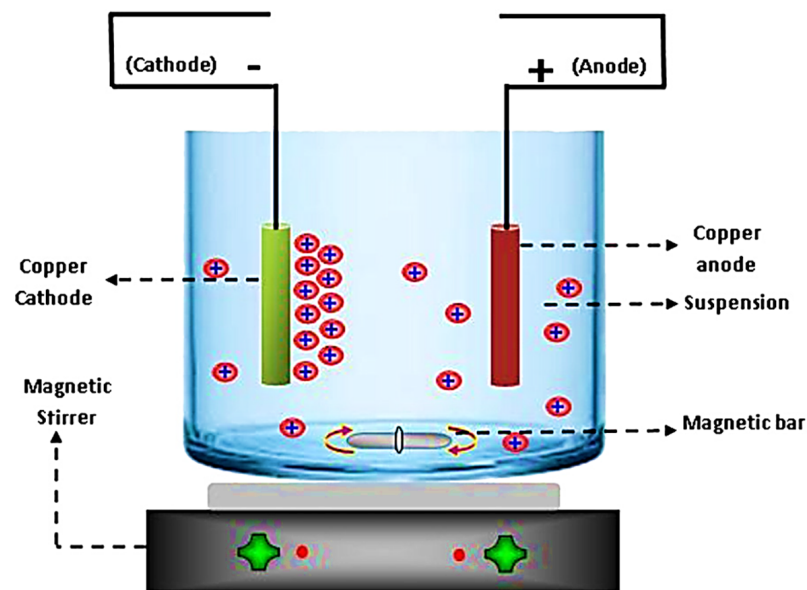

Figure 9. Schematic of the electrophoretic deposition setup [74]. Reproduced with permission from Elsevier (Creative Commons CC-BY-NC-ND license).
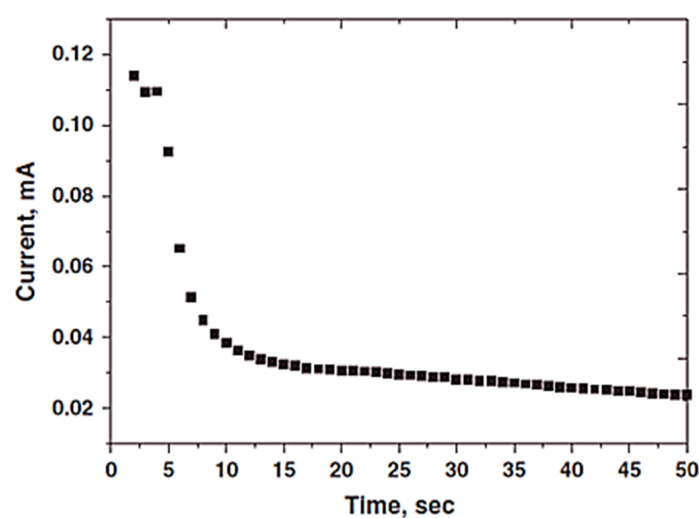
Figure 10. Deposition time vs. deposition current [74]. Reproduced with permission from Elsevier (Creative Commons CC-BY-NC-ND license).
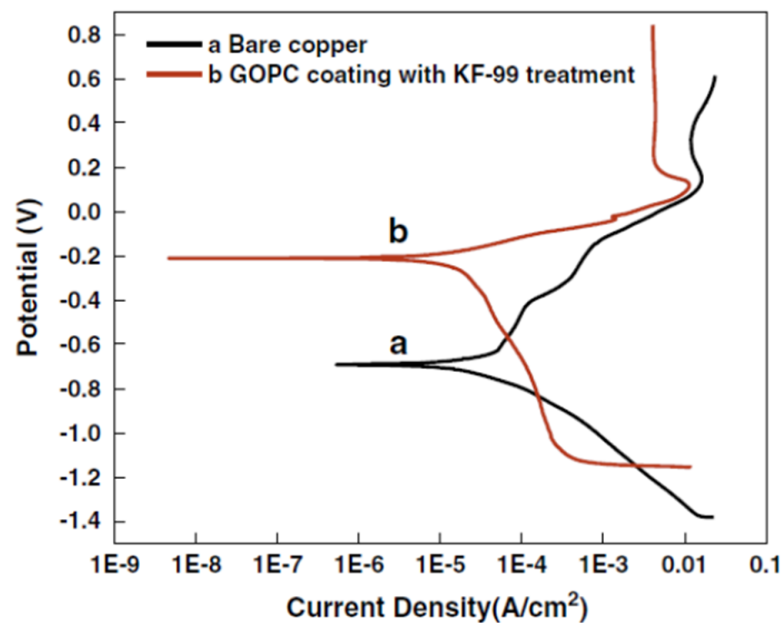
Figure 11. Tafel plots of (**a**) bare copper and (**b**) GOPC (graphene oxide polymer composite) coated copper [74]. Reproduced with permission from Elsevier (Creative Commons CC-BY-NC-ND license).
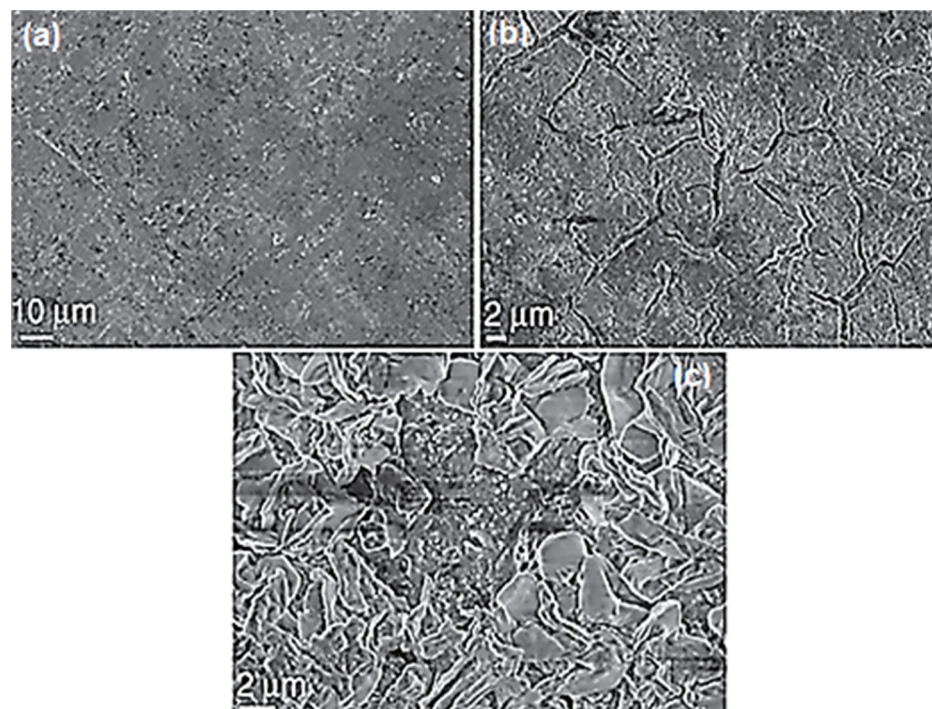
Figure 12. Field Emission Scanning Electron Microscopy images of: (**a**) bare copper substrate before corrosion test; (**b**) copper substrate after corrosion test; and (**c**) GOPC (graphene oxide polymer composite) coated copper coating after corrosion test [74]. Reproduced with permission from Elsevier (Creative Commons CC-BY-NC-ND license).

Qi et al. [75] reported on the poly(methyl methacrylate)/graphene oxide coatings through atom transfer radical polymerization. The poly(methyl methacrylate)/graphene oxide coatings had permeation reservations and superior physical characteristics [76]. Harb et al. [77] also prepared the poly(methyl methacrylate)/graphene oxide nanocomposite coating through in situ polymerization. Here, 3-(trimethoxysilyl)propyl methacrylate was used to form the silica domains in poly(methyl methacrylate)/graphene oxide coatings

by the hydrolytic condensation. The anti-corrosion coatings were deposited on the carbon steel surface by dip coating. The 3–7 μm thick coatings have consistent and defect free surfaces. The EIS studies in 3.5% NaCl solution depicted anti-corrosive barrier with an impedance modulus of 1 GΩ cm^2.

