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Recent progress in materials used towards corrosion protection of Mg and its alloys

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ABSTRACT

Magnesium has little resistance to corrosion and therefore its production and use are quite limited. The problem of corrosion associated with these alloys has been alleviated to some extent by the advantages obtained from fine coatings. An additional dense barrier against corrosion is created, using coatings obtained from sol-gel. As an alternative for Cr-based conversion coatings, rare-earth elements-based ones are been increasingly investigated for Mg and its alloys due to being eco-friendly. Because of chemical inertness, low friction, and high hardness, diamond-like carbon (DLC) coatings have exhibited the best protection for Mg and its alloys. In this review, we shed light on recent advancements in novel coatings for Mg alloys including hybrid, rare-earth conversion, composite polymeric (polymer composite is a multi-phase material in which reinforcing fillers are integrated with a polymer matrix), and DLC coatings.

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1. Introduction

The earth's 8th most abundant element is Mg, which makes up about 0.13% of the mass of oceans and 1.93% of the mass of the earth's crust. Owing to some favorable advantages of Mg, it is considered a promising metal for various applications. Mg has a high value of strength to weight ratio, which is 1/4 of that of iron and 2/3 of that of aluminum. Other properties of Mg include good dimensional stability, acceptable machinability, great thermal conductivity, good electromagnetic shielding, high damping characteristics, as well as recyclability. Because of these characteristics, Mg is used in different applications such as computer and automobile parts, household equipment, handheld tools, sporting goods, mobile phones, and aerospace components. Inherent biocompati-

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bility and low weight of Mg made it also a suitable material for implant applications [1-4].

However, extensive use of Mg in many applications is hindered because of several undesirable features including high chemical reactivity, weak corrosion resistance, weak creep resistance, as well as poor resistance to wear. The poor corrosion resistance is the main challenge limiting the use of Mg [5, 6]. Coating the base metal is considered an effective solution for the prevention of corrosion [7, 8]. The protection of the substrate is provided by the barrier formed between the metallic substrate and the surrounding environment and/or through some chemicals that act as corrosion inhibitors [9, 10]. The coating properties for rendering satisfying corrosion protection include uniformity, good adherence, being pore-free, and having self-healing ability for the occurrence of physical damage is possible [1].

The development of sol-gel coatings based on organic-inorganic hybrids with a low thickness in the range of 0.2-10 μm has been an appealing method for alleviating corrosion problems [11, 12]. Sol-gel coatings enhance the adherence of the organic paint system to the metal surface; therefore, they create additional dense barriers for corrosive substances. Another practiced coating is rare-earth conversion coating. This process is known for its simple electrolytic substances that usually contain chloride, sulfate, and nitrate of rare-earth metals such as neodymium, lanthanum, and cerium. Because of the simple electrolyte, its maintenance and recycling are easy. Another important issue is that these coatings are eco-friendly surface treatments [13-15].

Recently, composite polymer coatings and their performance in the corrosion protection of Mg have attracted the attention of many researchers. The good corrosion resistance of super-dispersed polytetrafluoroethylene (SPTFE) [16-18], a composite of diethylenetriamine, polyetherimide, and hydroxyapatite [19], and composites graphene oxide (GO)-containing 4-ethylene dioxythiophene (PEDOT) has been reported for the protection of Mg and its alloys. Coatings based on diamond-like carbon (DLC) are also among recently-developed coatings for the protection of Mg alloys owing to their chemical inertness, low friction, and high hardness [20]. An interlayer metallic film such as Si or Cr is usually used between the substrate and DLC film for the enhancement of coating adhesion [21, 22]. In this article, new coating materials for the protection of magnesium and its alloys including rare earth conversion coatings, hybrid coatings, DLC coating, and composite polymeric coatings, have been discussed and recent advancements in these fields have been reviewed.

