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Smart self-healing coatings on biomedical magnesium alloys: A review

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ABSTRACT

With promising mechanical property and biodegradability, magnesium (Mg) alloys are considered as the potential candidates in biomedical application. Rapid degradation of Mg alloys compromises the mechanical performance and interfacial bioactivity, hindering clinical adoption. Deposition of a surface coating is an effective technique to improve the corrosion protection and bio-efficacy of Mg biomedical implants. Because of the natural degradability and corrosion, the surface is inevitably damaged in the complex physiological environment. Therefore, it is essential to form a self-healing coating that can repair damage to restore the stable mechanical structure and functions of biomedical Mg alloys. This paper reviews the recent advances in coating technology and the related properties including biodegradation behavior, self-repairing activity, biocompatibility, and other biological effects, and the healing mechanism is discussed. Self-healing coatings suitable for Mg alloys include conversion coatings, encapsulation coatings, and multilayered coatings, and their properties *in vitro* and *in vivo* are reviewed by focusing on drug-controlled and prolonged release, sterilization, cytocompatibility, osteogenesis, hemocompatibility, and angiogenesis. This review aims at providing guidance for the future research and development of practical healing coatings for biomedical Mg implants.

1. Introduction

Magnesium (Mg) and its alloys are temporary implant materials with exceptional mechanical properties and biodegradability [1–3]. Owing to the superior biocompatibility, Mg alloys have been used in biomedical applications [4]. The release of Mg^{2+} ions is crucial to activating enzyme activity and sustaining cell growth and neuromuscular activity [5,6]. However, excessively rapid degradation of Mg compromises the mechanical properties and even produces implant failure, consequently hampering wide clinical applications to orthopedic implants and cardiovascular stents [7–9]. In addition, the corrosion process causes hydrogen emission and local buildup besides local alkalinization from Mg release near the implant/tissue interface, possibly impacting the health of patients [10,11].

Since it is essential to improve the corrosion resistance of Mg alloys [12], microstructural treatment and surface modification have been proposed [13,14]. The general corrosion rate can be reduced by alloying, but Mg alloys are susceptible to localized corrosion [15]. Compared to alloying, surface modification can restrain the degradation activity of Mg without altering the favorable bulk attributes [16]. Various coating techniques including micro-arc oxidation (MAO) [17], layered doubled hydroxide (LDH) formation [18,19], plasma electrolytic oxidation (PEO) [20,21], phosphate conversion [22,23], and polymer incorporation [24] have been proposed and developed for Mg alloys to tailor the corrosion resistance and other properties. Although these coatings improve the anti-corrosion capability of Mg alloys, they are temporary and can be damaged mechanically leading to localized corrosion and even premature failure [25,26].

To address these issues, inspired by the healing process of natural living organisms, smart coatings with self-healing characteristics have been proposed and developed for Mg alloys [27–29]. Generally, most damaged coatings need cumbersome repair or substitution, otherwise premature failure can occur [30,31]. Therefore, self-healing coatings which can repair defects autonomously with or without external stimuli are desirable to retard degradation and prolong the service time of biomedical implants [32,33]. Introduction of the self-healing ability to

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polymer-based materials for defect repair was first proposed by C. Dry [34] in 1996 and it has since spurred much research [35]. Although the concept of self-repairing coatings was originally applied to carbon steels, scientists have also studied their application to biomedical metals such as Mg, Zn, and Fe.

The aim of this review is to summarize the latest developments in smart self-healing coatings on biomedical Mg alloys by focusing on the coating technology, mechanism, and properties. The different types of self-healing coatings are described in Fig. 1.

2. Self-healing coatings on Mg alloys and clinical requirements

Biomedical Mg alloys have bioactive surfaces which can promote osseointegration and reendothelialization and resist infections. In addition, the self-healing mechanisms supplementing the basic clinical features have been proposed [36,37]. Construction of surface coatings on Mg-based materials can enhance the corrosion protection and interfacial biocompatibility in contact with body fluids and can also be used as delivery carriers for therapeutic agents [38,39]. Studies have focused on controlling corrosion, non-toxic byproducts, as well as bioactivity [40,41]. In short, self-repairing coatings must possess excellent biocompatibility, avoid immune reactions, and produce effective anti-inflammatory responses within 3–7 days after surgical implantation [42]. Self-recovering coatings must also possess sufficient adhesion strength to bear the dynamic stress in patients because the lack thereof can cause mechanical injury or implant failure [43,44]. Moreover, the bacteria resistance is important to biomedical coatings in order to mitigate post-surgical infections [45,46]. Hence, coatings with intelligent self-healing properties are desirable.

3. Technology-driven self-healing coatings on biomedical Mg alloys

In general, physical barrier coatings can satisfy the anticorrosion requirements for biodegradable Mg alloys but they must degrade together with the Mg alloy substrate after tissue recovery [47,48]. Until now, most coatings need to be manually repaired or replaced thus requiring high maintenance costs. Based on protective surfaces in nature, self-healing coatings with biomimetic self-curing functions have been developed [49,50]. When these coatings undergo degradation, the surface damage can be repaired with minimal or no external intervention [51–53]. The self-repairing activity can enhance the protection capacity and extend the service life of biomedical Mg-based implants [[54]- [55,56]]. Conceptually, smart healable coatings can partially or completely repair damages and preserve the original functions automatically or independently [57,58]. Table 1 summarizes recent developments in this area.

3.1. Conversion coatings

Conversion coatings consist of compounds produced *in situ* by chemical or electrochemical reactions on the metal surface and it is relatively simple to fabricate a surface with enhanced adhesion strength [82–84]. Generally, such a treatment produces oxide, hydroxide, and/or phosphate on the surface of Mg alloys [85–87]. Compared to a deposited coating, this *in situ* growth environment provides improved adhesion strength [88–90]. The common methods include the use of chemical bonds and intermetallic compounds [91,92]. According to the chemical species, a chemically bonded coating reacts with the Mg substrate chemically [93–95] to provide enhanced adhesion strength, albeit a certain selectivity in the coating materials and chemical reactants. Chemical conversion coatings are produced by chemical bonding. In

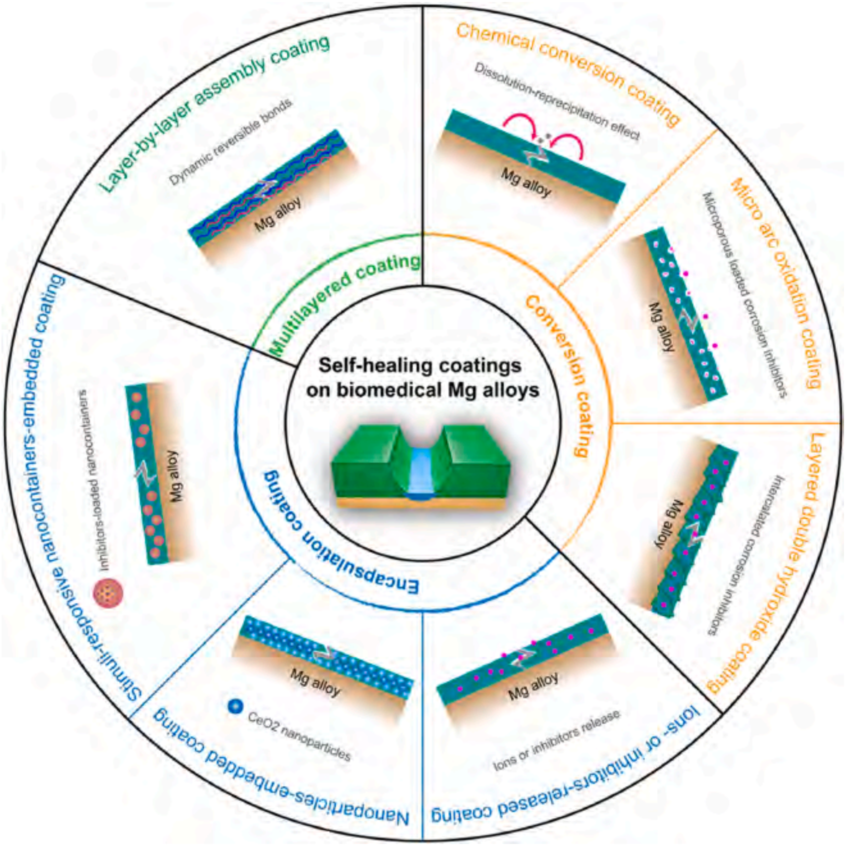

Fig. 1. Schematic representation of smart self-healing coatings on biomedical Mg alloys.

Table 1
Recent developments of smart self-healing coatings on biomedical Mg alloys.

Coatings	Substrate	Methods	Self-healing mechanism	Application	Refs
HAp and OCP	AZ31	Dip-coating	Formation of magnesium and calcium phosphates	Bone-fixation devices and stents	[59]
DCPD	Pure Mg	Dip-coating	DCPD uses as inhibitor and Hanks provides foreign-aid film-forming ions	Implants	[60]
Sr-HA	AZ31	Electrodeposition	Dissolution-precipitation mechanism	Osteogenesis	[61]
HA + PEO	Mg plate and screw	PEO and hydrothermal treatment	HA reacts with Ca	Osteogenesis	[62]
PA/Ca^{2+}	Mg–Sr	LbL assembly	Ca^{2+} can re-permeate and strengthen chelation between PA and Mg^{2+} ions	Cytocompatibility	[63]
DCPD/PA	Pure Mg	Dip-coating	Formation of apatite and Mg-PA chelating compounds	Implants	[64]
AgAC	Mg–3Zn-0.5Sr	MAO	The addition of CH3COOAg led to the formation of Ag_2O and Ag_2CO_3 phases	Bacteria killing and osteogenesis	[65]
MAO/CSCe	Mg–1Ca	MAO and spin-coating	Formation of cerium oxide/hydroxide	Cytocompatibility	[66]
LDH-SiO_3	AZ31B	Hydrothermal treatment and anion exchange	Formation of Mg silicate precipitates	Cytocompatibility	[67]
LDH/Bg	AZ31	Hydrothermal growth and sol-gel spin-coating	Formation of stable MgF_2 and $Ca_{10}(PO_4)_6F_2$ products	Bone-repairing biomaterials	[68]
MAO-LDH	AZ31	Low-temperature in-situ grown	Diffusion behavior of LDH coating	Cytocompatibility	[69]
Sol-gel/PCTyr Schiff-base	ZE21B	Dip-coating	Chelation and deposition of PCTyr Schiff base and Mg in defects	Cardiovascular	[70]
Silk-KP	Mg–1Ca	Spin-coating	Formation of $Mg_3(PO4)_2$ salt	Osteogenesis	[71]
Poly (TA-Zn)	ZE21B	One-step dip-coating	Existence of hydrogen bonds, Zn–COOH coordinative bonds, and dynamic covalent disulfide bonds	Bacteria killing and reendothelialization	[72]
(CIP/PAH/SiO_2/PAH)$_{20}$	AZ31	SSLbL assembly	SiO_2 migration	Bacteria killing	[73]
SiO_2/CeO_2	AZ31	SSLbL assembly	Formation of Ce-rich passive coating	Orthopedic implants	[74]
Silk-PA	Mg–1Ca	Spin-coating	PA complexes with Mg^{2+} and Ca^{2+} ions	Osteogenesis	[75]
DCPD/cFMSNs@PLA/MgO	WE43	Combination of MAO, spin- and dip-coatings	F^- reacts with Mg^{2+} to form precipitation film	Osseointegration	[76]
PDA/8-HQ@PEDOT/PLA	AZ31	Dip-coating	Formation of $Mg(HQ)_2$ passive layer	Cytocompatibility	[77]
Silk-HNT/PA	Mg–1Ca	Spin-coating	Deposition of corrosion inhibitors	Osteogenesis	[78]
(AgNPs/PEI)$_5$	AZ31	Dip-coating	Diffusion-driven rebonding of the cross-linked polysiloxane structure	Bacteria killing	[79]
Ce + HT	Pure Mg	Electrodeposition and hydrothermal treatment	Formation of $Ce(OH)_4$ and insoluble CeO_2	Osteogenesis	[80]
Ta/TaN	AZ91	Nitrogen plasma immersion ion implantation and high-vacuum magnetron	Reactions between tantalum intermediate layer and electrolytes and penetrating ions through defects as well as formation of oxide compounds	Bacteria killing and cytocompatibility	[36]
PEO/PDA	AZ91	Combination of PEO and electrodeposition	Zipper-like and pore block mechanism by binding between $Mg(OH)_2$ and catechol groups	Cytocompatibility	[81]
Gd_2O_3	AZ31	Electrophoretic deposition	Formation of apatite formation	Bacteria killing and cytocompatibility	[37]

comparison, intermetallic binding involves an intermetallic compound membrane at the coating-substrate interface with no additional materials required [96]. Under the effects of force and heat, an intermetallic coating with the same composition and crystal structure as the substrate is produced and the coating is combined with the compound in the substrate to improve the adhesion strength. Previous research has emphasized the potential application of conversion coatings by utilizing a bonding membrane sandwiched between the coatings and Mg-based substrate [97,98]. Multiple conversion coatings with self-healing capabilities have been developed, for example, chemical conversion coatings, micro-arc oxidation (MAO) coatings, and layered double hydroxide (LDH) coatings.

