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Journal of Composites and Compounds

# Application of composite conducting polymers for improving the corrosion behavior of various substrates: A Review

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ABSTRACT

One of the most important problems in the manufacturing industry is metal corrosion. Recently, conductive polymers (CPs) have attracted attention due to their economic viability and widespread industrial applications. Upon adsorption, long-chain carbon bonds of polymers provide a blockage for large surface areas of corroding metals. The adsorbed thin films create a barrier between the surrounding environment and the metal substrate. Polypyrrole (PPy), polyaniline (PANI), and polythiophene (PTh) are conducting polymers that are utilized to protect metals and metal alloys against corrosion. A proper selection of synthesis parameters for CPs can improve the anticorrosion behavior of the coatings for metals and metal alloys. This paper has an overview of conducting polymer composite coatings on substrates based on steel, copper, magnesium, aluminum, and their alloys.



Peer review under responsibility of JCC Research Group

ARTICLE INFORMATION

*Article history:*
Received 28 July 2020
Received in revised form 07 September 2020
Accepted 13 October 2020

*Keywords:*
Corrosion resistance
Polyaniline
Polypyrrole
Polythiophene
Metal substrates

Table of contents



## 1. Introduction

In the electronic and metallurgical industries, corrosion control is a challenge with great importance worldwide [1]. To protect metals from corrosion, various methods have been used. A widely practiced technique is the application of conducting polymer coatings [2, 3]. An active area of research in electrochemistry in the last decades has been the electrodeposition of CPs on the surfaces of metallic electrodes. In contrast to other coatings, such as paints, CPs do not contain toxic and hazardous constituents for the environment. Additionally, compared to other coatings that only provide physical barriers against corrosive envi-

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*DOR: 20.1001.1.26765837.2020.2.5.7.1*
*https://doi.org/10.29252/jcc.2.4.7*

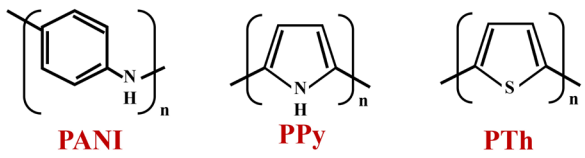


**Fig. 1.** FChemical composition of PANI, PPy, and PTh.

ronments, CPs provide physical and electronic barrier effects and electromagnetic interference (EMI) shielding which enhances the protection behavior [4-6].

These composite materials are strongly adsorbed onto active sites of the metal substrate leading to suppression of the dissolution process and production of a protective film layer. In fact, corrosion protection of CPs is a kind of anodic protection. According to studies, coating a metal with a conducting polymer places the potential of the electrode in the passive zone in the absence of redox reactions [7]. CPs have been applied on the surface of metals such as zinc [5], copper [6, 7], aluminum [8, 9], iron [10, 11], stainless steel (SS) [12], mild steel (MS) [13], etc. Mostly, polyaniline, polypyrrole, and polythiophene are used for coating metal substrates [8-19]. This review article has an overview of conducting polymers and composites and the state-of-the-art findings in the field of composite conducting polymers coated on various metal substrates are presented.

## 2.1. Conducting polymers

CPs can be used as a protective coating for the prevention of metal surface corrosion and enhancement of PE values [20]. Intrinsically conducting polymers are organic polymers with electrical conductivity. These compounds can either be semiconductors or show metallic conductivity. Their great advantage is the processability of conductive polymers, mainly by dispersion. Generally, these polymers are not thermoplastics and thereby they are not thermoformable. However, they are organic compounds like insulating polymers. Mechanical properties of conducting polymers are not similar to other commercial polymers, but they exhibit high electrical conductivity [21]. Using organic synthesis methods as well as advanced dispersion techniques, the electrical properties of these polymers can be fine-tuned [22, 23].

Different CPs are commercially available including PANI, PPy, PTh [24-26]. The chemical composition of these polymers is illustrated in Fig. 1. To synthesize CPs, electrochemical or chemical oxidation methods are used [27, 28]. PANI and its derivatives are extensively utilized for anticorrosion coatings due to facile synthesis, enhanced environmental stability, as well as various redox states that allow property regulation. Localized/delocalized polarons and bipolarons may be present in the PANI structure in various proportions, which depends on the methods of synthesis and isolation. PANI is practically applied to protect concrete steel bar reinforcement [28, 29].

Among all known CPs, one can consider PPy as promising material due to its high conductivity, easy and flexible preparation, good mechanical properties, and stability. Potential technological application of PPy include membrane separation [30], electronic and electrochromic devices [31], light-weight batteries [32], chromatographic stationary phases [33], sensors [34], and counterelectrode in electrolytic capacitors [35]. In recent years, it has been reported in several studies that PPy can protect metals and their alloys from corrosion [36-38].

An important class of conjugated polymers is PTh polymers that have a wide range of applications including field-effect transistors, electrochromic, and conducting films [39]. Few reports have demonstrated the use of PTh for the corrosion protection of metals. Among CPs, some PTh derivatives have shown good performance, which ultimately depends on the environment nature that CPs are in contact with. It is feasible to generate PTh and its derivatives on other CPs such as PPy by applying proper voltage. The combination of these two conducting polymers has led to better corrosion performance [40].

It is possible to formulate CP-based coatings to inhibit corrosion of metals even in damaged areas where the surface of the metal is directly exposed to the corrosive environment. Conducting polymers can be whether in the reduction-nonconductive state or oxidation conductive state. Under appropriate conditions, they can easily switch between the two states. Redox processes occur in CPs; therefore, the expelling/binding of dopants (counterions) is conducted in response to the metal surface potential variation. The potential variation is initiated by local electrochemical reactions resulting from the corrosion. Based on the local corrosive conditions, the dopants can be expelled or inserted by the CP, which often act as inhibitors that prevent the local corrosion reactions upon release [41, 42]. This is considered as a strategy suggested for taking advantage of CP-based corrosion-resistant coatings [43].