2. Mg and its alloys

Among engineering metals, Mg is the lightest metal and its density is 1.74 g.cm^{-3} [23, 24]. Its density is about four times less than steel (7.86 g.cm^{-3}) and 35% less than Al (2.7 g.cm^{-3}) [25-28]. Mg metal is obtained by either the electrolysis of seawater magnesium chloride melts or magnesium oxide reduction with silicon. About 1.3 kg magnesium is founded in 1 m^3 of seawater

(0.3%) [29]. Mg has better vibration and noise dampening properties than Al and shows excellent castability and good ductility [30-32]. By alloying magnesium with zirconium [33], zinc [34], thorium [35], manganese [36], aluminum [37], or rare earth metals [38], the ratio of strength to weight increases, making these materials suitable choices where lightweight and the reduction of inertial forces are required. Due to this characteristic, denser cast iron, steel, copper-based alloys, and even Al-based alloys are replaced with Mg-based alloys [39, 40]. Mg is very appealing to the electronic and audio industry owing to its significant electromagnetic interference shielding [41, 42].

Important commercial Mg alloys are the AM series (Mg-Al-Mn), AZ series (Mg-Al-Zn), EZ series (Mg-RE-Zn), AE series (Mg-Al-RE), WE series (Mg-RE-Zr), and ZK series (Mg-Zn-Zr). Different casting processes are used for the production of Mg alloys. Gravity casting (permanent mold and sand casting) together with high-pressure die-casting are the most applicable fabrication techniques. Thixomolding, Thixocasting, and squeeze casting are other production methods [43-46]. According to the Mg consumption analysis by the International Magnesium Association (IMA), automotive components fabricated based on die casting Mg alloys are increasing significantly. In other words, the dominant production approach for Mg alloys with long-term potential growth is expected to be high-pressure die-casting [47].

Although the most common casting technique is die casting, the low density of Mg together with other advantages are important for aerospace applications using the sand cast method. Special Zr-containing magnesium casting alloys with yttrium, zinc, silver, and rare earth elements are utilized between 250 ˚C to 300 ˚C [48-50]. Also, wrought products including forgings, plates, sheets, and extrusions have been applied for numerous applications [39].

2.1 Application of Mg and its alloys

By applying magnesium alloys, it is possible to design lightweight engineering systems such as positive implications in reducing energy consumption. Moreover, these alloys have been used in the form of battery electrodes and viable biodegradable materials [51, 52]. The microelectronics industry benefits from Mg alloys in various components used in computer disk drives, CVD/DVD chassis, cellphone, and camera casings [53-55].

The primary reason to use Mg alloys in the automotive industry is environmental and energy concerns. As a result of a high ratio of strength/weight, Al and steel alloys can be replaced with Mg alloys in the automotive industry [56, 57]. A considerable number of research on auto manufacturing companies has been focused on the development of magnesium and its alloys [47, 58]. The first auto company that used Mg in its products was Volkswagen. The company used 22 kg of Mg in each Beetle model [59].In 1928, Porsche first used magnesium in the car engine [60]. The Mg average usage in 2005, 2010, and 2015 was 3 kg, 20 kg, and 50 kg per car, respectively [61].

In addition to fiber-reinforced polymers and composites, modern aircraft need structural metals. Al and its alloys have been already optimized as the traditional aerospace materials and there are limitations in the further enhancement of their strength together with the reduction of weight and the component dimension. Although Mg alloys have high strength-to-weight ratios and other promising properties, their high surface reactivity is a significant challenge [62-64]. In aerospace applications, the coating of Mg with nickel has also been suggested. It was shown that coating nickel on ZM21 alloy by direct electroless plating provided coatings with good soldering, optical, environmental, and mechanical characteristics [36].

Recently, Mg alloys have been of great interest as new degradable biomaterials. The properties that offer Mg alloys as promising materials for temporary implants include: (1) Mg is a necessary element for human metabolism. In terms of the abundance of the cations in the human body, cationic magnesium is in fourth place and the stored amount of this element in our body is 25 g. Half of this value is stored in the bone tissue. (2) The density of Mg and its alloys (1.74-2.0 g.cm^{-3}) is lower than the density of Ti alloys (4.4-4.5 g.cm^{-3}) and close to the density of bones (1.8-2.1 g.cm^{-3}). (3) The elastic moduli of Mg alloys (41-45 GPa) are close to that of the bone resulting in the alleviation of the stress shielding effect. Moreover, these alloys show greater fracture toughness compared to ceramic biomaterials. Mg is also a cofactor for different enzymes in the body and acts as a stabilizer for the structures of RNA and DNA [65-70]. (4) The standard electrode for magnesium has a potential of -2.37 V, and in a physiologic environment containing Cl, Mg

Fig. 1. Schematic of the applications of Mg and its alloys.

metal shows even poorer corrosion resistance. Hence, biodegradable Mg alloys can be developed by benefitting from their high-rate corrosion in physiologic environments [70-72]. Fig. 1 shows the schematic illustration of the Mg and its alloys applications in various fields.