3.1.1. *Chemical conversion (CC) coatings*

A chemical conversion coating can be viewed as a membrane transforming the active surface into a passive state [99–101]. Briefly, a chemical conversion coating comprises a barrier layer with excellent bonding strength with the Mg substrate and is formed by the reaction between the metal atoms and anions in the medium. Chen et al. [102] have provided a general review of chemical conversion coatings in technical applications and shown that the coating performance relies on the proper pretreatment to functionalize the coatings. Chemical conversion coatings include chromate conversion coatings [103,104], vanadium conversion coatings [105–107], stannate conversion coatings [108–110], phosphate conversion coatings [111–113], phytic acid conversion coatings [114,115], and tannin conversion coatings [116,117]. Nevertheless, only chromate, phytic acid, and phosphate conversion coatings can be used as self-healing coatings on Mg alloys [118,119]. The self-healing activity of chemical conversion coatings mainly depends on the dissolution-reprecipitation effect at defective locations related to the nature of the cladding materials and degradation behavior [120–122]. Except for magnesium oxide/hydroxide, chemical conversion coatings include a mixture of other oxides and hydroxides formed in solutions. Self-healing chemical conversion coatings have attracted attention because they are cost-effective and suitable for a wide range of biomedical applications. Generally, self-healing chemical conversion coatings are divided into two categories, inorganic and organic conversion coatings.

3.1.1.1. Inorganic conversion coatings. Early research on self-repairing coatings largely concentrated on chromate conversion coatings which are now used commercially [123]. Belin et al. [124] have reported that the self-repairing mechanism that involves hexavalent chromate reacting with Mg to form Cr^{3+} and then reacting with hydroxide ion in the corrosive medium. After the chemical reaction, the original Cr^{6+} is converted into the $Cr(OH)_3$ sediment which can be deposited onto damaged sites to recover the original structure and functions. Recently, Zhang et al. [125] have proposed a Cr(III) conversion coating composed of amorphous/nanocrystalline Cr and Cr_2O_3 with the excellent self-repairing capability. However, CrO_3 is formed during corrosion. Chromate conversion coatings possess advantages such as easy preparation, low cost, and healing [126]. However, due to carcinogenicity and high toxicity, clinical application of self-healing chromate conversion coatings is limited [127] and it is thus important to develop non-toxic Cr-free conversion coatings on Mg alloys with self-healing characteristics for biomedical applications.

Phosphate is used to prepare self-healing coatings on Mg alloys. Phosphate conversion coatings consist of trace metallic complexes that enhance the biocompatibility and cellular response [128]. The calcium-phosphorus conversion coating is especially meaningful as a result of the formation of hydroxyapatite (HAp). Among the various Ca–P phases, HAp is closest to the mineral phase of natural bone and is highly recognized as an osteoconductive mineral [129]. Besides, HAp provides corrosion protection in the physiological environment. The HAp coating can be fabricated on Mg-based alloys by the sol-gel method [130] or electrochemical deposition [131], but the adhesion strength is relatively low. The bonding strength of chemical conversion coatings can be improved by *in situ* coating preparation [132]. Osseointegration of the HAp membrane mainly relies on the coating surface and compared with the whisker and flake HAp, nanoplates and nanospheres-containing coatings exhibit improved cell adhesion [133]. HAp coatings possess a controllable morphology and uniform thickness, but fabricating such self-healing coatings without a fragile surface and spatial defects remains a challenge.

Hiromoto et al. [59] have fabricated octacalcium phosphate (OCP) and HAp on AZ31 and pure Mg (Fig. 2). Release of Mg^{2+} (Fig. 2A) is inhibited on the scratched samples and as shown in Fig. 2B, no significant differences are observed among different samples on the 1st day, indicating that corrosion is not accelerated. Both the HAp and OCP coatings are self-repairing due to the formation of quasi-corrosion protective layers containing magnesium phosphate and calcium phosphate in the scratches (Fig. 2C and Table 2). The self-healing mechanism is illustrated in Fig. 2D. When the phosphate membranes are scratched, an anodic reaction (Eq. (1)) occurs on the exposed Mg, but a cathodic reaction (Eq. (2)) occurs in the corrosive solution. Subsequently, Mg^{2+} ions react with the produced hydroxyl and dissolved phosphate to generate $Mg(OH)_2$ and $Mg_3(PO_4)_2$ sediments to seal the scratches. However, the self-healing property relies solely on the immersion experiments with pH variations, but they are not rigorous enough to fully validate the effectiveness of the phosphate coatings. Therefore, it is necessary to demonstrate the self-healing properties of phosphate coatings by other measurements.

$$Mg \rightarrow Mg^{2+} + 2e^- \tag{1}$$

$$2H_2O + 2e^- \rightarrow 2OH^- + H_2\uparrow \tag{2}$$

Dong et al. [60] have fabricated a dicalcium phosphate dihydrate (DCPD) coating with the self-healing ability on the Mg alloy by a dip-coating method. The DCPD coating has excellent corrosion resistance and self-healing properties. The scratches are sealed by corrosion products containing Ca and P. The scratched DCPD coatings release more hydrogen than the unscratched sample, but there is no significant difference between the scratched and unscratched groups in the overall corrosion process, indicating that the DCPD coatings have excellent self-healing ability. DCPD provides layer-forming ions to suppress local corrosion, which is considered an autonomous self-repairing mechanism. The corrosion inhibitors and ions in Hank's solutions can enhance healing and foreign corrosion inhibitors can improve self-curing. The discovery provides new opportunities for further research and application of efficient self-healing coatings on biodegradable Mg implants. Huang et al. [61] have prepared a strontium-doped hydroxyapatite (Sr-HA) coating with high solubility on the Mg substrate. The Sr-HA

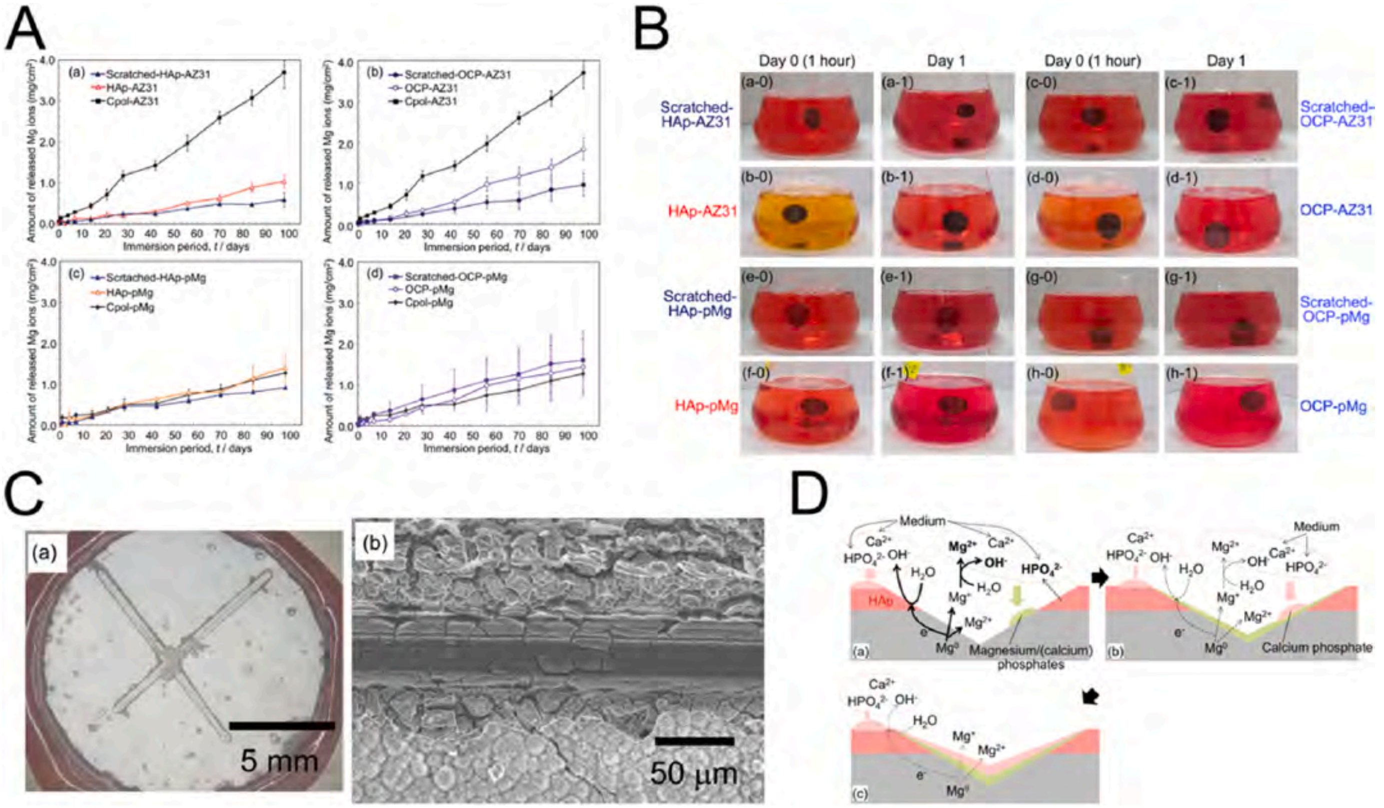

Fig. 2. (A) Mg^{2+} release plots; (B) Colors of phenol red in the cell culture of different samples; (C) Optical images and morphologies of the damages; (D) Schematic illustration of the self-healing mechanism [59]. Copyright 2015, Elsevier.

Table 2
Compositions (at%) of the corrosion products at the scratches on the scratched HAp-AZ31 sample [59]. Copyright 2015, Elsevier.

Specimen		Mg	Al	Zn	Ca	P	O	Na	Cl	Ca/P ratio
Scrached-HAp-AZ31	Corrosion products in scratches	21.2	5.3	0.5	0.1	3.4	69.5	–	0.3	0.04
	Remaining coating	0.7	0.4	0.9	10.5	11.9	75.6	–	–	0.9

coating acts as a physical barrier to delay the formation of pits and provides ion replenishment to seal cracks (Fig. 3A). By regulating Mg degradation and Sr release, improved osteocompatibility is observed *in vitro* (Fig. 3B) and *in vivo*. The implant exhibits only 2.7–5.6% reduction in the Mg volume 12 weeks after implantation as observed from ovariectomized rats (Fig. 3C). Compared to the HA-coated sample, the Sr-HA coating promotes early osteogenesis and bone tissues with higher trabecular thickness and hardness are formed around the SR-HA coating.

3.1.1.2. Organic conversion coatings. Compared with inorganic surfaces, organic conversion coatings have fewer spatial defects and cracks on the surface and organic conversion coatings offer superior corrosion protection for biomedical Mg alloys [134] in addition to excellent self-repairing activity [135]. Conceicao et al. [136] have fabricated a polyvinylidene fluoride (PVDF) coating on the hydrofluoric acid-pretreated Mg substrate. The self-recovering ability stems from the transformation from corrosion-induced conversion of $Mg(OH)_2$ to MgF_2. Zong et al. [137] have constructed a flower-like bis (8-hydroxyquinoline) magnesium ($Mg(HQ)_2$) coating on the Mg substrate. It shows the dissolution and recrystallization behavior during degradation and Fig. 4A reveals the self-repairing activity to suppress local corrosion. Kim et al. [62] have fabricated a dense hydrophilic coating by plasma electrolytic oxidation (PEO) using hyaluronic acid and carboxymethylcellulose to improve bonding. Hyaluronic acid bonds quickly to Ca^{2+} and apatite in body fluids resulting in healing of the outer layer in the damaged region, and magnesium oxide grows out of the inner cavity to seal the defects (Fig. 4B). After the screws are implanted for 1 month, both the heads and necks begin to degrade (Fig. 4C). The PEO + HA coating shows the smallest corrosion rate throughout, although the screw heads exposed to bones degrade. Fig. 4D shows the self-healing features of the coated groups and there is obvious new bone regeneration around the thread one month after implantation. In the PEO + HA group, although osteoblasts are formed uniformly and substantially around the threads 2 months after implantation, initial bone formation is irregular similar to the PEO group, and there is tight adhesion with the cartilaginous bone. Therefore, the polymer-ceramic composite layer is corrosion-resistant while offering the cellular activity and new bone formation capability for the absorbable magnesium screws.