## 3. Corrosion protection mechanisms of CPs

For the provision of electronic conductivity in CPs, oxidative polymerization and anion doping are performed into the polymer. The penetration of aggressive anions into CP coating is prevented by controlling the doping ions. When CP-coated metal substrates are immersed in aggressive environments, such as the sodium chloride solution, the chloride anions present in the medium is exchanged with doped anions in the CP coating. The corrosion protection mechanisms of CPs have not been precisely revealed. Four possible hypotheses have been proposed:

I) Mechanism of controlled inhibitor release: In this mechanism, the anion dopant may be released upon reduction from the oxidized and hence doped CP-based coating on a metallic substrate, which is driven by a coating defect. As far as doped PANI is concerned, the anions are released either through a reduction mechanism or a simple acid-dopant elimination if it is soluble in water [44, 45].

II) Mechanism of anodic protection: according to this mechanism, protective metal oxide layers are formed on the metal surface as a result of CPs providing corrosion protection [46].

III) It is proposed that an electric field is produced when there is a contact between a doped semiconductor or a conducting polymer and a metal resulting in a reduction in the corrosion rate due to the restriction of the electron flow from the metal to an oxidizing species [47].

IV) CPs create an adherent, dense, low porosity film on the metal surface limiting the access of oxidant agents and prevent metal surface oxidation [48] (Fig 2).

A denser CP layer provides a better barrier effect and decreases the rate of $H_2O$ and $O_2$ transport into the polymer. The reaction site on which $O_2$ reduction occurs moves from the metal/CP interface to the CP/solution interface by the enhancement of the compactness of the coating and its adherence to the substrate [49]. The change in the $O_2$ reduction site on the surface of the polymer leads to a decrease in reduction products such as OH across the metal/CP interface, and thereby prevent the coating disbondment and delamination [50]. Furthermore, oxygen reduction requires the local reoxidation of the coating and its active role in the case that local small-size defects or pinholes are generated. Therefore, the improvement of the barrier effect should not inactivate CPs. The open-circuit potential of the metal/CP-based coating/solution system will be is in the passive state as far as the conducting polymer is in the conductive form. The site of the $O_2$ reduction and its kinetics are important factors to determine the prolonged protective properties of the coating. Generally, it has been reported that the barrier effect is improved by

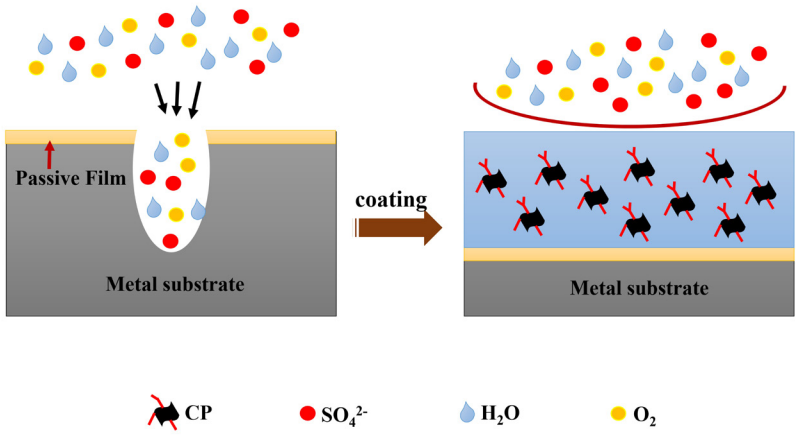


**Fig. 2.** Barrier effect of CPs for the diffusion of corrosive agents.

the dehydration of CP film electrodeposited on metal surfaces from an aqueous medium [43].

Mechanisms I and II are the most important contributing mechanisms that can rationalize corrosion inhibition by CPs. For a specific metal substrate/CP-based coating/solution system, the other two mechanisms contribute to the corrosion process simultaneously with the controlled inhibitor release or the anodic protection mechanism [10, 43].

### *3.1. Corrosion inhibitors*

Different ways of CP doping can be used for controlling the electrolytic environment near the surface of the metal substrate in case a scratch is formed. In this condition, a galvanic coupling exists between the CP coating and the metal. The anodic reaction involves the metal oxidation, while the cathodic reaction is the CP reduction resulting in the release of the doping anions. However, oxygen is reduced simultaneously on both the metal surfaces and CP coating resulting in the OH production and the CP reoxidation, respectively. Based on the nature of doping anions and the metal, a self-healing process may be triggered. In some metals such as steel, copper, and aluminum, the doping anions such as molybdates and phosphonic acid derivatives act as inhibitors, or oxide formation is initiated [43].

In the inhibition mechanism, a monomolecular barrier is formed by the organic species adsorption onto the surface of the metal. The presence of the adsorbed molecules results in the limitation of the cathodic and/or anodic corrosion reactions such as electron transfer and decreases the rate of corrosion [51, 52]. According to Brycki et al. [53], the inhibitor action involves the replacement of the adsorbed water from the surface of the metal by soluble organic species (Org):

$$Org_{sol}+nH_2O_{ads} \rightarrow Org_{ads}+nH_2O_{sol} \quad (1)$$

Several investigations have reported that monomeric aniline, as well as functionalized aniline derivatives, act as potential corrosion inhibitors for steel and iron [54-56].

### *3.2. Anodic protection*

The anodic protection activity involved the ways wherein general corrosion of the metal substrates and alloys is prevented by CPs mostly in solutions free of halides [43]. According to Kinlen et al. [57], the electroactive conducting polymer (ECPs) electrochemistry provides anodic protection for the substrate and also prevents cathodic debonding of the polymer coating. In this protection mechanism, the corrosion potential of the metal substrate in the electrolyte of interest moves to the passive region. The proposed reaction between the metal (M) and the oxidized state of the polymer coating ($ECP^{m+}$) is:

$$\frac{1}{n}M+\frac{1}{m}ECP^{m}+\frac{y}{n}H_2O \rightarrow \frac{1}{n}M(OH)_y^{(n-y)}+\frac{1}{m}ECP^{0}+\frac{y}{n}H \quad (2)$$

reoxidation of the ECP can occur by dissolved or atmospheric oxygen:

$$\frac{m}{4}O_2+\frac{m}{2}H_2O+ECP^{0} \rightarrow ECP^{m+}+mOH^{-} \quad (3)$$

## 4. Composite conducting polymers (CCPs)

Polymer nanocomposites have found increasing attention in various engineering applications [58-60]. The essential characteristic of this procedure is that CPs make it possible to maintain the substrate surface potential into a passive state wherein a protective oxide film is generated on the surface of metal substrates [43, 61]. As a result, CPs-based coatings are pinhole and defect resistant in such a way as that of the hexavalent chromium coatings. It is due to the replenishing of CP charges consumed by oxidation of metal by $O_2$ reduction within the CP coating. The corrosion process of metal is prevented by switching the CP-based coating into the oxidation state and thereby changing the potential into the passive region [62].