2.2 Corrosion process in Mg and its alloys

Although magnesium alloys exhibit good properties suitable for different applications, the problem of their poor corrosion resistance is still a concern [73-75]. Because of the low standard electrode potential of Mg, this metal is the most reactive one. A galvanic corrosion system is formed between Mg and another metal and even micro-galvanic corrosion could occur between Mg and some secondary phases and/or impurities in aqueous environments [53, 76]. Therefore, the discussion of the possible current density of galvanic corrosion and micro galvanic corrosion over-potential is useful. The negative difference effect (NDE) of Mg and its alloys, which is different from most metals like copper and iron, lies at the center of the magnesium corrosion process. Overall, shedding light on the main causes of the Mg corrosion process could lead to progress in corrosion-related research. The galvanic current as well as its distribution can determine the galvanic corrosion rate as follows [74, 77]:

$$I_g=(\Phi_c-\Phi_a)/(R_a+R_c+R_s+R_m) \tag{1}$$

Where the galvanic current between the cathode and anode was denoted by , the potentials of the open circuit for the anode and cathode are represented by and , the anode and cathode resistances were denoted by and , respectively. is the solution resistance between the cathode and anode and is the metal resistance from the surface of anode to the surface of the cathode across a metallic path. In fact, Mg alloys are mostly used as structural materials in atmospheric environments; however, there are limited reports about the atmospheric corrosion of these alloys. Most studies are oriented towards an electrochemical approach in solutions [78, 79]. Oxygen reduction and water reduction are thought to contribute to the cathodic reaction on Mg in the atmospheric environment and under immersion condition [80]:

Anodic reaction:

$$Mg(s) \rightarrow Mg^{2+}(aq) + 2\bar{e} \tag{2}$$

Cathodic reaction:

$$2H_2O + 2\bar{e} \rightarrow H_2\,(g) + 2OH^{-}(aq) \tag{3}$$

$$O_2 + 2\,H_2O + 4\bar{e} \rightarrow 4OH^{-}(aq) \tag{4}$$

The general reaction has been shown as the following:

$$Mg^{2+}(aq) + 2\,H_2O\,(aq) \rightarrow Mg\,(OH)_2\,(s) + H_2 \tag{5}$$

$$2Mg^{2+}(aq) + 2\,H_2O\,(aq) + O_2 \rightarrow 2Mg\,(OH)_2\,(s) \tag{6}$$

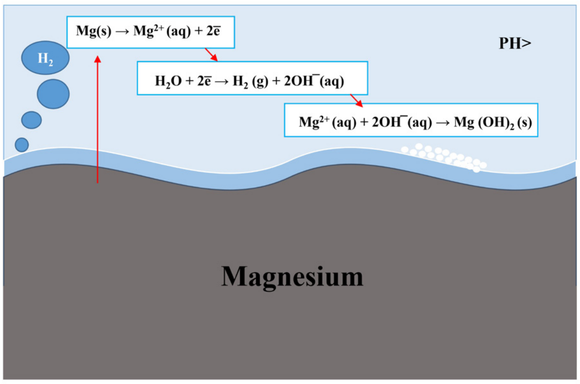

Fig. 2. Schematic of magnesium corrosion mechanism in the atmospheric environment.

At the ordinary atmosphere CO_2 level, a direct reaction occurs between brucite and $CO_{2,}$ and magnesite is formed. The reaction is:

$$Mg(OH)_2\,(s) + CO_2 \rightarrow MgCO_3\,(s) + H_2O \tag{7}$$

Fig. 2 shows a schematic illustration of the magnesium corrosion mechanism in the atmospheric environment. Compared to conventional metals like Mn, Al, and Zn alloys, Mg and its alloys have higher chemical activity due to their corrosion potential between -1.73 and -1.67 V-NHE. As a result, Mg alloys often act as an anode in contact with other metals and exhibit low corrosion resistance. Hence, galvanic corrosion is prevalent in Mg alloys that are in contact with other metals. Localized corrosion, fatigue corrosion, galvanic corrosion, stress corrosion cracking (SCC), and intergranular corrosion are different kinds of corrosion in Mg alloys [1, 81, 82]. These drawbacks constrain the application of Mg alloys in various engineering fields [83]. If unprotected Mg is in contact with an aqueous solution or moisture containing SO_x or Cl ions, it will be corroded severely due to the lack of sustainability and self-healing capability of the hydroxide-oxide-carbonate film formed on the surface of Mg [79, 83, 84]. Therefore, the prominent reasons for weak corrosion of these alloys are the formation of unstable, quasi-passive hydroxide film on their surface and the presence of impurities or secondary phases in their structure leading to internal galvanic corrosion [77, 79, 85].

2.3. Corrosion-resistant coatings for Mg and its alloys

Most light metals such as Ti and Al can form a passive oxide film on their surfaces; however, Mg cannot form such a protective film. Hydroxide/oxide/carbonate films are rapidly formed on the surface of Mg when exposed to the atmospheric environment [86-88]. These films have porous structures, are inhomogeneous and poorly bonded, and are unable to inhibit the corrosion of the underlying metal [89]. The formation of a coating on the metal surface that can form or add functional barrier layers leads to the isolation of the metal from the surrounding environment. This is an effective approach for the improvement of the corrosion resistance of Mg and its alloys. Besides, a good base is formed for applying subsequent organic coatings [90]. Some other coating treatment methods have been also developed such as vapor-phase processes, hybrid coatings, anodizing, conversion coatings, and electrochemical plating (electroplating) [1, 91-93].

There are two basic problems regarding single-layer coatings applied on the surfaces of Mg alloys: Firstly, conversion coatings are unable to provide Mg alloys with prolonged protection against corrosion [1, 13]. Secondly, the presence of an interface between the substrate and coatings cannot provide long-term protection. Hydroxide/oxide/carbonate precipitate forms a layer on the magnesium surface, weakening the adherence of applied coatings to the metal substrate. Thus, as the corrosive agent penetrates the interface between the substrate and the coating, the top coat is rapidly delaminated. The high volume of corrosion products under the top coat results in the detachment of the coating from the sub-

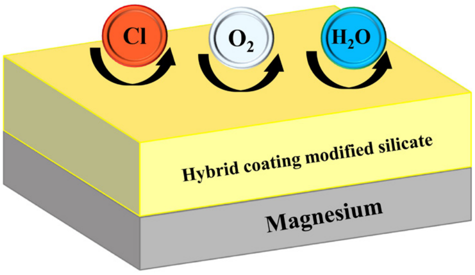

Fig. 3. Illustration of the protective effect of the silicate-modified hybrid coating against corrosion.

strate [94, 95]. Considering these issues, it is concluded that applying a single-layer coating on the surface of Mg alloys cannot provide enough corrosion protection [85].

3. New coating materials for corrosion protection

3.1 *Hybrid coatings*

Combining organic and inorganic materials in a single-phase hybrid network makes it possible to tune the desired coating characteristics for various applications [96]. The synthesis of materials containing hybrids of inorganic and organic compounds by various methods is a new field of research in materials science. The considerable attention paid to this field has led to the new hybrid material emergence with promising properties for novel applications [97-99]. The strength and nature of organic/inorganic interactions determine the properties of hybrid materials. The interaction strength increases when strong bonding, such as covalent bonding, replace weak van der Waals forces. Organic/inorganic hybrids may be categorized into two groups depending on the interaction type or the chemical bonding nature between the components: hybrids with poor interactions between the two constituents are classified into class I, and the ones with strong chemical interactions are classified into class II [100, 101]. Structural properties can be measured for distinguishing between different organic/inorganic hybrids. An ancient painting of Maya blue is an old example of a class I hybrid material made by ancient Mexican people. They prepared Maya blue by encapsulating the natural blue indigo within clay mineral channels. This approach made it resist the severe environments for more than twelve centuries; this could not be achieved by simply mixing the two components [102]. The sol-gel technique could be used as a proper synthetic process to prepare the thin-film hybrid coatings. It is possible to produce transparent hybrid coatings in which molecular-scale interactions exist between the constituents with the sol-gel process [103, 104].