Phytic acid (PA) is an organic phosphate polymer which can form insoluble compounds with metal ions such as Mg^{2+}, Ca^{2+}, and Zn^{2+} due to the biocompatibility, negative charges, and high chelation ability [138–140]. It is a desirable candidate to construct self-healing coatings on Mg alloys. Zhang et al. [141] have designed a PA conversion coating by dipping the AZ31B Mg alloy in the PA solution. The cracks in the conversion film heal gradually and the corrosion resistance of Mg is improved by a heat treatment. The amorphous magnesium phytate coating is converted to crystalline $Mg_2P_2O_7$. The transformation of this macromolecular complex into a small molecular organic architecture is accompanied by a volume increase, leading to self-repairing of surface cracks. Liu and co-authors [142] have prepared a PA/Ca^{2+} hybrid coating on Mg and the incorporation of Ca^{2+} re-permeates and strengthens the chelation between the PA molecules and Mg^{2+} to improve self-curing of damages. The *in vitro* tests illustrate that this conversion coating has good biocompatibility and bioactivity. Dong et al. [64] have constructed a hybrid DCPD/PA coating with anti-corrosion and self-healing properties. The pH increase stimulates release of Ca^{2+} and PO_4^{3-} to promote the formation of apatite and also mitigates corrosion in body fluids. The PA-containing complexes are regulated by enrichment of Mg^{2+} and converted into Mg-PA chelating compounds with improved self-repairing capability. In summary, chemical conversion coatings have excellent mechanical stability but suffer from low efficiency. Therefore, better self-healing coatings are required.

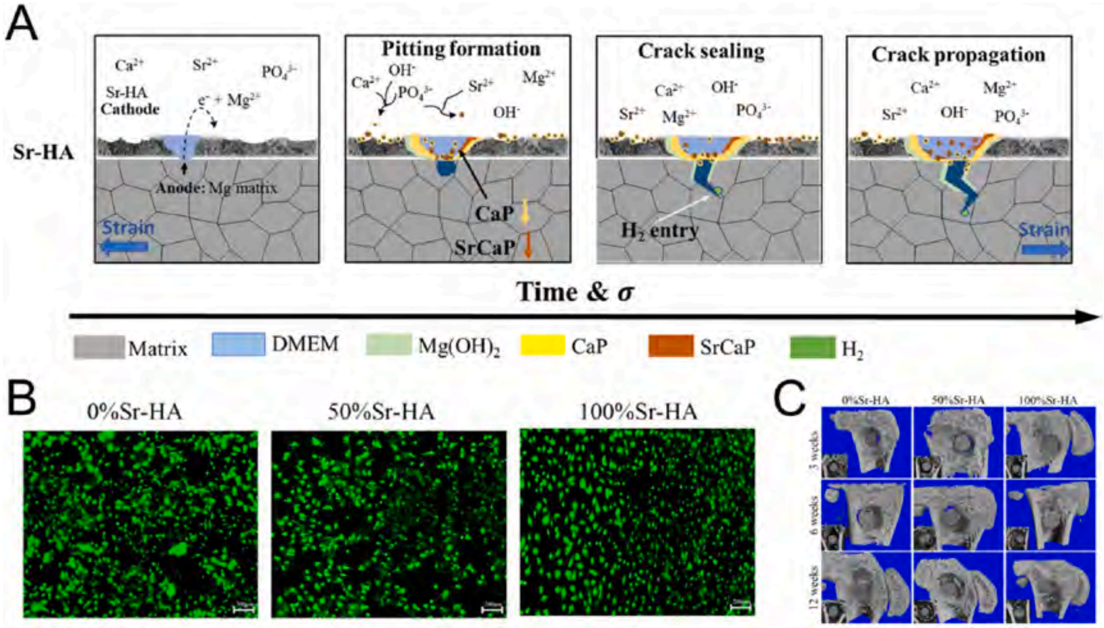

Fig. 3. (A) Schematic presentation of the corrosion and self-healing mechanisms; (B) Fluorescent images of pre-osteoblasts cultured on the surfaces; (C) Micro-CT and histomorphometry analysis [61]. Copyright 2023, Elsevier.

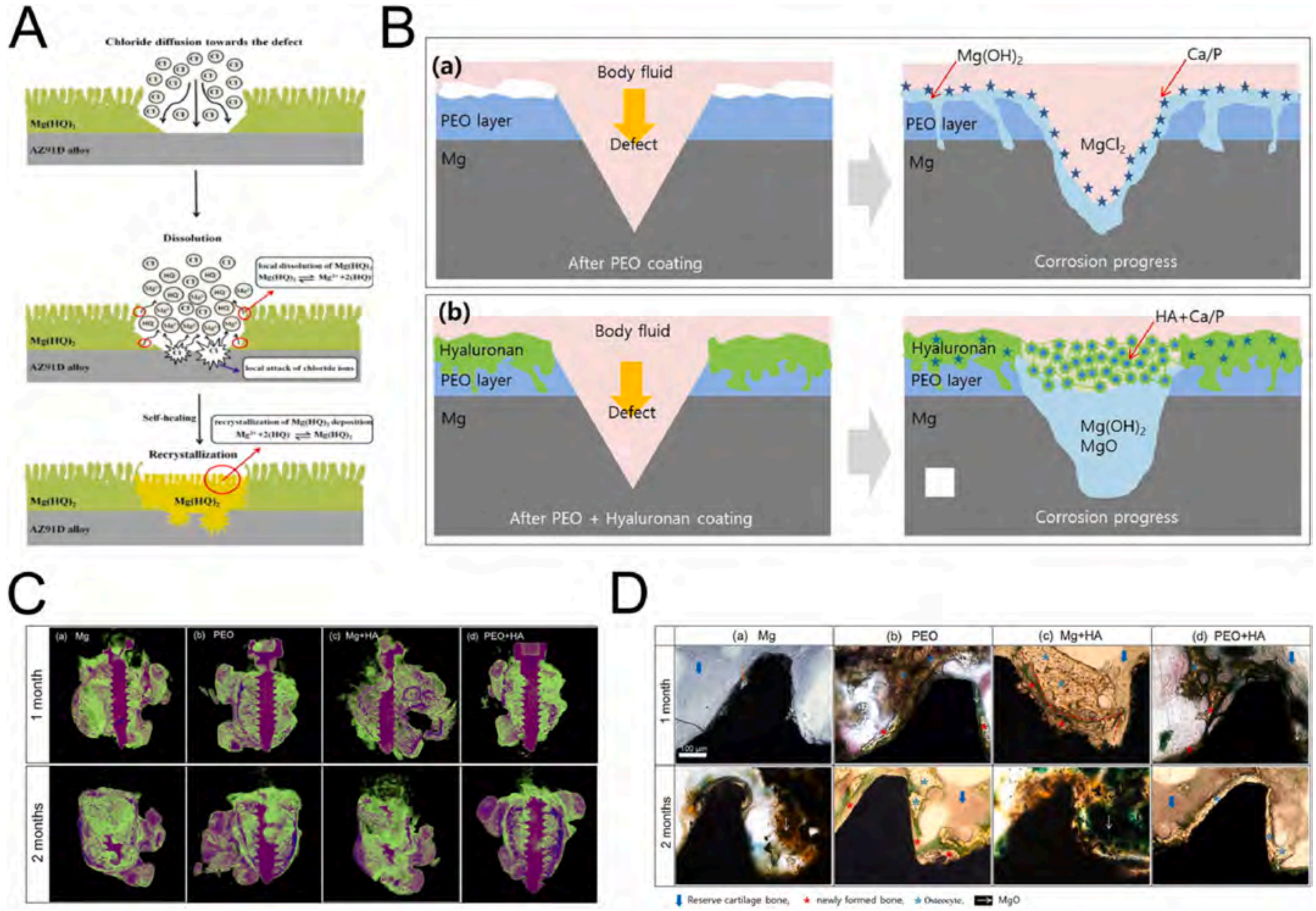

Fig. 4. (A) Schematic illustration of the self-healing mechanism [137]; (B) Schematic presentation of the healing progress; (C) Micro-CT images of the rat femur; (D) Histological images around the screw [62]. Copyright 2014, 2019, Elsevier.

3.1.2. *Micro arc oxidation (MAO) coatings*

Micro arc oxidation (MAO) is developed based on anodic oxidation and the resulting coatings possess higher bonding strength and wear resistance compared to those produced by traditional anodic oxidation techniques such as electrochemical deposition or electrophoretic deposition [143]. The micro-arc oxidation process relies on the matching of the electrolyte with plasma parameters such as the electrolyte chemistry, reaction time, applied voltage, and pulse frequency [144]. Under the action of instantaneous high temperature and pressure generated by the arc discharge, the modified ceramic coating is grown on the surface of the Mg alloy. Generally, the surface of the micro-arc oxidized Mg has micropores [145], but a porous coating is not recommended for long-term implantation because corrosive ions in the physiological environment can penetrate the pores to corrode the underlying Mg substrate. Therefore, a pure MAO coating is not sufficient to shield the Mg substrate.

Researchers have attempted to reduce the risk of quick degradation by incorporating corrosion inhibitors [146–148]. Generally, corrosion inhibitors can be combined with ions such as Mg^{2+} and OH^{-} around the fracture to form an internally soluble precipitate, which is then sedimented at the damaged site and can be adsorbed at the artificial defects to generate a protective thin film to realizing effective self-repairing. Chen et al. [65] have reported an Ag-containing self-healing antibacterial micro-arc oxidation coating on biodegradable Mg-based alloys. Addition of CH_3COOAg leads to the formation of Ag_2O and Ag_2CO_3 phases which seal the micropores and improve the corrosion protection ability. As shown in Fig. 5A, the MC3T3-E1 osteoblasts seeded on the AgAC-2 and AgAC-3 samples exhibit low cytotoxicity and good binding strength due to the controlled release of Mg^{2+}. The samples incorporated with 2 and 3 g L^{-1} of CH_3COOAg exhibit excellent bacteria-killing capability against *E. Coli* on account of Ag^{+} release during corrosion as shown in Fig. 5B.

Preparing an organic film on the inhibitors-incorporated MAO conversion coating can further enhance the corrosion protection for Mg-based materials. Liu et al. [149] have prepared a dual self-healing coating by embedding the corrosion inhibitor M − 16 into the micropores of the MAO layer on the α-Mg substrate followed by spraying of a disulfide-bonded modified polyurethane film. This coating delivers excellent performance in releasing inhibitors into the scratched area and closing the damaged area by thermally inducing the disulfide exchange reaction in the outer polymer coating. This approach provides insights into the design of self-healing coatings on Mg/Mg alloys. Jia et al. [66] have developed an inhibitor encapsulation, anticorrosion, self-healing, and cytocompatibility coating on the Mg–1Ca Mg substrate by combining MAO with spin-based assembly as shown in Fig. 5C. Trivalent cerium (CeIII) is the inhibitor and the polysaccharide chitosan is the carrier. The ceramic coating bonds well with the substrate and the pores act as "interlocking" sites for the polymer. The Ce–NH_2 coordination between the inhibitor and chitosan is believed to be the cause of particle fixation and prolonged delivery. Compared to the bare Mg alloy, the coating has good self-healing properties and protects the substrate from biological corrosion (Fig. 5D). As shown in Fig. 5E, the self-repairing mechanism includes two steps: (i) The protonated coating is disintegrated due to corrosion causing the local pH to increase; (ii) The chitosan chain expands to the active site and produces cerium oxide/-hydroxide to repair the damage. The coating is also biocompatible