Expanded studies have focused on the anti-corrosive features of CPs; however, there are still numerous problems to be resolved regarding the fulfillment of mechanical and physico-electrochemical requirements of high performance anticorrosive CP-based coatings exposed to various practical conditions. Anticorrosive CP-based coatings have some limitations such as poor adhesion to the metallic substrate, anion-exchange properties, poor barrier effect due to porous structure, and irreversible consumption of stored charges within the coating, which can oxidize the substrate and form a passive oxide layer. The mentioned drawbacks show their effect more significantly under harsh environments. In case chloride ions are present, these ions can either penetrate through the CP coating or undergo anion-exchange (replacement of chlorides with CP doping anions) and reach the metal-substrate interface. Extended localized corrosion may be induced by chloride ions and during the redox reactions, the charge stored in the CP layer might be irreversibly consumed.

Using CP-based composites consisting of a conducting polymer and different inorganic fillers like metal oxides has been offered as an efficient strategy for the elimination of the disadvantages. In CP-based composites, the CP self-healing properties are combined with qualities of inorganic materials. Therefore, the composite coatings exhibit improved physicochemical and mechanical properties including enhanced hydrophobicity, barrier effect, and adhesion [43, 63-65]. The improvement of these properties leads to the enhancement of corrosion protection. Nano-

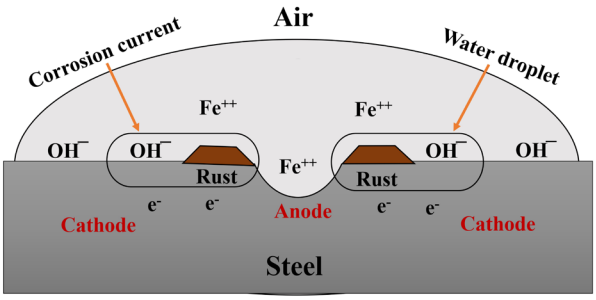


**Fig. 3.** Corrosion mechanism of steel.

technology has gained dramatic attention in recent years and is expected to make advancements in the design and development of commercially viable CP-based composite coatings [66]. It seems that CP-based nanocomposite can combine the properties of CPs and inorganic materials more effectively compared to microcomposites [67].

## 5. CCPs coatings on metals

### *5.1. CCPs coated on steels*

Structural steel is corroded through an electrochemical process in the presence of oxygen and moisture. Rust is produced by the oxidation of iron in the steel, which has a volume of six times the original material [68]. The corrosion mechanism is presented in Fig. 3. There are numerous reported focusing on the corrosion protection effect of CP coatings on metals, especially iron and mild steel, and stainless steel and significant advancements have been made [69-76]. Most conducting polymers form conducting films directly on the substrate surface by anodic oxidation [77, 78]. Changing from an insulating state to a conducting state by different doping methods including injection of charge at the interface of a metal and the conducting polymer, photo doping, electrochemical doping, and chemical doping by charge transfer [71]. Due to the capability of these polymers in charge storing and transport, they can anodically protect metals against fast corrosion [79]. The corrosion protection mechanisms of CPs are complex and affected by various parameters [80-85]. Some theories have proposed that a passive oxide film is formed on the metal surface by oxidation-reduction processes, while others have predicted that the barrier mechanism is responsible for provided protection [61, 86, 87].

There are many studies targeting to investigate the corrosion protection of steel by CPs and CCPs. Sathiyanarayanan et al. [88] synthesized the PANI-$TiO_2$ composite coating (PTC) on steel and studied its corrosion protection behavior. To prepare PTC, aniline and $TiO_2$ were chemically oxidized by ammonium persulfate in a medium containing phosphoric acid. According to the results, the redox property of PTC led to maintaining the steel potential in the passive region. The resistance of the PTC coating in a 3% NaCl solution after 60 days was more than 107 $cm^2$ and in the salt spray test for 35 days was 109 $cm^2$. However, in both cases, the resistance of the coating was less than 104 $cm^2$. It was proposed that the corrosion protection is due to the passivation of steel resulting from the presence of polyaniline. Lenz et al. [73] incorporated $TiO_2$ pigment into PPy during the electrochemical synthesis of the CP-based coating on AISI 1010 steel. Weight loss and salt spray tests demonstrated that the PPy/$TiO_2$ composite significantly increased anti-corrosion properties compared to PPy films. The composite coatings were suggested as a primary coating that can be applied on mild steel instead of phosphatized layers.

According to Radhakrishnan et al. [89], composite coatings composed of PANI and nano-$TiO_2$ prepared by in-situ polymerization on steel plates showed superior corrosion resistance than did PANI coatings in aggressive environments. It was reported that the corrosion resistance improvement for the nanocomposite coating containing 4.18 wt% $TiO_2$ nanoparticles was beyond 100 times. It was proposed that the improvement is the result of the high surface area accessible for the dopant release due to nano-size additive, redox properties of PANI, charge transport prevention by the $TiO_2$ nanoparticles, and an increase in diffusion barrier. In a research study by Patil et al. [90], polyvinyl acetate (PVAc)-ZnO-PANI hybrid composite coatings (PVAc as the major matrix) were deposited on steel plates by the dip-coating method. In comparison with the coatings that contained either ZnO or PANI, the coatings that contained both the components exhibited higher corrosion resistance. The PVAc-ZnO-PANI coating showed the $I_{corr}$ value of two-order lower than that of PVAc–ZnO and PVAc coatings. The improvement was reported to be the result of the redox behavior of PANI, enhancement of barrier properties by nanoparticles, as well as the formation of protective oxide layers near the substrate. Hosseini et al. [91] electrodeposited the polypyrrole phosphate (PPy-P) coating by cyclic voltammetry (CV) method on ST12 mild steel. The deposited PPy–P films demonstrated higher corrosion resistance compared to the PPy coating.