Few studies have focused on the theoretical aspects of anti-corrosion polymer/graphene nanocomposite coatings [78]. Yuan et al. [79] investigated the polyimide/graphene oxide nanocomposite by molecular dynamics simulation method. The interfacial interactions observed between the polymer and nanofiller endowed polyimide/graphene nanocomposite coating with fine corrosion resistance properties. Gupta et al. [80] used MD simulation studies for the anti-corrosion behavior of the polymer/graphene oxide coatings. In order to simulate the polymer/graphene oxide coatings in the presence of corrosive species, the simulations were carried out on a system containing 491 water molecules configurations of nanocomposite on an Fe surface (Figure 13). The authors [80] observed that the nanocomposite inhibitor was pulled down onto the metal surface and high electronic cloud density was generated. Moreover, the high surface area of the nanocomposite effectively formed the high contact area protective inhibitor film, which separated the metal surface from corrosive media and so protected it from corrosion.
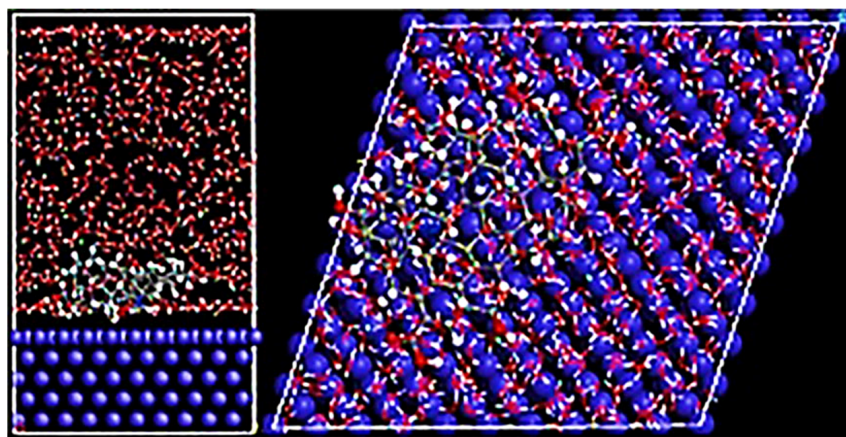
Figure 13. MD simulation: side and top views of the final adsorption of polymer/graphene oxide on iron surface [80]. Reproduced with permission from Elsevier.

Among conducting polymers, polyaniline has been used as a preferred contender in the coating application [81,82]. Kraljić et al. [83] fabricated the polyaniline and graphene based nanocomposite coating through in situ polymerization. The polyaniline/graphene coating possesses a large surface area, nanofiller dispersion, and barrier features to avoid the permeability of water and oxygen molecules through the coatings [84]. Sun and co-workers [85] formed the anti-corrosion pernigraniline/graphene nanocomposites. The coatings revealed high scratch resistance and barrier performance towards the corrosive media. Change and co-researchers [86] designed the polyaniline/graphene nanocomposite coatings for corrosion defense of steel. Figure 14 shows the chemical oxidation polymerization of aniline monomers with 4-aminobenzoyl functionalized graphene nanosheets using desired chemicals and oxidants. The polyaniline/graphene nanocomposite coatings offer fine barrier properties against O$_2$ and H$_2$O molecules. In fact, the authors [86] concluded that the fine dispersion of graphene nanosheets developed lengthened diffusion pathways for the percolating corrosive species, so preventing the erosion process.
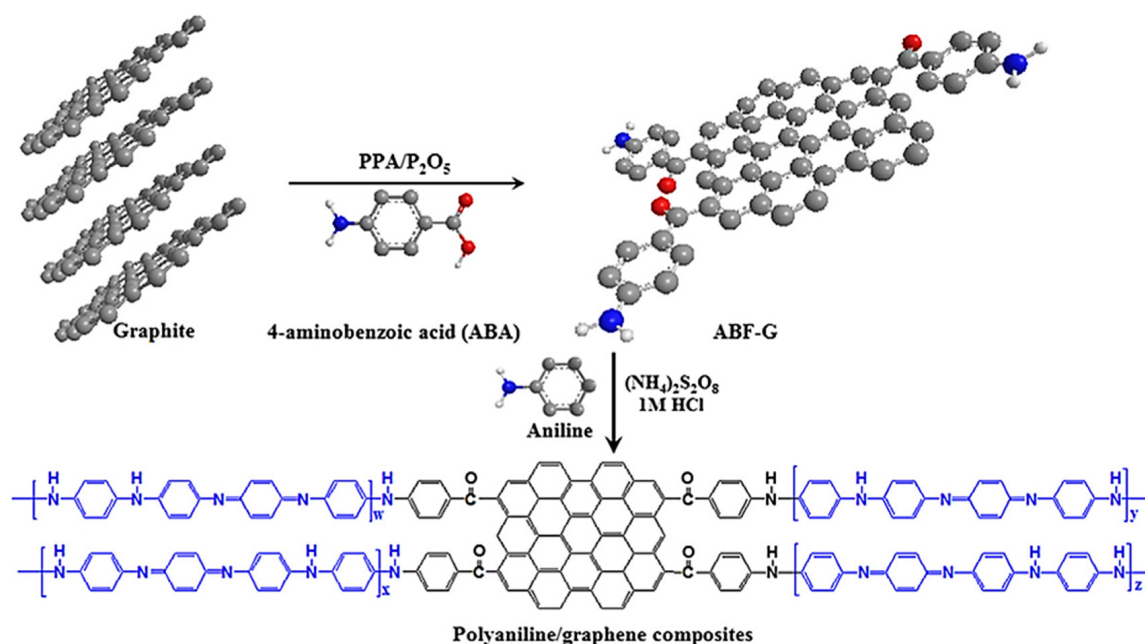
Figure 14. Preparation of polyaniline/graphene nanocomposite. PPA = polyphosphoric acid; ABF-G = 4-aminobenzoyl group-functionalized graphene-like sheets [86] Reproduced with permission from Elsevier.