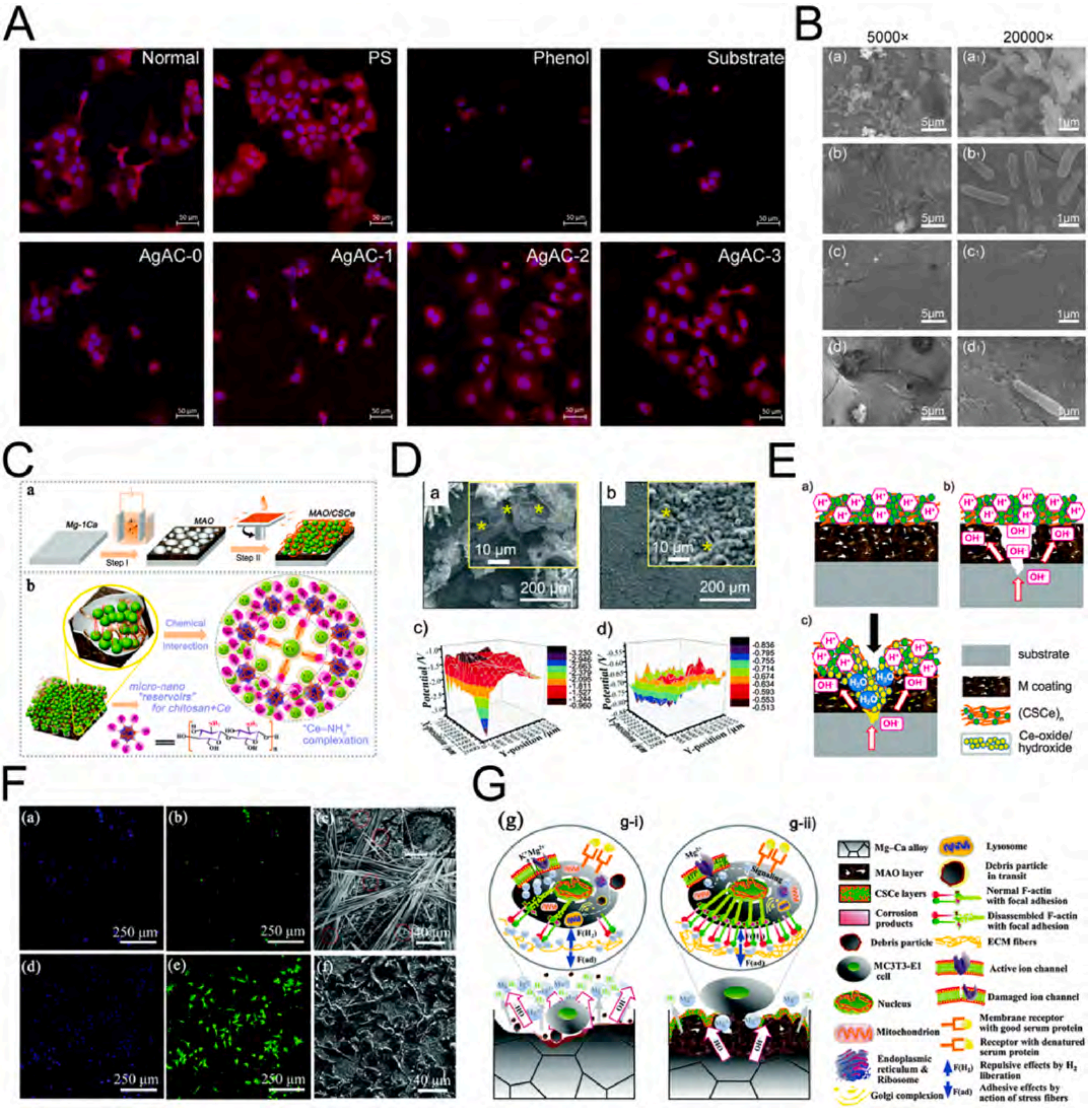

Fig. 5. (A) Fluorescent images of MC3T3-E1 attached on the specimens; (B) Morphology of *E. coli* on the samples [65]; (C) Schematic presentation of the coating preparation; (D) Morphology and SKP maps of the samples after immersion; (E) Schematic of the mechanism of self-healing; (F) Staining and morphology of MC3T3-E1 cells on the samples; (G) Schematic displaying the possible factors affecting cell adhesion on the surfaces [66]. Copyright 2021, Taylor & Francis; 2016, Royal Society of Chemistry.

showing good adhesion and growth of osteoblasts (Fig. 5F). The possible materials/cellular interface and cellular changes upon materials exposure are shown in Fig. 5G. Briefly, for the bare Mg, the high local pH can denature membrane proteins, thereby disrupting protein-based transport channels and disabling active ion transport. The high local concentration of Mg^{2+} alters the osmotic pressure of the cell membrane. In addition, large amounts of corrosion products are produced, especially those with sharp morphologies providing mechanical/physical means to damage cells. Continuous H_2 evolution together with denatured serum proteins mitigates cell anchoring contributing to the detachment of adherent cells and causing the motor of adhesion-actin fibers to break down and inactivate the adherent spots. At the same time, excessive endocytosis of the detached debris particles from the bulk metal and their lysis in lysosomes may dramatically alter the cellular microenvironment giving rise to an inflammatory cascade response or even necrosis. To sum up, the size and distribution of micropores affect the corrosion resistance, adhesion, and biocompatibility of MAO coatings and therefore, additional chemical and mechanical pretreatment and post-treatment techniques must be considered for MAO coatings.

3.1.3. Layered double hydroxide (LDH) coatings

Layered double hydroxide (LDH), a general term for hydrotalcite and hydrotalcite-like compounds, is an inorganic compound derived from a group of anionic clays with a ductile brucite arrangement. Its structure contains a positively charged layer of mixed metal hydroxide [150–152]. There are several *in situ* and *ex situ* deposition techniques to produce LDHs coatings, including urea hydrolysis, hydrothermal treatment, steam coating, co-precipitation, and spin-coating [153–155]. LDH has an interlayer layered nanostructure that gives them unique anion exchange properties to capture, store, and release anionic corrosion inhibitors in the interlayer architecture. Therefore, LDH can be used to prepare self-healing coatings on Mg-based substrates. Chen et al. [156] have constructed aspartic acid intercalated Mg–Al LDHs nanostructured coatings on the AZ31 Mg alloy by a hydrothermal treatment with self-healing and anticorrosion properties. When the scratched coating is immersed in the corrosive medium, corrosive chloride ions penetrate the LDHs interlayer and release intercalated aspartic ions, which chemically adsorb to the scratches to form protective films. After immersing for 20 days, LDH dissolves and aspartic combines to form an insoluble precipitate but the process is time-consuming.

To shorten the self-healing time and provide better corrosion resistance, researchers have developed various schemes. Ding et al. [157] have developed tungstate-intercalated LDHs coatings with superhydrophobic, self-healing anti-corrosion properties on the AZ31B Mg substrate. The composite coating has remarkable self-healing properties and the coating recovers from the peak current density to the noise level in only 8 h because the superhydrophobic coating prevents the intercalation inhibitor (tungstate) from being wasted by ion exchange in the initial stage of immersion, thus improving the utilization of the inhibitor. Chen et al. [158] have loaded the corrosion inhibitor 8-hydroxyquinoline (8-HQ) onto graphene oxide (GO) and then incorporated it into Mg–Al LDHs. The graphene oxide blocks the corrosive medium and changes the direction of the LDHs nanosheets to horizontal growth to avoid vertical growth of nanosheets for denser nanosheet films. Li et al. [159] have introduced a thiophene derivative corrosion inhibitor into the Mg–Al LDH self-healing coating using dip-coating (Fig. 6). With the highly positive shift in the corrosion potential, the stable physicochemical adsorption of *N*-alkyl-N, *N*-dime-thyl-*N*-(3-thienylmethylene) ammonium bromide (NTA) provides the self-healing capacity at damaged Mg sites.

The self-healing LDH-SiO_3 coating is very promising in expanding the clinical use of magnesium-based implants by providing a viable way to control degradation and enhance cytocompatibility. Li et al. [67] have developed a silicate-loaded MgAl LDH self-healing coating on the biomedical AZ31B using a hydrothermal technique and anion exchange for improved corrosion protection and cytocompatibility. The self-healing mechanism is mainly due to the release of SiO_2^{3-} from the LDH conversion coating and leaching of Mg^{2+} during natural degradation leading to the formation of the Mg silicate precipitate that seals the corroded regions. The LDH-SiO_3 coating improves the attachment and proliferation of MC3T3-E1 pre-osteoblasts due to inhibition of Mg^{2+} release and alkalization by the LDH-SiO_3 coating. Ouyang et al. [68] have designed a hierarchical layered double hydroxide (LDH)/bioactive-glass (Bg) coating on the AZ31 Mg substrate by the combination of the *in situ* hydrothermal growth and sol-gel spin-coating. The self-repairing effect and cooperative mineralization are produced when the LDH containers are pre-sealed with fluoride and paired with the bioactive glass (Bg). The self-healing behavior of the LDH/Bg hybrid coating is achieved by local redeposition of corrosion-produced Mg^{2+} and dissolved Ca^{2+} with the replaced F^- ions generating stable MgF_2 and $Ca_{10}(PO_4)_6F_2$ products in the pits. This smart coating takes advantage of the physical barrier and self-healing activity to suppress biodegradation and promote bio-remineralization on Mg. This study discloses a general approach to design smart anti-corrosion coatings and targeted drug delivery systems. Li et al. [69] have developed a self-healing Mg–Al LDH coating on the MAO-coated AZ31 Mg by a low-temperature *in situ* method. The MAO coating is completely covered by the nanosheet-modified LDH coating with a thickness of 3.8 μm. Compared to the bare Mg, the corrosion current density of the hybrid MAO-LDH coating decreases by 4 folds. The passivation zone in the anodic polarization branch indicates strong self-healing in addition to excellent corrosion resistance. The indirect extraction test of MC3T3-E1 pre-osteoblasts suggests that the composite-coated osteoblasts have the acceptable cell compatibility.

3.2. Encapsulation coatings

Encapsulation coatings are embedded with functional repairing agents and external stimuli (such as light, microwave, sound wave, electric field, and magnetic field) or internal stimuli (e.g., pH, temperature, redox potential, and enzyme) can trigger the transition from a passive self-healing coating to an active one [160]. In contrast to chemical conversion coatings, the fabrication of encapsulation coatings is based on intelligent and rational designs [161]. To achieve this goal, advanced and complex processing is involved to produce excellent corrosion inhibition and controllability for biomedical materials.

3.2.1. Ion- or inhibitor-releasing coatings

For effective encapsulation, the ions or corrosion inhibitors are directly doped into the host coating and then deposited on the Mg substrate. When local degradation occurs, the ions or inhibitors interact with Mg^{2+} or OH^- to produce insoluble precipitates to repair the physical damages. The key to smart coatings is the choice of the effective ions or corrosion inhibitors and optimization of the loading efficiency. Many ions or corrosion inhibitors have been proposed [162–164]. Polymers, LDHs, and plasma electrolytic oxidation (PEO) coatings are the effective carriers for ions or inhibitors. Song et al. [165] have prepared Ce^{3+}/La^{3+} co-doped gelatin-chitosan hybrid coatings on AZ91D and Li et al. [166] have prepared PA-modified silane coatings on the Mg substrate. Qian and co-authors [167] have fabricated a superhydrophobic self-healing coating with the dual actions using benzotriazole (BTA) and shape memory polymer. As shown in Fig. 7A, the

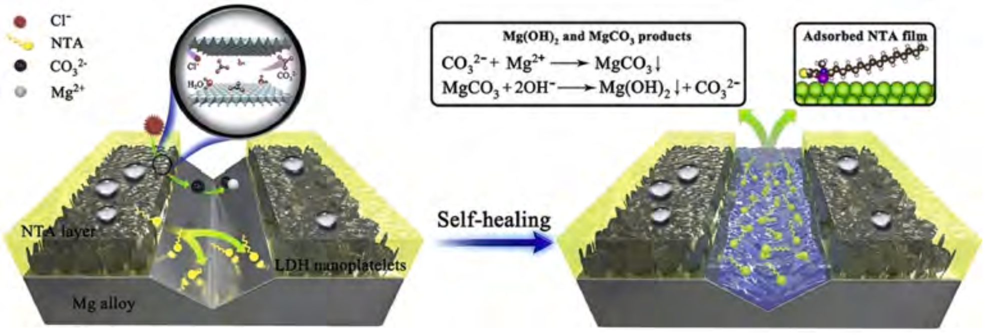

Fig. 6. Schematic presentation of self-healing mechanism by BTA adsorption on the damaged MA-Al LDH coating [159]. Copyright 2019, Elsevier.