To coat 304 stainless steel for bipolar plates used in a proton exchange membrane fuel cell, Ren et al. [92] used galvanostatic deposition to produce an inner layer of PPy with large groups of dodecylsulfate ions, and then a PANI external layer containing small groups of $SO_4^{2-}$ was applied via cyclic voltammetric deposition. According to results, the increase in pitting corrosion potential and corrosion potential of the bare steel for the single PPy and PPy/PANI coatings was more than 500 mV and 400 mV (saturated calomel electrode), respectively. Compared to the single PPy coating, the bilayer composite coating showed more effective corrosion reduction through providing passivity protection as well as a physical barrier with acceptable contact resistance.

Jiang et al. [46] deposited PPy-graphene oxide (GO) composite coatings on 304 stainless steel bipolar plates by in-situ electrodeposition to protect them against aggressive environments. The analysis in the simulated PEMFC environment exhibited that during potentiostatic polarization, the polarization current density of the substrate was significantly reduced by the conductive PPy-GO composite coating. The addition of GO to the PPy matrix led to the enhancement of the adhesion strength and an increase in the diffusion pathway of corrosive agents and therefore, restriction of their inward penetration. The best corrosion resistance was obtained for the composite coating containing 1 mg $mL^{-1}$ of GO in the electrodeposition electrolyte. The corrosion enhancement in the composites is the result of the improved anodic protection and physical barrier. Jadhave et al. [93] added poly-o-anisidine (POA) and PANI nanoparticles to alkyd paint formulation for protecting the mild steel surface. In comparison with the POA/alkyd coatings, corrosion protection of the PANI/Alkyd coatings was remarkably higher.

Epoxy/graphene composite coatings with hydrophobic surfaces were prepared by Chang et al. [94]. The water droplet's contact angle with the epoxy surface and hydrophobic epoxy/graphene surface were 82° and 127°, respectively. The improvement of the corrosion resistance by applying the composite coating was reported to be due to the physical barrier effect, a decrease in the adsorption of water/corrosive media resulting from the coating hydrophobicity, and high aspect ratio of graphene nanosheets leading to enhancement of the oxygen barrier property. Sumi et al. [95] synthesized PANI-$Fe_2O_3$ composite by an *In-situ* method and added it to a commercial alkyd resin as an anti-corrosive coating for mild steel. The composite coating was proposed to offer passivation protection and better barrier performance. The complimentary cathodic reaction of the nonconductive leuco-PANI to the conductive emeraldine-PANI was explained to be also responsible for the improved corrosion resistance in the acidic medium. Table 1 summarized research

**Table 1.**
Research reports on using CCPs for corrosion protection of steel

| Authors | CPs | Additive | Coating technique | Medium | Corrosion behavior |
|---|---|---|---|---|---|
| Jadhav et al. (2020) [96] | PPy | $Fe_2O_3$ | Electrochemical method | NaCl | Better corrosion resistance was observed by the coating of $Fe_2O_3$/PPy. |
| Sun et al. (2020) [97] | PANI | - | Electrochemical deposition | NaCl | The density of corrosion current decreased 5 times and the coating exhibited effective protection for 140 days. |
| Deyab et al. (2020) [98] | PANI | Zn-Porphyrin | Electrochemical deposition | $H_2SO_4$ | The composite of PANI/Zn-Pr with 1.0% of Zn- Pr rendered the highest anti-corrosion activity (99.41%). |
| Chen et al. (2020) [24] | PPy | Polydopamine –functionalized carbon powders | Electropolymerization | $H_2SO_4$ | The PPy/C-PDA coating showed good protection performance for the 304SS bipolar plate in PEMFC. |
| Rajkumar et al. (2020) [99] | PPy | $TiO_2$, ZnO, and $SiO_2$ | Incorporation in resin | NaCl | The PPy coating provides the denser passivation film at the interface of PPy and $TiO_2$. |
| Chen et al. (2019) [100] | PPy | $TiO_2$ and V-$TiO_2$ | Electrochemical method | HCl | comparing V-$TiO_2$/PPy and $TiO_2$/PP composite coatings, the V-$TiO_2$/PPy showed better corrosion resistance performance. |
| Kong et al. (2019) [101] | PANI | Chitosan | - | HCl | Using the PANI/CTS in 0.5 M HCl solution was effective for corrosion protection of Q235 steel and at high PANI/CTS concentrations, the highest inhibition efficiency was obtained. |
| Babaei-Sati et al. (2019) [102] | PPy | $Al_2O_3$ | Electrodeposition | $H_2SO_4$ | PPy/$Al_2O_3$ nanocomposite with declining the density of corrosion current by 18 times, exhibited excellent performance in the protection of MS. |
| Shi et al. (2019) [26] | PANI | SiO2 | Drop casting technique | $H_2SO_4$ | The silicone-$SiO_2$@PANI coating with a 4:1 weight ratio of $SiO_2$/PANI exhibited the highest resistance against corrosion (2.24×107 Ω $cm^2$) after immersion in a corrosive medium for about 180 days. |
| Jaouhari et al. (2019) [103] | PPy | Zinc phosphate | Galvanostatic electrodeposition | NaCl | The ZP/PPy coatings showed excellent corrosion resistance and increased the ZP/PPy coating thickness. |
| Liu et al. (2019) [104] | PANI | $TiO_2$ | Electrochemical deposition | NaCl | The epoxy coating with $TiO_2$/PANI particles showed high corrosion protection compared to the blank coating after subjecting to a corrosive environment. |
| Wang et al. (2019) [38] | PANI | Nb: $TiO_2$ nanofibers | galvanostatic method | HCl | The presence of Nb: $TiO_2$ nanofibers in the coating of PANI led to the provision of better in-situ anodic protection and physical barrier effect. |
| Abd El-Lateef et al. (2019) [105] | PANI | $Tl_2O_3$-$SiO_2$ | Electrochemical deposition | HCl | The PANI coating could prevent the carbon steel corrosion and provide maximum yielding of 89% and this amount after the modification with $Tl_2O_3$-$SiO_2$ nanocomposites was improved and reached 96%. |
| Ramezanzadeh et al. (2018) [83] | PANI | GO-$CeO_2$ | Electrodeposition | NaCl | The deposition of $CeO_2$ and PAni improved the properties of active and barrier corrosion inhibition of GO nanosheets. |
| Contri et al. (2018) [106] | PPy | Montmorillonite (Mt) | Electrodeposition | $H_2SO_4$ | The Epoxy/Mt-PPy (5 wt%) could prevent carbon steel corrosion. |
| Jadhav et al. (2018) [107] | PPy | Mica | Incorporation in resin | NaCl | The pigment-based composite coating of Mo-doped PPy/mica exhibited better protection against corrosion with the steel passivation by the anions of molybdate. |
| Salem et al. (2018) [108] | PANI | - | Electrochemical deposition | NaCl | The possibility of delamination and blister formations were reduced by composite coatings. |
| Jiang et al. (2018) [109] | PANI | $Ni(OH)_2$ | Cyclic voltammetry technique | NaCl | The $Ni(OH)_2$ particle deposition in a matrix of PANI prevented access to aggressive media. Also exhibited long-term anti-corrosive behavior. |
| Arabzadeh et al. (2017) [110] | PPy | - | Cyclic voltammetry method | HCl | The sample synthesized with the scan rate of polymerization equal to100 mV/s was the best coating. |
| Ladan et al. (2017) [111] | PPy | $TiO_2$ | Dip coating | NaCl | Co doping $TiO_2$/PPy decreased the charge transfer across the interface of electrolyte/AISI 1018 steel. |
| Yan et al. (2017) [112] | PPy | $Al_2O_3$ | Cyclic voltammetry technique | NaCl | The PPy-$Al_2O_3$ composite coating exhibited good performance in the corrosion protection of 316SS. |
| Yan et al. (2017) [113] | PPy | $SiO_2$ | Cyclic voltammetry technique | NaCl | The PPy-$SiO_2$ coating exhibited good performance in the corrosion protection of 316SS. |
| Qiu et al. (2017) [114] | PANI | GO | Pulse-current deposition | Phosphate buffer | The 98.4% corrosion inhibition efficiency and 99.3% protection efficiency was obtained by using the PANI-GO composite coating. |