A group of the hybrid materials that have the potential to be used in a variety of industrial materials is organically modified silicates (or mosils). They can be used for catalyst supports, porous materials for chromatography, corrosion protection coatings, anti-fogging coatings, anti-soiling, abrasion-resistant, colored glasses, hard coatings, and optical materials with high reflection due to the facile process depending on the molecular scale [105]. The mechanical behavior of these materials is also promising, owing to the combination of tough organic phases and rigid inorganic phases [106, 107]. In these systems, dispersed organic groups throughout the film enhance the coating hydrophobicity, render them water-repellent, and improve the corrosion resistance [108-110]. Fig.3 schematically shows the protective effect of the silicate-modified hybrid coating against corrosion.

To enhance the corrosion resistance of AZ31 Mg alloy, Li et al. [111] prepared a hybrid coating of poly(lactic-co-glycolic acid) (PLGA) and dicalcium phosphate dihydrate (DCPD). First, a DCPD coating was applied on the substrate using electrochemical deposition and then a PLGA coating was deposited to form the coating. This hybrid coating was effective to increase the magnesium alloy resistance against corrosion. Gao et al. [112] developed an effective biomimetic approach to coat AZ91 with a hydroxyapatite/graphene oxide (HA/GO) hybrid. According to the results, the hybrid coating decreased the corrosion current density of the metallic substrate by one order of magnitude in comparison with bare alloy.

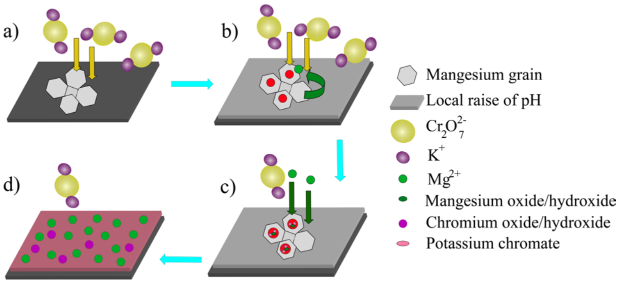

Fig. 4. Illustration the deposition mechanisms of the Cr conversion coating on the metal in the treatment bath (a) immersion of the pretreated alloy, (b) reaction of redox, (c) the coating deposition, and (d) complete coating deposition..

Peres et al. [113] added different amounts of silica nanoparticles to (3-glycidoxypropyl)trimethoxysilane (GPTMS) and tetraethylorthosilicate (TEOS) hybrid films to improve the corrosion properties of AZ31. The results suggested that the hybrid coating provided corrosion protection for Mg alloys and adding the SiO_2 nanoparticles further increased the corrosion resistance. Lamaka et al. [114] used the sol-gel approach to form an organic-inorganic hybrid coating on AZ31B magnesium alloy. The coating was applied through copolymerization of Ti or Zr alkoxides and epoxy-siloxane. The additive of tris (trimethylsilyl) phosphate was also utilized for the provision of additional corrosion protection. It was proposed that hydrolytically stable chemical bonds of Mg–O–P was formed in the coating doped with tris(trimethylsilyl)-phosphate, and increased the magnesium alloy resistance to corrosion.

Zhang et al. [115] employed a chemical conversion route for applying a crack-free hybrid coating of HA/phytic acid (PA) on the AZ31 surface. After the formation of the precursor coating, the coating was hydrothermally treated in the saturated CaO solution. It was shown that the hybrid coating had stable barrier properties and could effectively protect the Mg alloy. To enhance biological behavior and obtain tunable degradation, Kang et al. [116] deposited a hybrid coating of poly(ether imide) (PEI)-SiO_2 on Mg. An increase in the content of SiO_2 resulted in the enhancement of coating hydrophilicity and thereby the corrosion rate of the substrate increased. In these systems, dispersed organic groups throughout the film enhance the coating hydrophobicity, render them water-repellent, and improve the corrosion resistance. The magnesium alloys coated with such films have a good resistance to corrosion.