Polyaniline has been blended with epoxy matrix to form the corrosion resistance coatings. Lin et al. [87] developed the epoxy coatings reinforced with poly(styrenesulfonate) polyaniline/reduced graphene oxide and reduced graphene oxide to form epoxy/reduced graphene oxide (epoxy/rGO) and epoxy/poly(styrenesulfonate)-polyaniline/reduced graphene oxide (epoxy/PSS-PANI/rGO) nanocomposites. The epoxy/rGO and epoxy/PSS-PANI/rGO nanocomposites have been tested for anti-corrosion and mechanical properties. The curing capacities of the epoxy/rGO and the epoxy/PSS-PANI/rGO nanocomposites obtained in that work [87] can be seen in Figure 15. Obviously, the curing capacity of the epoxy/PSSPANI/rGO nanocomposites was found to be higher than the epoxy/rGO nanocomposites. For example, at similar 0.1 wt.% loading, the curing capacity of the epoxy/PSS-PANI/rGO was ~97.6%, i.e., 2.5% higher than that of the epoxy/rGO nanocomposite. The mechanisms proposed for the anti-corrosion phenomenon of epoxy/rGO and epoxy/PSS-PANI/rGO nanocomposites are presented in Figure 16 [87]. The epoxy/PSS-PANI/rGO coating developed a better passivation layer, tortuosity, and interfacial matrix–nanofiller adhesion to prevent the corrosion occurrences. On the other hand, epoxy/rGO has been found least effective towards the corrosion prevention.