investigation on using CCPs for corrosion protection of steel substrates.

*5.2. CCPs coated on magnesium and its alloys*

Because of biocompatibility, easy biodegradation, and excellent mechanical properties, Mg alloys have been extensively investigated for biomedical applications. Nevertheless, in a physiological environment, these alloys show a high corrosion rate leading to an increase in the pH value, which adversely affects cell differentiation, proliferation, and viability on the implant surface and thereby induces blood clots together with chronic tissue inflammatory responses [115-119]. Two main strategies for the improvement of the corrosion resistance of Mg and its alloys are surface modification and alloying [120, 121]. CPs and CP-based composites have been developed for the corrosion reduction of Mg-based substrates. The corrosion mechanism of Mg with and without CP coatings is shown in Fig. 4. PANI-$TiO_2$ composites were deposited on the ZM 21 alloy by Sathiyanarayanan et al. [122]. To synthesize the coatings, aniline underwent oxidative polymerization in phosphoric acid with $(NH_4)_2S_2O_8$, in the presence of $TiO_2$. Compared to the PANI coating, the composite coating exhibited more effective protection performance as a coating for the ZM 21 alloy. In another research, Guo et al. [123] applied a composite coating of PPy/ZnO to protect biodegradable Mg alloys for orthopedic implant applications. Results indicated the improved corrosion protection, antibacterial property, as well as cytocompatibility of the composite coating suggesting it as proper material for orthopedic implants.

Wang et al. [124] presented the corrosion protection performance of composite coatings based on PANI and coal. PANI/coal powder was synthesized by *in situ* polymerization, the coatings were composed of epoxy, and PANI/coal was deposited on the surfaces of Mg alloys. A significant decrease in the corrosion rate and corrosion current density of the PANI/coal coatings was observed suggesting that the coating is a promising candidate for the enhancement of corrosion resistance of Mg alloys in aggressive environments. In a research study by Li et al. [125], a PPy/$V_2O_5$ composite film was deposited on magnesium by mild vapor phase polymerization (VPP) technique. Corrosion investigations in 3.5 wt% NaCl revealed that the prepared composite film reduced the corrosion rate of Mg. The VPP method was offered as a technique with great potential to synthesize CP-based coating for the protection of reactive metals. Table 2 summarized research investigation on using CCPs for corrosion protection of Mg-based substrates.

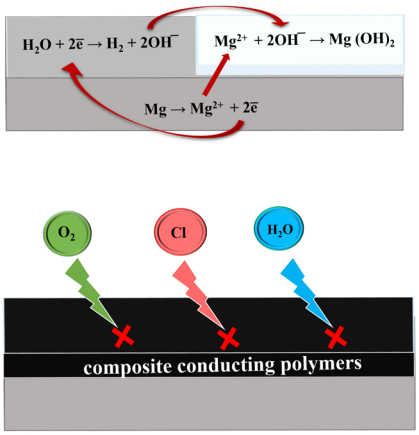


**Fig. 4.** Corrosion mechanism of brae Mg and CP-coated Mg.

*5.3. CCPs coated on alumnum and its alloys*

Al is an important metal due to its high technological value and its application in the household and aerospace industries [134-137]. Al-