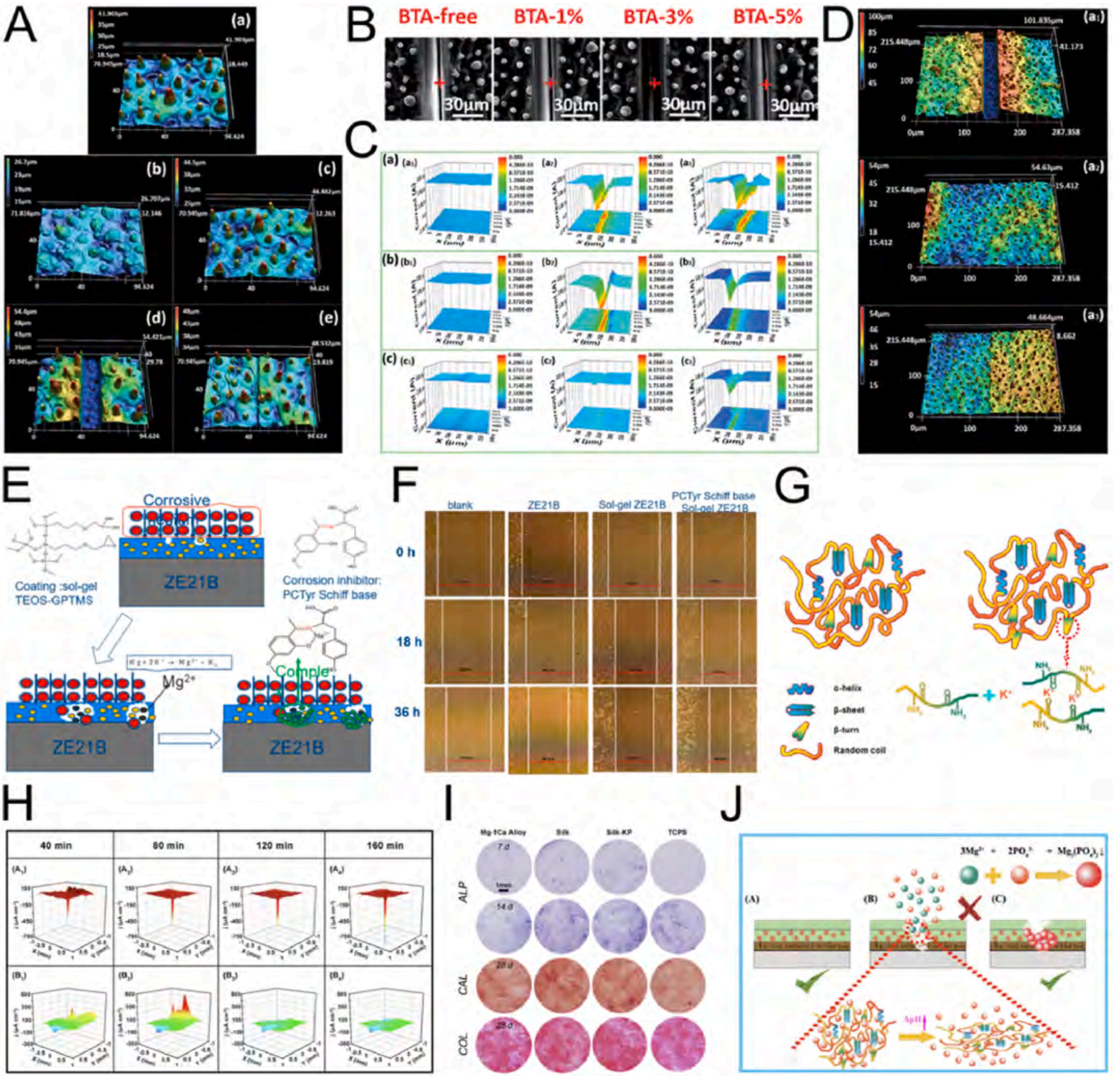

Fig. 7. (A) CLSM images of superhydrophobic coatings; (B) Morphology; C) SECM mappings of different coatings after immersion; (D) CLSM images of the scratched coatings [167]; (E) Schematic presentation of self-healing mechanism; (F) HUVECs migration assays [70]; (G) Schematic illustration of the secondary structural changes of silk fibroin composited with K^+; (H) SVET 3D maps of the electric current densities; (I) Optical images of the ALP activity, collagen secretion, and calcium deposition; (J) Schematic presentation of self-healing mechanism [71]. Copyright 2017, Royal Society of Chemistry; 2021, 2019, Elsevier.

healed surface exhibits complete recovery of the surface structure and roughness. As the BTA contents in the coatings increase from 0 to 5%, the physical scratches heal gradually in the corrosive medium as shown in Fig. 7B and C. Even during outdoor exposure, the coatings self-heal in sunlight without an external heating source (Fig. 7D). Li et al. [70] have produced a paeonol condensation tyrosine (PCTyr)-loaded sol-gel coating on the ZE21B Mg substrate by the dip-coating method and the good self-repair stems from chelation and deposition of the PCTyr Schiff base and Mg in the damaged area as shown in Fig. 7E. Besides, good biocompatibility and angiogenesis are observed *in vitro* (Fig. 7F).

Compared with other inhibitor ions such as MnO^{4-}, and VO^{3-}, PO_4^{3-} ions can chelate with Ca^{2+} and Mg^{2+} from the corrosive medium and the magnesium and calcium phosphate precipitates heal defects. Xiong et al. [71] have constructed a self-repairing coating containing silk and K_3PO_4 inhibitors on the Mg–1Ca substrate. The secondary structural changes of silk fibroin composited with K^+ ions are illustrated in Fig. 7G. After immersion for 160 h, the cathodic and anodic current densities at the defect sites are close to the zero planes indicative of self-healing (Fig. 7H). Multiple cell responses *in vitro* observed from the MC3T3-E1 cells such as spreading, adhesion, proliferation, and differentiation confirm the superior cytocompatibility and osteogenesis activities as shown in Fig. 7I. The self-healing mechanism of the silk-KP coating is illustrated in Fig. 7J. Zhang et al. [72] have developed a poly (thioctic acid) self-healing coating on the ZE21B Mg substrate with improved corrosion protection, bacteria killing, hemocompatibility, and reendothelialization. On the heels of these promising results, the cardiovascular stent with the poly (TA-Zn)-REDV hybrid coating ha large clinical potential in procedures related to arterial stenosis.

3.2.2. Nanoparticles-embedded coatings

Nanoparticles incorporated into coatings can enhance the mechanical and anticorrosion properties of Mg alloys. Coatings with nanoparticles possess better fluidity than ions/inhibitors-incorporated surfaces leading to superior self-healing properties. Because the coating is mostly made of polymers, the nanoparticles-incorporated one is also known as the inorganic-organic hybrid coating. Calado et al. [168] have designed a CeO_2 nanoparticles-incorporated siloxane hybrid coating on the AZ31 Mg substrate. Cerium improves and stabilizes the corrosion protection of Mg and the CeO_2 nanoparticles show no harm in terms of adhesion and coating integrity, as no cracks or defects are detected from the modified coating. The improved corrosion protection of the cerium dioxide-modified coatings may be due to the stability of the magnesium corrosion products which prevent the electrolyte from entering the substrate and thus repair the corrosion defects. Yabuki et al. [169] have prepared a TiO_2 particle-casein self-healing coating on the Mg substrate. In the coating, casein stabilizes TiO_2, while TiO_2 particles served as the enhancement phase to improve the self-healing properties and corrosion resistance. The self-recovering activity of the Ce-contained coatings is attributed to leaching of casein and TiO_2 particles when the pH of the local corrosion sites increases as shown in Fig. 8A. Cerium can mitigate the attack by corrosive chloride by forming protective Ce-oxide/hydroxide. Ji et al. [73] have developed a multilayered film composed of ciprofloxacin (CIP) and SiO_2 nanoparticles on the AZ31 Mg substrate by the spin-spray layer-by-layer (SSLbL) assembly method. The multilayers show excellent corrosion protection, self-repairing ability, bacteria-killing capacity, and prolonged drug release as shown in Fig. 8B. Zhao et al. [74] have fabricated a self-recovering coating with SiO_2 and CeO_2 nanoparticles by SSLbL assembly on the AZ31 Mg

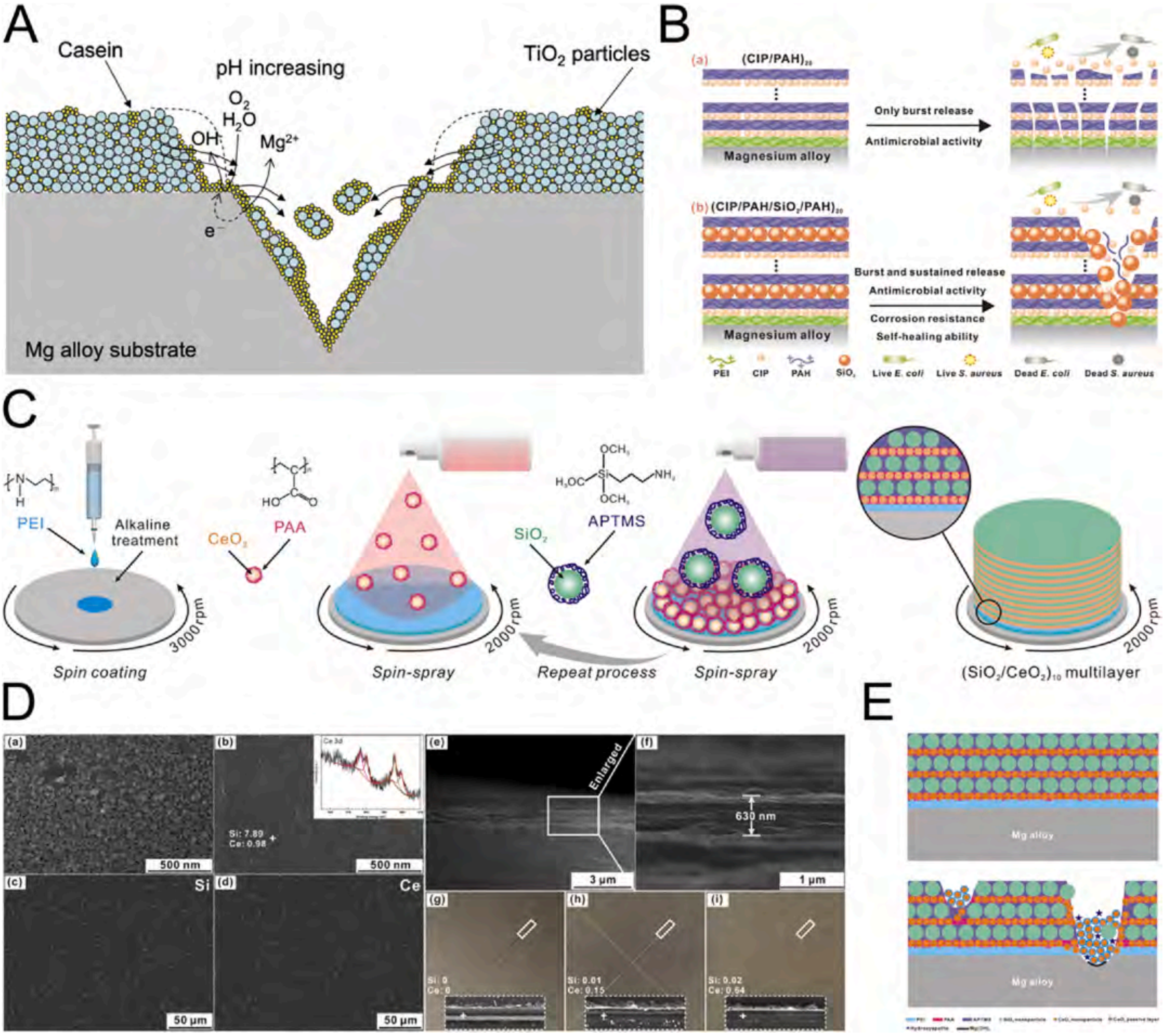

Fig. 8. (A) Schematic illustration of the self-healing mechanism of the TiO_2 particle-casein coating [169]; (B) Schematic presentation of the corrosion behavior, antibacterial activity, and release characteristics [73]; (C) Schematic illustration of the preparation process of multilayers on Mg alloys; (D) Morphology, XPS data, and cross-sectional images of the samples; (E) Schematic of the corrosion mechanism and self-healing activity [74]. Copyright 2010, 2019, 2018, Elsevier.

substrate as shown in Fig. 8C. The nanoparticles improve the corrosion resistance of the Mg alloy. As shown in Fig. 8D, smoother and denser multilayered coatings with a thickness of 630 nm are produced. In addition, the nanoparticles-embedded coatings have excellent self-healing properties. In the multilayered sructure, SiO_2 forms a physical barrier against the corrosive medium, while the CeO_2 nanoparticles are corrosion inhibitors to repair physical damages as shown in Fig. 8E.

3.2.3. Stimuli-responsive nanocontainers-embedded coatings

Nanocontainers are nanocarriers loaded with ions or inhibitors. Structurally, stimuli-responsive nanocontainers-incorporated coatings consist of four components: the guest coating (such as polymers), nanocontainer (such as mesoporous silica, zeolite, nanotubes, and metal-organic frameworks), repair agents (such as ions or corrosion inhibitors), and stimuli-responsive materials (such as polymers) [170, 171]. Typically, smart nanocontainers encapsulated with healing agents are evenly embedded in the host coating [172,173]. When local cracks occur, the healing agents are leached under certain stimulation to form a protective layer around the scratches [174–176]. These coatings combine the benefits of functional agents and nanoparticle-incorporated coatings and can avoid the initial boosted leaching of the corrosion inhibitors.

Xie et al. [177] have fabricated fluoride ion-loaded MCM-41 type mesoporous silica nanocontainers which are incorporated into a nickel coating on the AZ31 substrate. The released fluoride ions react with Mg^{2+} to produce MgF_2 which precipitates at the defect sites when corrosion products are generated. Although it is of great significance to repair scratches, unfortunately the modified coatings do not have responsive drug-controlled release functions. Ding et al. [178] have studied the stimulus-responsive activity of smart nanocontainers and rapid self-repair of nanocontainers-embedded coatings on Mg alloys. Supramolecular assemblies are used to modify the outer layer of magnetic nanocapsules (Fe_3O_4@$mSiO_2$) linked with disulfide bonds with the inner hollow encapsulating the 8-HQ corrosion inhibitors. The smart nanocontainers are triggered by the dual OH^- and Mg^{2+} stimuli and the nanocontainers-embedded coatings exhibit fast self-healing.