3.2. Rare earth conversion coatings

Due to the simple and low-cost procedure, conversion coating treatment has been considered for different applications. The conversion coatings for Mg are typically applied for the provision of enhanced paint-base properties and corrosion protection [117]. Because of the excellent corrosion resistance as well as a simple coating process, chromate conversion coatings have been favored greatly in recent years. Nevertheless, the application of these coating is limited due to the hexavalent chromium ion toxicity. Other conversion coatings on Mg alloys such as stannate [118-121], phosphate-permanganate [122, 123], phosphate [124-126], and rare earth metals [127-130] conversion coat-

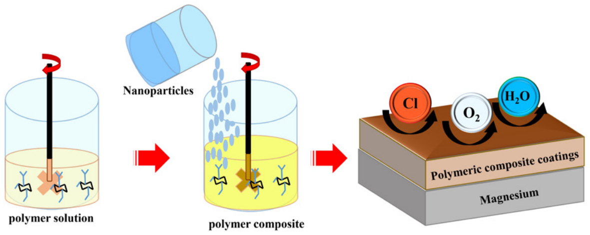

Fig. 5. Illustration of the polymer composites' production containing nanoparticles and their anti-corrosion properties.

ings have been investigated. The process of preparing rare-earth conversion coatings is accompanied by simple electrolytic constituents, mainly, chloride, sulfate, and neodymium, cerium, nitrate, and lanthanum rare earth metals. Therefore, its maintenance and recycling would be easy. Another important advantage of the rare-earth conversion coatings is that they are considered as environment-friendly treatment [13].

Rudd et al. [127] coated Mg and WE43 alloy with praseodymium, lanthanum, and cerium conversion coatings. The coating decreased the magnesium dissolution in a buffer solution with pH=8.5, however, after 60 min of immersion, a deterioration in the coating was observed. The reason was stated to be the formation of corrosion products of Mg hydroxide together with mixed coatings composed of rare earth/Mg oxide and hydroxides. In another research, Brunelli et al. [131], produced a conversion coating of cerium for the corrosion protection of Mg, AZ91, as well as AM50. It was reported that by reducing the cathodic and anodic currents and ennobling the corrosion potential, the corrosion resistance of bare Mg and its alloys could be increased. Furthermore, a further increase in the corrosion resistance was achieved by acid pre-treatment. The result of corrosion investigations in the chloride environment showed that after five days, the coated samples were unaffected while localized corrosion was observed in the untreated samples.

Montemor et al. [132] studied the effect of conversion film of rare-earth metals (cerium and lanthanum) on the corrosion behavior of the AZ31 Mg alloy. The AZ31 substrate showed reduced corrosion activity in the presence of chloride ions. Moreover, the treatment time affected the efficiency of corrosion protection. On the other hand, Li et al. [133] reported weak adhesion of cerium conversion coatings to the AZ31 substrate and limited corrosion resistance enhancement of the coating. Lin et al. [13] also found the inherent weakness in adhesion of cerium conversion coatings to the surface of AZ31 leading to coating partial detachment when dried at room temperature. Cross-sectional observations exhibited three layers with porous, compact, and fibrous structures formed sequentially on the surface of the substrate. Among the interfaces, the interface between the fibrous and compact layers was identified to be the weakest bonding.

According to Lin et al. [134], conversion coatings of lanthanum and cerium could improve the corrosion resistance of AZ63 alloy. An increment in immersion time resulted in the improvement of the inhibition effect provided by the dual rare-earth film. Laleh et al. [135] used the micro-arc oxidation (MAO) method to apply oxide coatings on AZ91D magnesium alloy and then the samples were soaked in a Ce bath for sealing the pores of the MAO coatings. The results revealed that the pore sealing for 10 min exhibited a remarkable improvement of the substrate corrosion resistance. Fig.4 shows a schematic illustration of the deposition mechanisms of the Cr conversion coating on the metal in the treatment bath.

Many works were reported on rare-earth coatings for Mg and its alloys. According to the results of corrosion studies, the rare-earth coatings can provide good corrosion resistance.

3.3. Polymeric nanocomposite coatings

Polymers and polymer composites are widely used in different engineering applications [136-139]. A range of physical and tribological features are offered by polymeric nanocomposites [140]. The addition of nanoparticles in polymers leads to the improvement of properties such as anti-corrosion performance, thermal conductivity, mechanical strength, and electronic and optical properties [141, 142]. Fig. 5 shows a schematic illustration of the production of polymer composites containing nanoparticles and their anti-corrosion properties.