**Table 2.**
Research reports on using CCPs for corrosion protection of Mg and its alloys

| Authors | CPs | Additive | Coating technique | Medium | Corrosion behavior |
|---|---|---|---|---|---|
| Najibzad et al. (2020) [126] | PANI | Praseodymium | Dip coating | NaCl | The improvement in the performance was observed with applying 2000 ppm concentration compared to other concentrations. |
| Guo et al. (2020) [123] | PPy | $ZnO_2$ | Cyclic voltammetry technique | NaCl | An increase in the resistance of the corrosion was observed. |
| Jothi et al. (2020) [127] | PPy | Gelatin | Electrodeposition | NaCl | The coating exhibited good performance in providing the corrosion resistance of AZ31. |
| Samadi et al. (2020) [128] | PANI | Praseodymium | Electrochemical methods | NaCl | The composite of PANI/Pr31 that exhibits anti-corrosion behavior can be used as environmentally-friendly and non-toxic corrosion protective coating. |
| Li et al. (2020) [125] | PPy | $V_2O_5$ | Vapor phase polymerization (VPP) | NaCl | For the synthesis of the protective coating of CPs on reactive metals, the method of mild VPP may be effective. |
| Maurya et al. (2019) [129] | PANI | Graphene | Incorporation as pigments in epoxy resin | NaCl | The amount of the resistance value $>10^6$ Ω $cm^2$ was estimated using the composite coatings. |
| Yufeng Li et al. (2018) [130] | PANI | $SiO_2$ | Electrochemical methods | NaCl | For the Mg-Li alloy, the good resistance was obtained with the coating and the density of the corrosion current and impedance value was $6.7\times10^{-7}$ A $cm^{-2}$ and $5\times10^4$ Ω $cm^2$, respectively. |
| Gao et al. (2018) [131] | PANI | - | Electrochemical methods | NaCl | The improvement was observed with the PANI-PhA addition because of the synergistic effect of silane, PhA, and PANI. |
| Wang et al. (2017) [124] | PANI | Coal | Electrodeposition | NaCl | This sample exhibited excellent resistance to corrosion. |
| Saremi et al. (2016) [132] | PPy | NaF and polyethylene glycol (PEG) | Cyclic voltammetry technique | NaCl | The observed improvement in the corrosion behavior of the PPy coating with PEG and NaF was due to the inhibition fluoride effect, which is considered a barrier for magnesium alloys. |
| Chen et al. (2010) [133] | PANI | $SiO_2$ | Electrochemical methods | NaCl | In the solution of 3.0 wt% NaCl, the coating of PANI-$SiO_2$ showed better performance in keeping the potential values in the noble potential compared to the coating of pure epoxy. |
| Sathiyanarayanan. (2007) [122] | PANI | $TiO_2$ | Electrodeposition | NaCl | For the protection of the Mg ZM 21 alloy, the composite coating was better than Polyaniline coating. |

**Table 3.**
Research reports on using CCPs for corrosion protection of Al and its alloys

| Authors | CPs | Additives | Coating technique | Medium | Corrosion behavior |
|---|---|---|---|---|---|
| Tomaev et al. (2019) [146] | PPy | Aluminum Oxide | Galvanostatic | $H_2SO_4$ | The improvement in the electrochemical potential was obtained by PP coating, and the incensement in the surface impedance was provided by oxide coating. |
| Kumar et al. (2017) [147] | PPy | $CeO_2$ | Galvanostatic | NaCl | For the corrosion protection of the Al in aircraft infrastructures, the nanocomposites of PPy with nanoparticles of $CeO_2$ could be effective. |
| Hosseini et al. (2017) [144] | PPy | $TiO_2$, $Mn_2O_3$, and ZnO | Cyclic voltammetry technique | oxalic acid | An excellent improvement in the corrosion protection was observed by applying the synthesized polypyrrole with nanoparticles of $TiO_2$. |
| Hussein et al. (2016) [143] | PANI / PPy | CNT and $Ni_2LaO_4$ | Cyclic voltammetry technique | oxalic acid | The barrier effect increased with the nanoparticles of NiLa oxide. Also, the reaction of oxygen reduction catalyzed by these particles led to improving the Al passive state. |
| Ates et al. (2015) [148] | PANI | $TiO_2$, Ag, and Zn | Cyclic voltammetry method | NaCl | The nanocomposite film of PANI/Ag exhibited the highest efficiency of protection (PE = 97.54%). |
| Ates et al. (2015) [149] | PANI / PPy | $TiO_2$ | Cyclic voltammetry | oxalic acid | According to the results, the efficiency of corrosion protection of the nanocomposites coated on the electrode of Al1050 was larger compared to PPy (94.9 %), PANI (96.4 %), and uncoated Al1050 electrodes. |
| Alvi et al. (2015) [150] | PANI | ZnO | Cyclic voltammetry method | HCl | Due to the electronic properties and chain conformation of the ZnO-PANI, it provided excellent protection against corrosion for Al and steel. |
| Jensen et al. (2014) [151] | PPy | Aluminum flake | Electrochemical methods | KCl | The composite coatings exhibited the reduction of dissolved oxygen over the scribe with no corrosion product concomitant buildup. |
| Gupta et al. (2013) [152] | PANI | Lignosulfonate | - | NaCl | The low corrosion amount was obtained with the coating of 5 wt% Pani-LGS/epoxy. |
| Jadhav et al. (2013) [153] | PPy | Aluminum flake | Incorporation in epoxy resin | Electrolyte solution | For the larger defect protection of the AA 2024-T3 substrate, the composite of the wire PPy/Al flake was effective. |
| Yan et al. (2013) [142] | PPy | Al flake | Incorporation in epoxy resin | DHS solution | The best performance of the protection was obtained by doping the vanadate in the composite coating. |
| Shabani et al. (2011) [154] | PANI | Montmorillonite | Electrosynthesis | NaCl | Using nanocomposite-coated compared to uncoated Al led to a decrease in the amount of the corrosion current ($i_{corr}$) from 6.55 μA $cm^{-2}$ to 0.102 μA $cm^{-2}$. |
| Hosseini et al. (2011) [134] | PANI | Montmorillonite | Electrochemical methods | NaCl | Epoxy blend with polyaniline and MMT showed the highest corrosion protection for 100h. |
| Castagno et al. (2010) [155] | PPy | Montmorillonite (MT) | Electrochemical techniques | NaCl | The PPy/MT films with 1% clay provided good performance in the protection of corrosion for Al. |
| Hosseini et al. (2009) [156] | PPy | Montmorillonite | Electrochemical methods | NaCl | The coating provided good protection of Al corrosion with a combination of epoxyblend with MT and PPy advantages. |
| Wu et al. (2007) [157] | PANI | Silicate–NiZn ferrite | Electrochemical and salt-spray | NaCl | With the incorporation of the NiZn ferrite/PANI particles, the denser configuration of the ormosil hybrids was obtained which could prevent the Al alloy substrate corrosion. |
| Shah et al. (2001) [145] | PANI/ PPy | - | Galvanostatic and potentiostatic technique | oxalic acid | The low corrosion rates were observed in moderate to high applied electrochemical current densities. |