Nanocontainers-embedded coatings have attracted attention in biomedical applications. Xiong et al. [75] have designed a pH stimuli-responsive self-healing coating on the Mg–1Ca alloy in the form of a sandwich with a fluoride precoating (bottom), silk-phytic acid (PA) coating (middle), and silk fibroin coating (top). In the coating, PA is loaded into the intermediate coating as an inhibitor by utilizing the strong chelating ability to dissolve Mg^{2+} and Ca^{2+}. The silk-PA exhibits self-healing and pH responsiveness in conjunction with improved corrosion resistance. The coating also possesses biocompatibility and

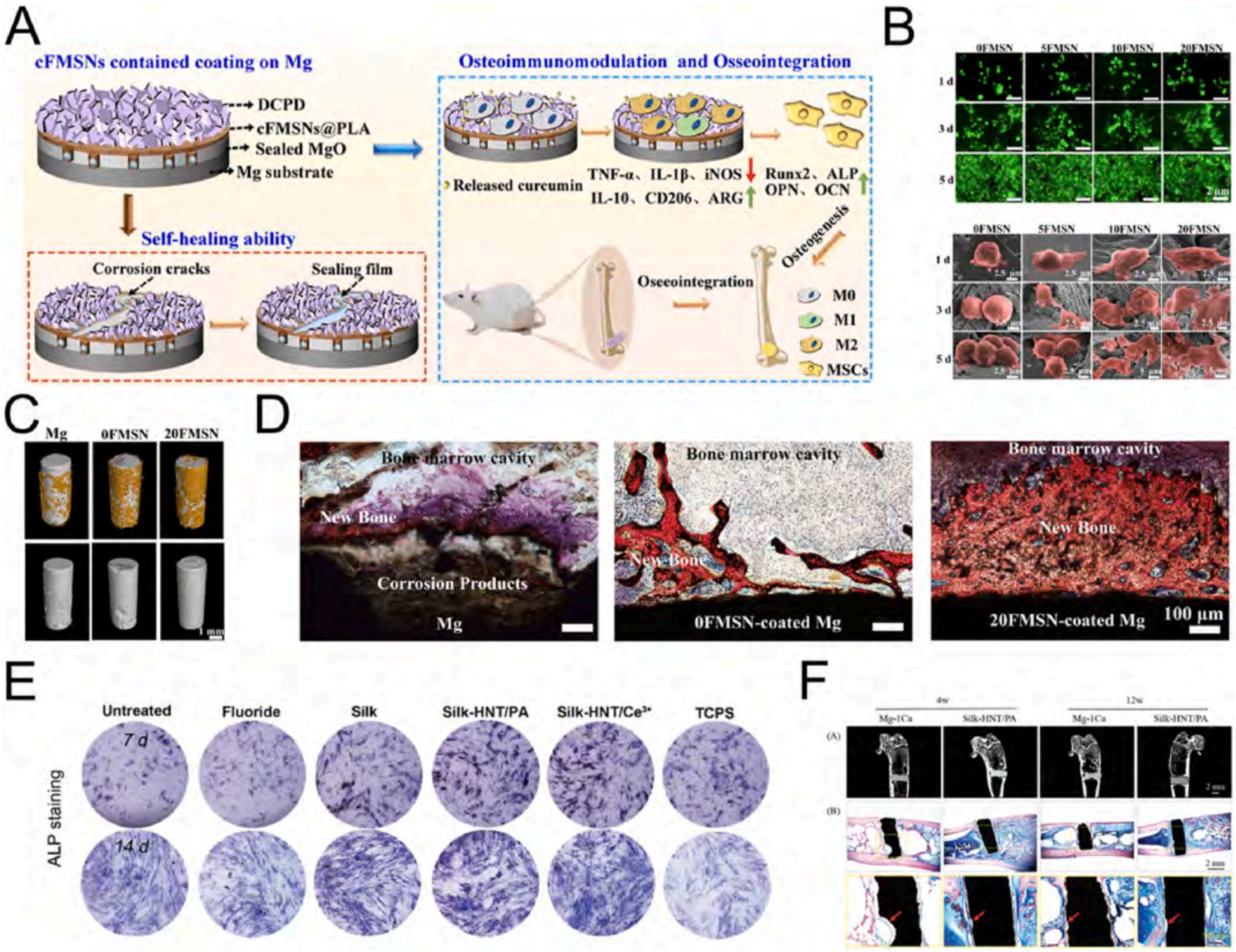

Fig. 9. (A) Schematic illustration of the coating preparation, osteo-immunomodulation, and osseoi-ntegration; (B) Live/dead staining fluorescent images and FE-SEM images of RAW264.7 cultured on the specimens; (C) Micro-CT images of the samples; (D) Histological stained images of the specimens [76]; (E) ALP staining of the specimens; (F) *In vivo* assays of Mg–1Ca and Silk-HNT/PA rods after implantation [78]. Copyright 2020, Elsevier; 2022, Wiley.

osteogenic activity with good cell characteristics such as adhesion, diffusion, proliferation and differentiation. These findings reveal new opportunities for self-healing coatings with corrosion protection. Li et al. [76] have constructed a self-repairing coating with bone immunomodulatory functions on Mg by encapsulating curcumin and sustained-release F^- into mesoporous silica nanocapsules as shown in Fig. 9A. The hybrid coating has a three-layered structure consisting of an MgO inner layer, poly-$_L$-lactide intercalated F-encapsulated mesoporous silica nanocontainer containing curcumin, and DCPD outer layer. Here, F^- contributes to the self-healing functions and curcumin-controlled release produces the bone immunomodulatory effects as shown in Fig. 9B. Compared with the coatings with medium (10FMSN) and lowest (5FMSN) curcumin release, 20FMSN with the highest curcumin release modulate the surrounding immune microenvironment to promote bone differentiation and ECM mineralization of BMSCs achieving enhanced bone integration through contact osteogenesis (Fig. 9C and D). Zhang et al. [77] have prepared electrical-responsive biocompatible coatings based on electrically stimulated smart 8-HQ loaded poly (3,4-ethylenedioxythiophene) (8-HQ@PEDOT) nanocapsules by emulsion polymerization. After applying a higher negative voltage, the concentration of the 8-HQ inhibitor increases reflecting the distinct electrical response. The hybrid coatings possess excellent corrosion resistance and rapid

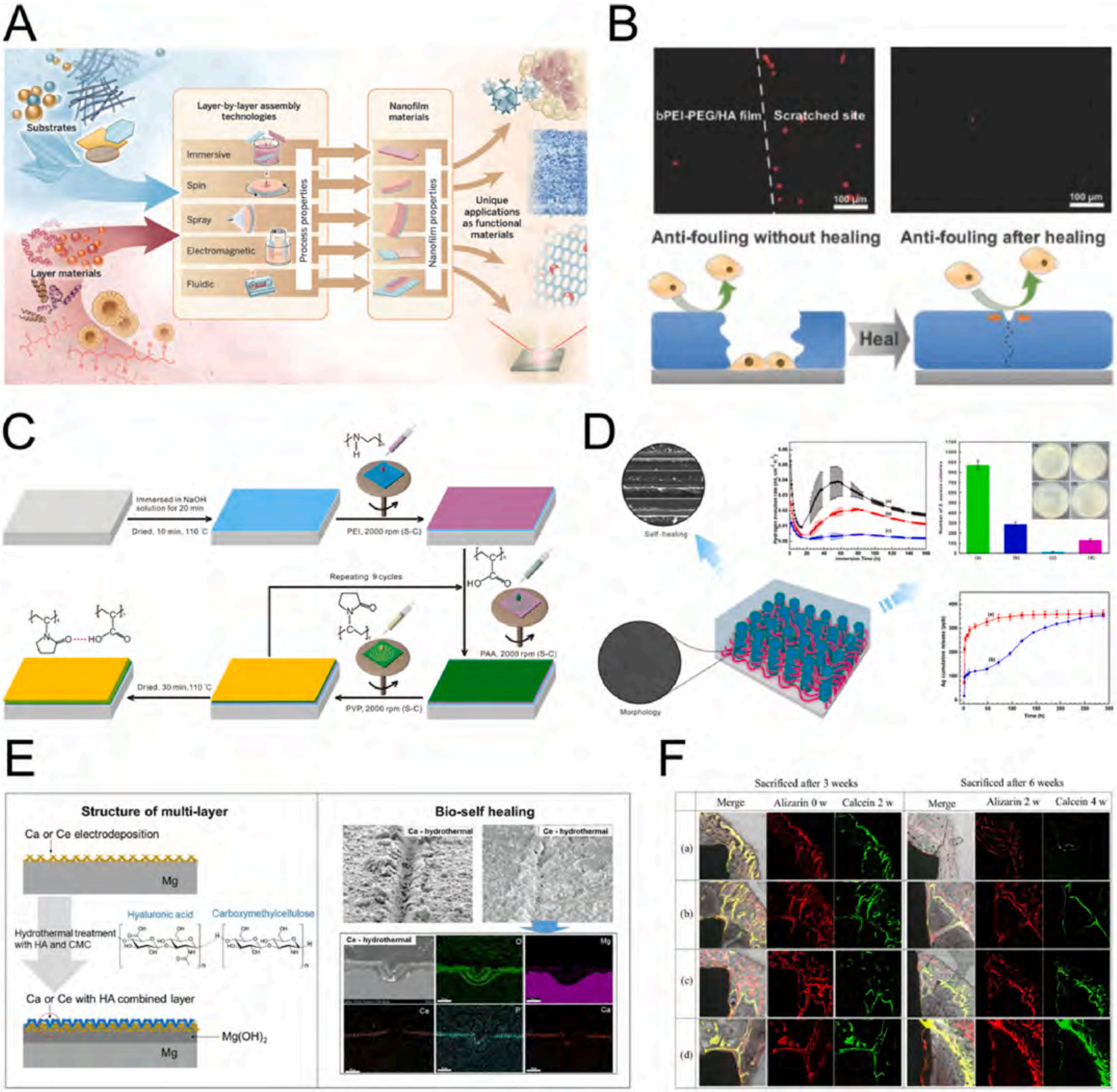

Fig. 10. (A) Layer-by-layer assembly of nanofilms for the preparation of functional materials [180]; (B) Cell adhesion on the scratched and healed films and schematic showing the damage and healing process of the films [184]; (C) Schematic presentation of the spin-assisted later-by-layer technique [185]; (D) Hybrid film developed on the Mg alloy with enhanced corrosion resistance and antibacterial properties in conjunction with self-healing and prolonged the release of Ag^+ [79]; (E) Structure of the multi-layers and scratch tests; (F) Fluorescence microscopy images of the samples after implantation [80]. Copyright 2015, The American Association for the Advancement of Science; 2015, Wiley; 2017, 2018, 2020, Elsevier.

self-healing properties in corrosive media. Many MG-63 cells survive on the coatings, as manifested by a hemolysis rate of 2.68%. The PDAs/8-HQ@PEDOT/polylactic acid coating has remarkable anti-corrosion, self-healing and biocompatibility properties boding well for biological implants. Xia et al. [78] have fabricated a pH stimulus-responsive self-repairing coating on the Mg–1Ca osteo-implant using filamentous proteins. The PA is encapsulated into the halloysite nanotubes (HNT) which are then spin-coated on the Mg substrate along with pH-responsive filamentous proteins. Upon pH increase, the PA inhibitors are released from HNT and cure the defects within 15 min on account of the random coil and alpha helix structure of filamentous protein stretches. In addition, the collagen-membrane fibrous structure of the silk protein has excellent osteogenic ability *in vitro* and *in vivo* as shown in Fig. 9E and F.

3.3. *Multilayered coatings*

Self-healing anticorrosion coatings can provide corrosion protection [179] and layer-by-layer (LbL) assembly of polyelectrolytes has been used to construct self-healing coatings on Mg. Multilayered thin films have garnered scientific interest due to their potential use in biomedicine [180]. The thickness and layer numbers can be tailored to cater to the corresponding functions and the performance of the films depend on the materials as well as assembly technology. Methods such as dipping, spinning, spraying, and multi-step synthesis have been proposed to fabricate multilayered coatings (Fig. 10A) [181–183].