In this way, various modified graphene nanofillers, processing routes, and mechanisms have been stated for organic polymer based anti-corrosive coatings [88]. The anti-corrosion efficiency was found to be reliant on polymer type, graphene content, graphene functionality, dispersion, matrix–nanofiller interaction/interface formation, and adhesion to the metal substrate [89]. Finally, Table 3 presents a brief summary of the important specifications of some corrosion resistant polymer/graphene nanocomposites.
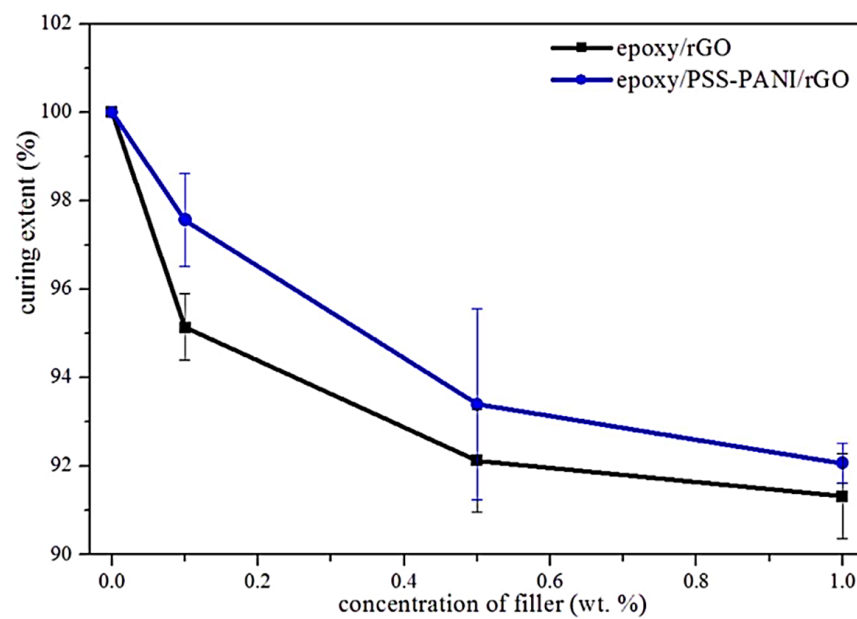
Figure 15. Curing extents of epoxy/reduced graphene oxide (epoxy/rGO) and epoxy/poly(styrenesulfonate)-polyaniline/reduced graphene oxide (epoxy/PSS-PANI/rGO) nanocomposites [87]. Reproduced with permission from Elsevier.
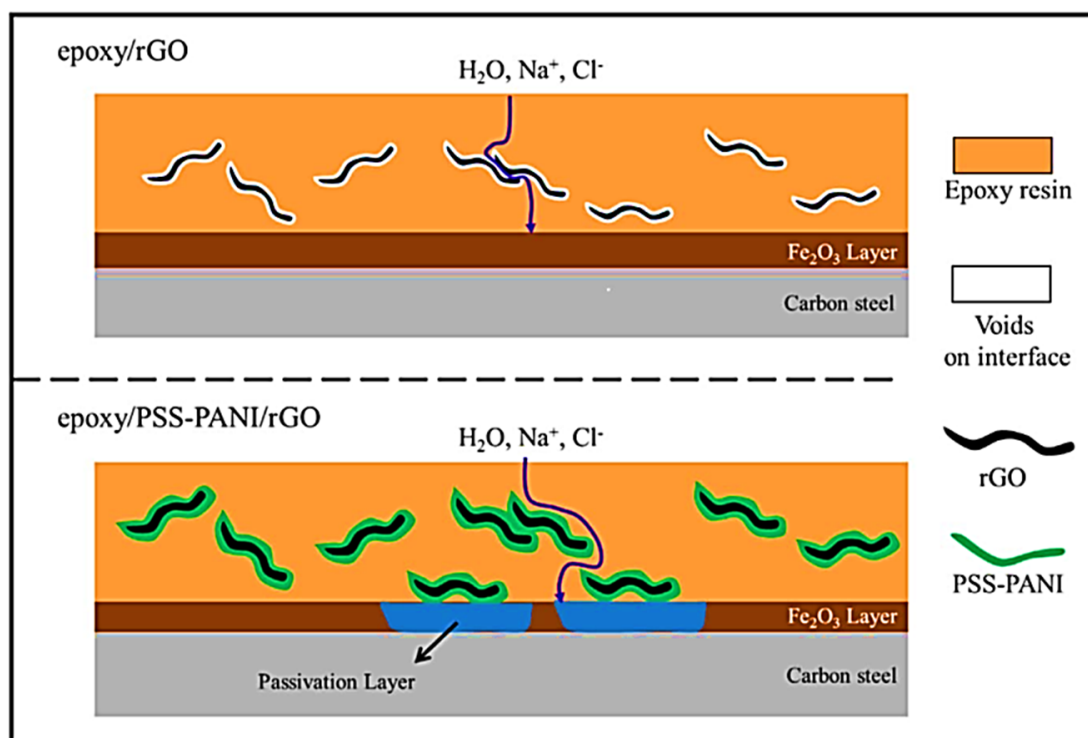
Figure 16. Schematic of proposed anti-corrosion mechanism of epoxy/reduced graphene oxide (epoxy/rGO) and epoxy/poly(styrenesulfonate)-polyaniline/reduced graphene oxide (epoxy/PSS-PANI/rGO) nanocomposite coatings [87]. Reproduced with permission from Elsevier.

Table 3. Specifications of corrosion resistant polymer/graphene nanocomposites.