though on the surface of reactive metals such as Al, a thin oxide film is formed protecting them from further corrosion, localized corrosion occurs on the surface of Al when it is exposed to corrosive environments containing complexing agents such as halides [138-141]. There have been several studies regarding the deposition of CP-based coatings on Al-based substrates to enhance their corrosion resistance. In a study by Yan et al. [142], PPy was first deposited on Al flakes in the presence of inhibiting dopants including vanadate, molybdate, or phosphate oxyanions. Then, the modified Al flakes were added to an epoxy primer to protect the AA 2024-T3 alloy. The composite coating showed good protection performance for the Al alloy through the mechanism of oxygen scavenger protection provided by PPy in the composite coating.

Hussein et al. [143] used the cyclic voltammetry technique to deposit PANI-NiLa and PPy-carbon nanotubes (CNTs) nanocomposite coating on aluminum. The thermal stability of PPy was enhanced by the addition of CNTs, while decreased in the presence of NiLa. The addition of CNTs and NiLa particles improved the protection role and adhesion of the PPy coating for aluminum. Compared to the PPy layer, the nanocomposite coatings had higher protection property for Al in the NaCl solution. PPy-NiLa nanocomposite coating demonstrated the highest corrosion protection. In another study by Hosseini et al. [144], ZnO, $Mn_2O_3$, and $TiO_2$, nanoparticles were dispersed in PPy by in-situ electropolymerization to

protect Al electrodes. It was found that the corrosion resistance of the nanocomposites was higher than that of bare PPy in harsh environments. The PPy/$TiO_2$ composite coating exhibited a remarkable improvement in corrosion protection. The great enhancement of protection properties was reported to be due to the high surface area of nano-additives for the dopant release, charge transport prevention by the $TiO_2$ nanoparticles, redox properties of PPy together with increased barrier effect to diffusion.

Kunal et al. [145] coated Al-2024-T3 substrate with PPy, PANI, and PPy/PANI composites via potentiostatic and galvanostatic techniques. Results showed that the corrosion rate reduction in the presence of CPs was about three orders of magnitude. Deposition time and applied current density as electrochemical processing variables were found to noticeably affect the corrosion behavior of the coated substrate so that low corrosion rates were achieved by applying moderate to high current densities. Table 3 summarized research investigation on using CCPs for corrosion protection of Al-based substrates.

*5.4. CCPs coated on copper and its alloys*

Cu is used in industrial and technological applications on a large scale because of its outstanding processability, thermal and electrical conductivity, wear and shock resistance, and ductility. Cu is the best selection for integrates circuits, especially microprocessors due to its improved electromigration performance as well as low resistivity [158-162]. Under neutral pH conditions, protective oxide or hydroxide layers form on the surface of Cu substrates [163-167]. In chloride-containing environments, the copper corrosion process and the protective layer formation are more complex [168, 169]. In oxidative environments, the mechanism of corrosion for copper involves the electrochemical reduction of water and oxygen at local cathodic zones and the dissolution of Cu at local anodic zones. The rates of reduction and dissolution reactions are slowed down by the formation of the protective film formation, and the diffusion rate of Cu chloride ions into the chloride solution influences the rate of these reactions. However, the diffusion and reduction of corrosive species like oxygen cannot be prohibited by the oxides or hydroxide layers [170, 171].

The enhancement of the corrosion resistance of copper has been an attractive topic for researchers [172, 173]. Applying CPs on copper substrates and their corrosion behaviors have been reported in the literature [174-182]. Beikmohammadi et al. [37] used the in situ electropolymerization technique to deposit PPy/$TiO_2$ composite coating on copper electrodes. It was proved that the addition of $TiO_2$ nanoparticles promoted the corrosion protection behavior of the coating compared to bare PPy in a harsh environment. As reported for the similar coatings for other metals, an increment of barrier to diffusion, charge transport prevention by the $TiO_2$ particles, high surface area of the titanium oxide nanoparticles for the dopant liberation, as well as redox properties of polypyrrole are responsible for the improvement.

Pan et al. [183] used Cu as substrate and electrochemically synthesized the conductive composite coating consisting of an outer PANI layer and an inner PPy layer. They found that the corrosion potential of Cu substrate increased via both the single PPy coating and the bilayered PPy/PANI. In addition, the corrosion current density decreased by an order of magnitude compared to uncoated Cu substrate. They also evaluated the Long-term protection of the coatings. It was shown that the PPy/PANI bilayer coating was better than the single polypyrrole coating that can be an effective physical barrier for inhibiting the penetration of corrosive species.