Chen et al. [184] have prepared a healing multilayer film by LbL assembly of PEGylated branched-chain poly (ethyleneimine) (bPEI) and hyaluronic acid (HA). Under physiological conditions, the artificial scratches on the coating surface are repaired quickly and owing to the grafted PEG and low-energy storage modulus of the coating materials, the films resist protein adsorption and cell attachment as shown in Fig. 10B. Zhao et al. [185] have developed a polyvinylpyrrolidone (PVP)/polyacrylic acid (PAA) LbL assembled composite coating by the spin-casting method as shown in Fig. 10C. Potentiodynamic polarization reveals a breakdown potential at 1.16 ± 0.01 V/SCE and good self-healing properties and corrosion resistance [186]. Zhao et al. [79] have prepared silver nanoparticles (AgNPs) and polymethyltrimethoxysilane (PMTMS) multilayered coatings on the AZ31 Mg alloy by LbL assembly and siloxane self-condensation reaction as shown in Fig. 10D. The scratch tests confirm self-repair of the polysiloxane films. EDS shows that the silicon concentration increases during self-healing reflecting diffusion-driven recombination of the crosslinked polysiloxane structure that has been damaged. Addition of AgNPs improves the bacteria-killing properties *in vitro*. Kim et al. [80] have fabricated hyaluronic acid and cerium multi-layer films on degradable Mg implants (Fig. 10E). $Ce(OH)_3/CeO_2$ penetrates the natural polymer HA to offer self-healing and resistance to initial corrosion. Ce shows no toxicity on osteoblasts and differentiation of osteoblasts is the high, especially the HA-HT group. As observed from the Ce + HT sample, implantation into the rat tibia results in stable bone marrow and osteoblast growth (Fig. 10F).

IN summary, multilayered coatings offer excellent flexibility and the layer number can be customized to address specific needs. However, owing to electrostatic interactions or connections (such as hydrogen bonding), the coatings can be damaged by corrosive ions leading to failure. In addition, the preparation of LBL coatings can be relatively complex and time-consuming.

The surface protection effiaacy of coatings can be modified by different ways in order to cater to different applications. Future research of smart coatings is expected to focus on researching and preparing hybrid and multilayered coatings using environmentally friendly and cost-effective processes. Intelligent design, efficient corrosion inhibitors, and healing agents will determine the practicality of the coatings. The advantages and disadvantages of different coating techniques are summarized in Table 3.

Table 3
Comparison of different coating techniques.

Techniques	Advantages	Disadvantages
Conversion coating	The conversion coating has stronger surface adhesion; *In-situ* conversion coating; Harder surface and stronger wear resistance are achieved	Poor toughness and density of conversion coating
Encapsulation coating	The encapsulation coating possesses stimuli-responsive activities; Sustained and controlled release of corrosion inhibitors can be provided; The self-healing behavior can be triggered and controlled precisely	The preparation process is complicated; The aggregation of Nanoparticles is difficult to resolve; The toxicity and degradation of the vector need to be verified
Multilayered coating	The operation process is a bottom-up assembly, which can effectively control the surface structure of the material at the molecular level and nanoscale; The preparation process is simple and the preparation cost is low; Rich types of application components can meet the different functional needs of materials; Not limited by the size and shape of the substrates; The prepared coating is nanoscale ultra-thin and the self-healing behavior is fast.	Material waste during the coating process; lower reliability and reproducibility of the coatings; Relatively long-time consumption

4. Conclusions and outlook

Mg-based alloys are promising temporary candidates for biomedical applications due to the good mechanical properties, biodegradability, and biocompatibility. These materials are expected to corrode completely under physiological conditions eventually, and the byproducts are benign to surrounding tissues [187]. However, excessively rapid degradation of Mg implants hampers wider clinical practice. Emission of hydrogen gas and local alkalization are the other significant drawbacks. Surface modification and coatings can improve the properties of Mg alloys while preserving the excellent bulk attributes. The degradation behavior and bioactivity of Mg implants can be controlled by fabricating suitable coatings. However, considerable challenges remain in the development of advanced functional coatings on biomedical Mg alloys. Bioabsorbable Mg-based implants should offer the proper corrosion protection, self-healing ability, antibacterial properties, biocompatibility, biological effects, and controlled drug release. Until now, it is nearly impossible to design self-healing coatings with all of these functions. Conventional monolayered coatings may not meet the requirements for corrosion protection, self-healing, and multi-functions, but versatile composite coatings can further improve the corrosion resistance and bioactivity. The following aspects should be addressed in future research and development.

1. The binding/fatigue strength, stability, and degradation reaction with surrounding environments are the major concerns for smart self-healing coatings on biomedical Mg alloys but there is insufficient information in the literature. Besides, there have been few studies on the changes of the mechanical strength during degradation *in vivo*. This area requires significant research.
2. Self-healing coatings are generally divided into two categories depending on inhibitor release and external stimuli. Long-term released inhibitors pose a threat to the physiological environment and patients' health, and stimulation does not always repair the defects in a timely and effective manner. Therefore, it is necessary to develop smart self-healing coatings that are eco-friendly and have

long-term durability with sustainable, controllable, and timely self-healing properties.

3. With the expansion of the application of Mg alloys, the development of multilayered and multifunctional coatings is becoming a research hotspot. The preparation method mostly determines its practicality. More features require more complex preparation processes and therefore, it is necessary to identify simple preparation technology for multifunctional coatings.
4. Handling of coated implants is tricky, especially magnesium implants, and coating defects can have serious impact. In this respect, self-reporting healable protective coatings that can automatically indicate coating damage and corrosion before implant fixation surgery are desirable. So far, the development of self-reporting coatings for magnesium alloys is insufficient. To make self-healing coatings more intelligent, self-reporting species should be added. As an animal ethical obligation to reduce and replace animal tests, advanced three-dimensional (3D) *in vitro* models should be developed from data obtained *in vivo* and *in vitro*. Versatile coatings require more complex preparation processes, but they need to be simplified to reduce costs and expedite clinical acceptance.
5. More biosafety assessment should be carried out. For example, chronic renal failure should be investigated using animal models to simulate patients with different levels of glomerular filtration rates (GFRs) and resolve contradictions. The long-term biosafety assessment of Mg implants in animals with renal failure is necessary to address the clinical questions.

Declaration of competing interest

The author (Rongchang Zeng) is an editorial board member for *Smart Materials in Manufacturing* and was not involved in the editorial review or the decision to publish this article.

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References

[1] L. Cai, H.R. Guo, Y.Q. Zhu, F.S. Du, J.T. Qi, L.Y. Cui, C.B. Liu, R.C. Zeng, Biodegradation mechanisms of pure Mg in presence of glucose, vitamin C, and citric acid, Smart Mater. Manuf. 1 (2023), 100014.
[2] L.Y. Han, Z.W. Zhang, J.W. Dai, X. Li, J. Bai, Z.H. Huang, C. Guo, F. Xue, C.L. Chu, The influence of alternating cyclic dynamic loads with different low frequencies on the bio-corrosion behaviours of AZ31B magnesium alloy in vitro, Bioact. Mater. 7 (2022) 263–274.
[3] L.Y. Li, Z. Yang, X.X. Pan, B.X. Feng, R. Yue, B. Yu, Y.F. Zheng, J.Y. Tan, G. Y. Yuan, J. Pei, Incorporating copper to biodegradable magnesium alloy vascular stents via a Cu(II)-Eluting coating for synergistic enhancement in prolonged durability and rapid Re-endothelialization, Adv. Funct. Mater. 32 (47) (2022), 2205634.
[4] J.W. Dai, C.Q. Wu, J.Y. Yang, L. Zhang, Q.S. Dong, L.Y. Han, X. Li, J. Bai, F. Xue, P.K. Chu, C. Chu, Poly-lactic acid coatings on the biomedical WE43 Mg alloy: protection mechanism and ion permeation effects, Prog. Org. Coat. 177 (2023), 107427.
[5] J.L. Wang, J.K. Xu, C. Hopkins, D.H. Chow, L. Qin, Biodegradable magnesium-based implants in orthopedics-A general review and perspectives, Adv. Sci. 7 (8) (2020), 1902443.
[6] K. Qian, Y. Zhang, Q.S. Dong, Y. Shao, Z.J. Cheng, J. Ju, F. Xue, C.L. Chu, D. D. Xia, J. Bai, Enhancement of corrosion resistance and antibacterial properties of PEO coated AZ91D Mg alloy by copper- and phosphate-based sealing treatment, Corros. Sci. 219 (2023), 111218.
[7] V. Tsakiris, C. Tardei, F.M. Clicinschi, Biodegradable Mg alloys for orthopedic implants - a review, J. Magnes. Alloy. 9 (6) (2021) 1884–1905.
[8] C.B. Liu, L. Cheng, B. Qian, L.Y. Cui, R.C. Zeng, Corrosion self-warning and repair tracking of polymeric coatings based on stimulus responsive nanosensors, Nanoscale 14 (23) (2022) 8429–8440.
[9] Y.B. Zhao, P. He, J.Y. Yao, M. Li, B. Wang, L.Y. Han, Z.H. Huang, C. Guo, J. Bai, F. Xue, Y. Cong, W.H. Cai, P.K. Chu, C.L. Chu, pH/NIR-responsive and self-healing coatings with bacteria killing, osteogenesis, and angiogenesis performances on magnesium alloy, Biomaterials 301 (2023), 122237.
[10] D. Jiang, Y.Z. Liu, B. Li, S. Chen, S.Q. Li, W.J. Yang, L.Y. Cui, R.C. Zeng, Insight into degradation and mechanical performance of polyelectrolytes-induced hydroxyapatite interlocking coating on Mg-3Nd-1Li-0.2Zn alloys, Appl. Surf. Sci. 614 (2023), 156041.
[11] L. Cai, D. Mei, Z.Q. Zhang, Y.D. Huang, L.Y. Cui, S.K. Guan, D.C. Chen, M. B. Kannan, Y.F. Zheng, R.C. Zeng, Advances in bioorganic molecules inspired degradation and surface modifications on Mg and its alloys, J. Magnes. Alloy. 10 (3) (2022) 670–688.
[12] Q.S. Dong, Y.Q. Jia, Z.X. Ba, Z.X. Tao, Z.Z. Wang, F. Xue, J. Bai, Exploring the corrosion behaviour of Mn-implanted biomedical Mg, J. Alloy. Compd. 873 (2021), 157939.
[13] C.B. liu, L. Cheng, L.Y. Cui, B. Qian, R.C. Zeng, Corrosion self-diagnosing and self-repairing polymeric coatings based on zeolitic imidazolate framework decorated hydroxyapatite nanocontainer on steel, Chem. Eng. J. 431 (2022), 133476.
[14] N. Singh, U. Batra, K. Kumar, A. Mahapatro, Evaluation of corrosion resistance, mechanical integrity loss and biocompatibility of PCL/HA/TiO2 hybrid coated biodegradable ZM21 Mg alloy, J. Magnes. Alloy. 10 (11) (2022) 3179–3204.
[15] A.M. Zhang, P. Lenin, R.C. Zeng, M.B. Kannan, Advances in hydroxyapatite coatings on biodegradable magnesium and its alloys, J. Magnes. Alloy. 10 (5) (2022) 1154–1170.
[16] S.A. Rahim, M.A. Joseph, T.S. Sampath Kumar, H. T, Recent progress in surface modification of Mg alloys for biodegradable orthopedic applications, Front. Mater. 9 (2022), 848980.
[17] S.Q. Liu, G.H. Li, Y.M. Qi, Z.J. Peng, Y.P. Ye, J. Liang, Corrosion and tribocorrosion resistance of MAO-based composite coating on AZ31 magnesium alloy, J. Magnes. Alloy. 10 (12) (2022) 3406–3417.
[18] Q. Guo, T.L. Wang, T.C. Zhang, L.K. Ouyang, S.J. Yuan, Superhydrophobic double layered MgAl-LDH/epoxy composite coatings for enhanced anticorrosion performance of magnesium alloys, Prog. Org. Coat. 174 (2023), 107300.
[19] J. Chen, T.T. Jiang, X.N. Lan, K. Kang, F.G. Cai, S.D. Ma, Characterization and corrosion resistance of a composite layered double hydroxides film on Mg-4Zn alloy in hank's solution, Mater. Today Phys. 20 (2021), 100474.
[20] K.V. Gaalen, C. Quinn, M. Weiler, F. Gremse, F. Benn, P.E. McHugh, T. J. Vaughan, A. Kopp, Predicting localised corrosion and mechanical performance of a PEO surface modified rare earth magnesium alloy for implant use through in-silico modelling, Bioact. Mater. 26 (2023) 437–451.
[21] M. Schneider, K. Kremmer, W. Fürbeth, S. Lederer, R. Sottor, Sepiolite reinforced PEO layer on AZ 31, Surf. Coat. Technol. 459 (2023), 129369.
[22] J.Y. Yang, Y.B. Zhao, J.W. Dai, L.Y. Han, Q.S. Dong, L. Zhang, J. Bai, F. Xue, P. K. Chu, C.L. Chu, Fabrication and growth mechanism of multilayered hydroxyapatite/organic composite coatings on the WE43 magnesium alloy, Surf. Coat. Technol. 452 (2023), 129125.
[23] H. Chai, L.F. Wang, X.Q. Cao, Q. Zhang, S. Arthanari, H. Lee, G. Huang, B. Xing, L. W. Zheng, H. Zhang, L.W. Lu, H.X. Wang, The effects of chemical conversion parameters on morphology and corrosion performance of calcium phosphate coating on AZ31 alloy, Mater. Chem. Phys. 296 (2023), 127338.
[24] M.M. Sabzehmeidani, M. Kazemzad, Insight into the microstructural characteristics and corrosion properties of AZ31 Mg alloy coated with polyurethane containing nanostructures of copper metal organic frameworks, Mater. Lett. 341 (2023), 134294.
[25] L. Liu, J.L. Lei, T. Liu, B. Shang, X.W. Zhao, J.X. He, F.S. Pan, L.J. Li, Artificial skin with fast and robust self-healing ability for durable intelligent protection of magnesium alloys, Chem. Eng. J. 458 (2023), 141482.
[26] S.K. Yang, R.X. Sun, K.Z. Chen, Self-healing performance and corrosion resistance of phytic acid/cerium composite coating on microarc-oxidized magnesium alloy, Chem. Eng. J. 428 (2022), 131198.
[27] Y.L. Wu, L. Wu, W.H. Yao, Y.H. Chen, Y. Zhou, L.Q. Wang, T. Wu, B. Jiang, A. Atrens, F.S. Pan, ZnAl-LDH@MXene modified by inhibitor as a nanofiller applied on Mg alloys coating protection, Carbon 204 (2023) 36–49.
[28] Y. Wang, D. Yu, K. Ma, C.N. Dai, D.Q. Wang, J.F. Wang, Self-healing performance and corrosion resistance of a bilayer calcium carbonate coating on microarc-oxidized magnesium alloy, Corros. Sci. 212 (2023), 110927.
[29] X.J. Li, Z.Y. Xue, W.T. Sun, J.H. Chu, Q.J. Wang, L.B. Tong, K.S. Wang, Bio-inspired self-healing MXene/polyurethane coating with superior active/passive anticorrosion performance for Mg alloy, Chem. Eng. J. 454 (2023), 140187.
[30] Z. Li, Y. Xu, J.R. Zhang, D.Q. Feng, Y. Fan, D.K. Xu, F.H. Wang, Living marine bacterium Tenacibaculum mesophilum D-6 inhibits crevice corrosion of X70 carbon steel, Corros. Sci. 215 (2023), 111012.
[31] J.W. Feng, Y.K. Pan, M. Yang, C. Fernandez, X.B. Chen, Q.M. Peng, A lactoglobulin-composite self-healing coating for Mg alloys, ACS Appl. Bio. Mater. 4 (9) (2021) 6843–6852.
[32] P.D. Tong, Y.L. Sheng, R.Q. Hou, M. Iqbal, L. Chen, J.G. Li, Recent progress on coatings of biomedical magnesium alloy, Smart Mater. Med. 3 (2022) 104–116.
[33] P.P. Lu, J.Y. Xu, W.J. Tian, C.C. Zhang, S.C. Niu, J. Zhao, W.H. Ming, L.Q. Ren, Robust antifogging coatings with ultra-fast self-healing performances through host-guest strategy, Chem. Eng. J. 465 (2023), 142868.