Polymer	Graphene Content	Metal Substrate	Dispersion	Anti-Corrosion Features	Ref
Epoxy	Graphene 0.2–0.8 wt.%	Steel plates; good adhesion	Fine graphene dispersion; barrier effects	Corrosion current density 8.08×10^{-9} Acm^{-2}; corrosion potential -0.49 at 0.4 wt.% nanofiller	[63]
Epoxy	1 wt.% Graphene oxide prepared with different aspect ration	Q235 steel	Better graphene dispersion; no defects and pores	Coatings tested in 3.5 wt.% NaCl; Bode and Nyquist plots	[70]
Waterborne polyurethane and polytetrafluoroethylene	0.5 wt.% graphene	Stainless steel	Zigzag graphene dispersion to block diffusion path of corrosive medium through coating to reach metal/coating	EIS studies	[73]
Isocyanate crosslinked with hydroxy functional acrylic adhesive matrix	Hydrophobic graphene oxide; 0.4 mg	Copper	Electrophoretic deposition	Electrochemical studies; potentiodynamic polarization; EIS studies; electrochemical degradation; Tafel plots; corrosion current density 3.49 μA/cm^2	[74]
Polymer	Graphene oxide	Fe surface	High contact area of protective inhibitor film	MD simulation	[80]
Polyaniline	Graphene 0.1–0.5 wt.%	Steel	Graphene nanosheets dispersion; lengthened diffusion pathway for percolating corrosive species	Barrier properties against O_2 and H_2O molecules	[86]
Epoxy/poly(styrenesulfonate)-polyaniline	Reduced graphene oxide up to 0.5 wt.%	Carbon steel	Better passivation layer/tortuosity; interfacial matrix–nanofiller adhesion	Anti-corrosion mechanism	[87]

5. Significance and Challenges of Polymer/Graphene Nanocomposites in Corrosion Protection

Epoxy (thermoset) and polyurethane, poly(methyl methacrylate) and other thermoplastics have been opted as non-conjugated matrices to form the nanocomposite with graphene nanofiller [90]. In particular, in conducting polymers, graphene nanoparticles have been included as an important dopant [91]. The conducting polymer/graphene nanomaterials have been used to design the metal protection coatings for numerous industrial applications [92]. In this regard, various conducting polymers have been employed. The polymer/graphene nanocomposite coatings have the capability to prevent corrosive species like toxic chemicals, moisture, and ionic/gaseous species from percolating to the metal surface. So, the polymer/graphene nanocomposite coatings may have an efficient barrier effect towards the corrosive media [93]. The fine graphene nanofiller dispersion in the polymers has been found indispensable to improve the corrosion protection features through the formation of inter-linked percolation network [94]. On the other hand, the aggregation of graphene nanofiller and uneven dispersion may cause seeping of the corrosive molecules to the underlying metal surfaces. Consequently, anti-corrosion polymer/graphene nanocomposites have been applied in several technical fields [95]. In the electronics industry, these nanocomposites have been found valuable for corrosion resistance [96]. Similarly, in the radiation shielding devices, polymer/graphene nanocomposites have been applied to improve the device performance as well as to avoid the corrosion processes involved [97–99]. In photovoltaic devices, the polymer/graphene nanocomposites serve the same purpose [100]. The anti-corrosion polymer/graphene nanocomposites have also been found beneficial for the smart memory devices [101,102]. Other corrosion resistance applications of the polymer/graphene nanocomposite have been found in the electronic sensors, biosensors, optical sensors, etc. [103,104]. In the biomedical sector, corrosion resistant polymer/graphene nanocomposites have been preferred for the artificial muscles and tissue engineering scaffolds [105,106].

The corrosion resisting performance of the polymer/graphene nanocomposite depends on the processing technique used, nanofiller dispersion, interface formation, and matrix–nanofiller interactions [107]. Moreover, the polymer–nanofiller compatibility, percolation network formation, and adhesion to the substrate point to the anti-corrosion capabilities. In this respect, several mechanisms have been projected for the polymer/graphene nanocomposite coatings causing the corrosion protection. However, it is still challenging to develop 100% effective corrosion resistant coatings to protect the underlying metal surface. The long-term use of polymer/graphene nanocomposite may produce surface defects and fragility in the coatings. Therefore, the polymer/graphene nanocomposite coatings need to be further modified to achieve outstanding electron transportation to facilitate the formation of a chemical diffusion barrier to the corrosive media. In a nutshell, high-performance polymer/graphene nanocomposite coatings need to be designed to meet the demands and standards of heavy industries based on metals or metallic components [12].

6. Conclusions

In this state-of-the-art review, a comprehensive valuation of the design strategies, essential properties, and potential of corrosion resistance polymer/graphene nanocomposite coatings has been presented. The graphene reinforcement of the polymeric composites is actually considered as an advanced strategy towards the development of high-performance anti-corrosion coatings. The polymer/graphene nanocomposite coatings show, generically, reasonable cost, high surface area, light weight, durability, scratch resistance, barrier properties, and high electrical conductivity for effective corrosion prevention performance. Lastly, the implications of innovative corrosion-preventive polymer/graphene nanocomposites for the metallic components and their industrial applications have been analyzed.

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