Metallic components used in engineering, energy, defense, and biomedical applications are encountered with the problem of corrosion [143]. In this regard, wear and abrasion resistance, anti-corrosion resistance, and barrier properties of polymeric nanocomposite are used. Restriction of electron flow from the metallic substrate to oxidizing agents occurs within doped conducting polymers preventing corrosion of the substrate. However, neat polymers as metal coatings are unable to provide good wear properties and corrosion protection [144]. Nanoparticles such as titania, silica, nanoclay, carbon black, carbon nanotube, graphene oxide (GO), graphene, nanodiamond, and fullerene nanoparticles have been added to various polymeric matrices to improve their corrosion resistance. To enhance interfacial adhesion and load transferring of matrix/nanofiller, functionalization has been considered [145]. Functional nanoparticles can increase the strength of nanocomposites and restrict the diffusion of corrosive species within the polymer matrix to prevent corrosion [146]. The underlying material can be also protected against friction and wear [147, 148].

Nazeer et al. [149] added GO and Ti nanoparticles to poly (butyl methacrylate) (poly(BMA)) to prepare a nanocomposite coating for AZ31 magnesium alloy. The results demonstrated that the nanocomposite coating was able to act as an excellent passivation layer to prevention diffusion and corrosion so that the corrosion current decreased significantly and the charge transfer resistance increased noticeably. Soleymani et al. [150] incorporated different amounts of baghdadite mineral to polycaprolactone (PCL)/chitosan (Ch) for improving the resistance to corrosion, biocompatibility, as well as bioactivity of the anodized AZ91 alloy. According to the results, the nanocomposite coating containing 3 wt % baghdadite showed hydrophobic behavior that led to decreasing the corrosion current density for the magnesium alloy and the enhancement of its corrosion resistance.

A corrosion control layer of 3,4-ethylenedioxythiphene (PEDOT)/GO nanocomposite was coated on magnesium in research by Catt et al. [151]. The results indicated that the corroding samples exhibited an increment in the polarization resistance, reduction of corrosion current, and more positive corrosion potential. It was reported that three factors contributed to corrosion protection of PEDOT/GO including redox coupling with magnesium corrosion resulting in the creation of a protective magnesium phosphate layer, inducing negative charges in the film, and the formation of an initial passive layer that prevents solution uptake. In the research of Rahimi et al. [152], cellulose nanoparticles (CNs) were added to polylactic acid (PLA) coating to protect the AZ31 alloy. The outcome of adding CNs to the coating was the enhancement of the PLA corrosion resistance. The most promising result of corrosion resistance improvement was attributed to the coating containing 5 wt. % of CNs.

Zhang et al. [153] coated AZ91D Mg alloy with polyaniline (PANI)/organophilic montmorillonite (OMMT) polymer nanocomposite. They reported that after 6000 h of immersion in the corrosive environment, the Mg alloy coated by PANI/OMMT maintained its high corrosion resistance. A dense Ch/hydroxyapatite (HA) composite coating was applied on AZ31 Mg alloy by Hahn et al. [154]. A high adhesion strength range of 24.6-27.7 MPa was reported for the coating and it exhibited higher corrosion resistance compared to the bare Mg substrate. To improve bone-implant integration and control the degradation of Mg-based substrates, Johnson et al. [155] developed PLGA/nanostructured hydroxyapatite (nHA) coatings using the spin coating method. Nano-scale features of the coatings were retained with the dispersion of nHA in the PLGA matrix. According to the corrosion behavior studies in the revised simulated body fluid, Mg substrates coated by the composite showed an enhancement of the corrosion potential and reduction of the corrosion current.

Nanocomposite coatings of the addition of inorganic nanofillers into the polymer matrix are a new method for corrosion protection that exhibit excellent mechanical performance and corrosion resistance in comparison to conventional composite coatings