In another study, Çakmakcı et al. [184] fabricated the poly(pyrrole)/

**Table 4.**
Research reports on using CCPs for corrosion protection of Cu and its alloys

| Authors | CPs | Additive | Coating technique | Medium | Corrosion behavior |
|---|---|---|---|---|---|
| Badi et al. (2020) [94] | PANI | Silver nanoparticles | Electrochemical methods | HCl | The coating containing the nanoparticles of PANI-Ag exhibited corrosion protection for 6061 Al alloys used in solar panel frames. |
| Wan et al (2019) [189] | PPy | Benzotriazole (BTA) or/ and silica | Cyclic voltammetry technique | NaCl | The composite film exhibited good performance in corrosion protection due to the synergetic effect of silica physical barrier and BTA active protection. |
| Beikmohammadi et al. (2018) [37] | PPy | $TiO_2$ | Cyclic voltammetry technique | NaCl | Nanoparticles of $TiO_2$ exhibited good performance in the improvement of polypyrrole films for the protection of copper. |
| Jafari et al. (2016) [190] | PPy | Graphene | Cyclic voltammetry technique | $H_2SO_4$ | The number of polymer pores decreased and the nanocomposite morphology after immersion in NaCl solution at a concentration of 5000 ppm for 2 hours, remained constant and unchanged. |
| Shabani et al. (2015) [191] | PPy | Zeolite | Electrodeposition | NaCl | By using this coating the corrosion current density declined and reached 0.34 μA $cm^{-2}$ and also the potential of corrosion shifted from −0.314 V to −0.141 V. |
| Pan et al. (2015) [183] | PPy/ PANI | - | Cyclic voltammetric and galvanostatic | acidic medium | The PPy/PANI bilayered coating provided better protection for the copper substrate than the PPy coating. |
| Davoodi et al. (2015) [192] | PPy | Multi-walled carbon nanotubes | Cyclic voltammetry technique | NaCl | Higher protection of corrosion was obtained by using the nanocomposite of PPy/functionalized MWCNT compared to PPy/MWCNT. |
| Dhibar et al. (2013) [193] | PANI | - | Electrochemical methods | HCl | The promising electrochemical properties were exhibited with doping of 2 wt% PANI. |
| Ozkazanc et al. (2013) [209] | PPy | Zinc and nickel | Electrodeposited | $H_2SO_4$ | The protection degree for electrodes of copper was enhanced. |
| K. Wu et al. (2009) [93] | PANI | Silicate/carbon black | Electrochemical methods | NaCl | The resistance of corrosion and barrier properties were enhanced by using the system of PANI/CB. |

poly(N methyl pyrrole) bilayer and poly(pyrrole-co-N-methyl pyrrole) copolymer composites via electrochemical synthesis. They applied them on Cu substrate through cyclic voltammetry from an aqueous solution of 0.1 M monomer and 0.3 M oxalic acid. They suggested that the monomer feed ratio strongly affects the performance of coatings, in which the most protective property was illustrated by copolymer fabricated with 8:2 concentration ratio. Electrochemical impedance spectroscopy and anodic polarization using 0.1 M $H_2SO_4$ solution were employed to evaluate the corrosion behavior of polymer composites. They implied that the bilayer and copolymer coatings had a higher protection effect than that of single PPy coatings.

Branzoi et al. [185] investigated the electropolymerized monolayer poly (N, N' dimethylaniline) (PNDMA), bilayer PNDMA/PANI, polyaniline (PANI), PANI/PNDMA coatings on Cu substrate.

They found that good corrosion protection was obtained by PNDMA-SDS/PANI coatings in aggressive media. In addition, better corrosion inhibition efficiencies were observed for bilayer coatings.

Singh et al. [186] used electrophoretic deposition (EPD), as a less time-consuming, inexpensive, and fairly facile method, to fabricate hydrophobic graphene oxide-polymer composite (GOPC) on copper. The efficacy of the coating under stringent environmental conditions was investigated via EIS and potentiodynamic polarization investigation. They implied that electrochemical degradation of the bare copper substrate was three orders of magnitude higher than GOPC coating. They realized that the GOPC coatings were impermeable to ion diffusion of corrosive liquid solution and oxidizing gas.

In another study, Kim et al. [187] fabricated graphene/polysiloxane (PSX) nanocomposite films possessing superior corrosion protection, high electrical, and dual function. A facile bar coating method using a metering rod was employed for the better in-plane ordering of filler networks in the coating. It was found that PSX-G composite coating films improved the charge transfer resistance dramatically (20,000%), higher electrical conductivity (1700 $Sm^{-1}$), and decreased rate of corrosion (1/40 th). This was due to complementary effects between the covering agent of graphene defects and inorganic polymer matrix as the anticorrosive layer as well as graphene conductive filler. They implied that the system could be potentially employed in industrial fields including energy storage systems, electromagnetic shielding (EMI), and anti-icing.

Singh et al. [188] applied a cathodic electrophoretic deposition (EPD) technique to fabricate anticorrosive graphene reinforced composite coating. They implied that the Cu substrate became resistant to electrochemical degradation by applying the composite coating. In this regard, the Tafel analysis showed that composite coating reduced the corrosion rate about an order of magnitude lower than that of bare substrate. Table 4 summarizes the studies focusing on the application of CCPs for corrosion protection of steel, Cu, Al, and Mg.

## 6. Conclusions and future insights

CCPs have been widely investigated for the protection of metal substrates such as steel, Al, Cu, and Mg. PANI, PPy, and PTh are common conducting polymers that have been developed as protective coatings for metals. Composite conducting polymers have been prepared with the incorporation of different components such as $ZnO_2$, $TiO_2$, NiLa, $Mn_2O_3$, etc. Corrosion inhibiting and anodic protection is the most important contributing mechanisms to the reduction of the corrosion rate of metals. It has been demonstrated that CCPs have superior corrosion protection properties than do conducting polymer coatings. This is the result of the high surface area of nano-additives for the dopant release, and the promotion of barrier effect against diffusion. It is expected that in future investigations, a variety of reinforcements will be at the center of attention and more focus will be placed on the application of CCPs on other metallic substrates and in different fields. Moreover, since the protection against corrosion by CPs is mostly based on the mechanism of anodic protection, the stabilization of the passive oxide film under the polymer coating and inhibition of the aggressive anions from penetration into the polymer film must be carefully considered.

## Acknowledgments

The authors received no financial support for the research, author-ship and/or publication of this article.

## Conflict of interest

The authors declare that there is no conflict of interest.

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