3.4. DLC coatings

DLC involves a broad range of amorphous carbon coatings having different hydrogen levels and sp^2 and sp^3 bonded carbon ratios. A variety of properties can be obtained by changing the ratio of the three components. There are similarities in the properties of DLC and diamond, however, DLC has an amorphous matrix in which sp^3 nodules exist in a matrix of bonded sp^2 [156, 157]. DLC coatings have a dense microstructure, chemical inertness, and high mechanical hardness [158]. Due to the structure of DLC coatings, high residual stress is built within these coatings [159]. Consequently, surface adhesion would be weak resulting in the delamination of the coatings at early stages. By the deposition of interlayers, the surface adhesion can be improved. The interlayers are thin coating layers that are applied for the promotion of DLC adhesion [160, 161]. The interlayer is required to have a strong bonding with both the DLC top coat and the substrate. Silicon nitride (Si_3N_4), chromium carbide (CrC), and titanium (Ti) are common interlayer materials. The interlayer can also contribute to the enhancement of the corrosion resistance because DLC could be both conductive and porous. Hence, the layer can act as a physical barrier between the corrosive environment and the substrate and decrease the delamination risk [162].

Recently, DLC coatings have been considered by many researchers for the promotion of the anti-corrosion and mechanical properties of magnesium alloys [161, 163]. Nevertheless, the direct deposition of hard materials like DLC coatings on Mg-based soft substrates is a challenge. Considerable differences in physical properties including thermal expansion coefficient, plasticity, and elastic modulus between the coating and substrate lie at the center of the problem [164-166]. Wu et al. [164] applied a three-layer coating composed of aluminum, aluminum nitride, and diamond-like carbon (from bottom to top) on the surface of AZ31. The results indicated the enhancement of the corrosion resistance of the coated alloy so that the presence of 3.5 wt% NaCl in solution reduces the current density from 2.25×10^{-5} A.cm^{-2} to 1.28×10^{-6} A.cm^{-2}. Anti-corrosion behavior of the Si-incorporated DLC (Si-DLC) films on AZ31 Mg alloy was studied by Choi et al. [20]. The prepared Si-DLC films yielded noticeable corrosion protection for the Mg alloy. The improved anti-corrosion performance of the Si-DLC films is the result of very low internal stress within the coatings. Wu et al. [167] deposited columnar layers of Cr and CrN with preferred textures of (110) and (111), respectively as interlayers for applying a DLC coating on AZ31. Although the adhesion between the substrate and coating was improved significantly by applying the interlayers, the enhancement of the DLC/AZ31 corrosion resistance was not observed. This was reported to be the result of the galvanic cell formation between interlayer and substrate in the through-thickness defects in a solution of 3.5 wt. % NaCl.

Masami et al. [168] also incorporated Si into DLC coatings to provide corrosion protection for AZ91 magnesium alloy. Results indicated that the Si-DLC coating had higher corrosion resistance in comparison with DLC coating. Applying Ti interlayer promoted adhesion between the AZ91 substrate and DLC coating, however, it negatively affected corrosion protection. Corrosion resistance, as well as adhesion strength of Mg alloy, was improved by treating the surface of the substrate with ozone. In a research study by Uematsu et al. [169], it was shown that the corrosion fatigue strength of AZ80A in distilled water was not improved by applying single-layer and multilayer DLC films. The films contained some defects facilitating the access of the corrosive water to the substrate. The thicker multilayer DLC coating showed no degradation of fatigue strength in the corrosive environment revealing its effectiveness for corrosion protection. According to the reports of Yamauchi et al. [170], it was found that the diamond-like carbon coating on the Mg-14 mass% Li alloy could not withstand the corrosive alkaline and acidic solutions.

4. Conclusions and future insights

Mg and its alloys suffer from low corrosion resistance. Mg alloys, unlike other light metals such as Ti and Al, are not capable of forming naturally passivating oxide films. Porous, poor-bonded oxide/hydroxide/carbonate layers are rapidly developed on the Mg surface under atmospheric exposure that are unable to protect the substrate. Different coatings are being used for Mg alloys to isolate the substrate from corrosive environments. Organic-inorganic hybrid coatings, polymeric nanocomposite, rare earth conversion coatings, and DLC coatings have been demonstrated to have great impacts on the promotion of corrosion resistance of Mg alloys. These new coatings have shown promising results in terms of corrosion protection of Mg alloys in different corrosive environments, however, they still need to be improved. Future studies might concentrate on the promotion of these coatings as well as the development of novel coating materials for the corrosion protection of magnesium and magnesium-based alloys. The development of inexpensive coating processes and accessible coating materials are favored for the industry and this issue should be considered in future research.

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Conflict of interest

The authors declare that there is no conflict of interest.

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