[34] C. Dry, Procedures developed for self-repair of polymer substrate composite materials, Compos. Struct. 35 (3) (1996) 263–269.

[35] N.K. Kumbhar, M.D. Joshi, V. Kumar, S.S. Hosmani, An impact of the recent developments in coating materials and techniques on the corrosion response of AZ91D alloy: a review, Adv. Eng. Mater. 25 (10) (2023), 2201680.

[36] A. Shanaghi, A.R. Souri, B. Mehrjou, P.K. Chu, Corrosion resistance, nano-mechanical properties, and biocompatibility of Mg-plasma-implanted and plasma-etched Ta/TaN hierarchical multilayered coatings on the nitrided AZ91 Mg alloy, Biomed. Mater. 16 (4) (2021), 045028.

[37] K. Saranya, S. Bhuvaneswari, S. Chatterjee, N. Rajendran, Biocompatible gadolinium-coated magnesium alloy for biomedical applications, J. Mater. Sci. 55 (25) (2020) 11582–11596.

[38] D. Li, D.N. Dai, G.G. Xiong, S.Q. Lan, C. Zhang, Composite nanocoatings of biomedical magnesium alloy implants: advantages, mechanisms, and design strategies, Adv. Sci. (2023), 2300658.

[39] Y.F. Zheng, X. Liu, D.N. Shen, W.T. Li, Y. Cheng, M. Yang, Y.H. Kou, B.G. Jiang, Perceiving the connection between the bone healing process and biodegradation of biodegradable metal implants through precise bioadaptability principle, J. Mater. Sci. Technol. 147 (2023) 132–144.

[40] X.D. Xue, C.Y. Liang, D.H. Wang, F. Peng, The research progress of self-healing coatings for magnesium/magnesium alloy, J. Alloy. Compd. 960 (2023), 170710.

[41] D.D. Zhang, F. Peng, X.Y. Liu, Protection of magnesium alloys: from physical barrier coating to smart self-healing coating, J. Alloy. Compd. 853 (2021), 157010.

[42] D.W. Lee, K. Park, J. Seo, Recent advances in anti-inflammatory strategies for implantable biosensors and medical implants, BioChip J 14 (1) (2020) 48–62.

[43] L. Tian, N. Tang, T. Ngai, C. Wu, R.Y. Ruan, L. Huang, L. Qin, Hybrid fracture fixation systems developed for orthopaedic applications: a general review, J. Orthop. Transl. 16 (2018) 1–13.

[44] M. Ebrahimi, Q.D. Wang, S. Attarilar, A comprehensive review of magnesium-based alloys and composites processed by cyclic extrusion compression and the related techniques, Prog. Mater. Sci. 131 (2023), 101016.

[45] L. Hernández, J. Ramón-Sierra, M. Soria-Castro, Á. Bacelis, G. Rodríguez-Gattorno, E. Ortiz-Vázquez, G. Acosta, Assessment of Mg(OH)2/TiO2 coating in the Mg-Ca-Zn alloy for improved corrosion resistance and antibacterial performance, J. Magnes. Alloy. 11 (1) (2023) 361–378.

[46] H.M. Feng, W. Wang, T. Wang, L. Zhang, W. Li, J. Hou, S.G. Chen, Preparation of dynamic polyurethane networks with UV-triggered photothermal self-healing properties based on hydrogen and ion bonds for antibacterial applications, J. Mater. Sci. Technol. 133 (2023) 89–101.

[47] Y.M. Zhang, N. Li, N. Ling, J.L. Zhang, L. Wang, Enhanced long-term corrosion resistance of Mg alloys by superhydrophobic and self-healing composite coating, Chem. Eng. J. 449 (2022), 137778.

[48] L. Wu, Y.N. Chen, X. Dai, W.H. Yao, J.H. Wu, Z. Xie, B. Jiang, Y. Yuan, F.S. Pan, Corrosion resistance of the GO/ZIF-8 hybrid loading benzotriazole as a multifunctional composite filler-modified MgAlY layered double hydroxide coating, Langmuir 38 (33) (2022) 10338–10350.

[49] J.J. Zhang, X. Zhao, J.F. Wei, B.C. Li, J.P. Zhang, Superhydrophobic coatings with photothermal self-healing chemical composition and microstructure for efficient corrosion protection of magnesium alloy, Langmuir 37 (45) (2021) 13527–13536.

[50] T. Liu, D.W. Zhang, R.J. Zhang, J.K. Wang, L.W. Ma, P. Keil, A. Mol, X.G. Li, Self-healing and corrosion-sensing coatings based on pH-sensitive MOF-capped microcontainers for intelligent corrosion control, Chem. Eng. J. 454 (2023), 140335.

[51] C.L. Zhou, M.F. Pan, S.J. Li, Y.X. Sun, H.J. Zhang, X.H. Luo, Y.L. Liu, H.B. Zeng, Metal organic frameworks (MOFs) as multifunctional nanoplatform for anticorrosion surfaces and coatings, Adv. Colloid. Interfac. 305 (2022), 102707.

[52] H.T. Zheng, Y.C. Zhang, X. He, H.H. Liu, A.Y. Chen, X.F. Xie, Infrared-light-driven self-healing MoS2/polyvinyl alcohol hydrogel with simultaneous enhancement of strength and ductility, J. Alloy. Compd. 918 (2022), 165801.

[53] W.P. Zhao, Z.X. Zhang, J. Hu, X.Q. Feng, J. Xu, Y.M. Wu, S.K. Yan, Robust and ultra-fast self-healing elastomers with hierarchically anisotropic structures and used for wearable sensors, Chem. Eng. J. 446 (2022), 137305.

[54] J.J. Zhang, J.F. Wei, B.C. Li, X. Zhao, J.P. Zhang, Long-term corrosion protection for magnesium alloy by two-layer self-healing superamphiphobic coatings based on shape memory polymers and attapulgite, J. Colloid Interf. Sci. 594 (2021) 836–847.

[55] Y. Wang, X.Y. Zhou, M.H. Yin, J.B. Pu, N.Y. Yuan, J.N. Ding, Superhydrophobic and self-healing Mg-Al layered double hydroxide/silane composite coatings on the Mg alloy surface with a long-term anti-corrosion lifetime, Langmuir 37 (27) (2021) 8129–8138.

[56] M.J. Anjum, J.M. Zhao, H. Ali, M. Tabish, H. Murtaza, G. Yasin, M.U. Malik, W. Q. Khan, A review on self-healing coatings applied to Mg alloys and their electrochemical evaluation techniques, Int. J. Electrochem. Sci. 4 (15) (2020) 3040–3053.

[57] X. Zhang, B.W. Tan, Y.T. Wu, M. Zhang, X. Xie, J.F. Liao, An injectable, self-healing carboxymethylated chitosan hydrogel with mild photothermal stimulation for wound healing, Carbohyd. Polym. 293 (2022), 119722.

[58] H.T. Yu, C. Chen, J.X. Sun, H. Zhang, Y.Y. Feng, M.M. Qin, W. Feng, Highly thermally conductive polymer/graphene composites with rapid room-temperature self-healing capacity, Nano-Micro Lett. 14 (1) (2022) 135.

[59] S. Hiromoto, Self-healing property of hydroxyapatite and octacalcium phosphate coatings on pure magnesium and magnesium alloy, Corros. Sci. 100 (2015) 284–294.

[60] Q.S. Dong, X.X. Zhou, Y.J. Feng, K. Qian, H. Liu, M.M. Lu, C.L. Chu, F. Xue, J. Bai, Insights into self-healing behaviour and mechanism of dicalcium phosphate dihydrate coating on biomedical Mg, Bioact. Mater. 6 (1) (2021) 158–168.

[61] C.Y. Huang, D. Li, J.L. Song, K. Chen, X.L. Wang, F. Zhao, X.N. Gu, X.H. Xie, Y. B. Fan, Dual-functional coatings on magnesium alloys: enhancing corrosion behaviour under stress and osteogenic effect in osteoporotic rats, Appl. Mater. Today 30 (2023), 101723.

[62] Y.K. Kim, Y.S. Jang, S.Y. Kim, M.H. Lee, Functions achieved by the hyaluronic acid derivatives coating and hydroxide film on bio-absorbed Mg, Appl. Surf. Sci. 473 (2019) 31–39.

[63] L. Liu, Q.Y. Yang, L. Huang, X.M. Liu, Y.Q. Liang, Z.D. Cui, X.J. Yang, S.L. Zhu, Z. Y. Li, Y.F. Zheng, K.W.W. Yeung, S.L. Wu, The effects of a phytic acid/calcium ion conversion coating on the corrosion behaviour and osteoinductivity of a magnesium- strontium alloy, Appl. Surf. Sci. 484 (2019) 511–523.

[64] Q.S. Dong, J.W. Dai, K. Qian, H. Liu, X.X. Zhou, Q.Q. Yao, M.M. Lu, C.L. Chu, F. Xue, J. Bai, Dual self-healing inorganic-organic hybrid coating on biomedical Mg, Corros. Sci. 200 (2022), 110230.

[65] Y. Chen, J.H. Dou, Z.F. Pang, Z.Q. Zheng, H.J. Yu, C.Z. Chen, Ag-containing antibacterial self-healing micro-arc oxidation coatings on Mg–Zn–Sr alloys, Surf. Eng. 37 (7) (2020) 926–941.

[66] Z.J. Jia, P. Xiong, Y.Y. Shi, W.H. Zhou, Y. Cheng, Y.F. Zheng, T.F. Xi, S.C. Wei, Inhibitor encapsulated, self-healable and cytocompatible chitosan multilayer coating on biodegradable Mg alloy: a pH-responsive design, J. Mater. Chem. B 4 (14) (2016) 2498–2511.

[67] J. Li, N.A. He, J.Y. Li, Q.Y. Fu, M.C. Feng, W.H. Jin, W. Li, Y. Xiao, Z.T. Yu, P. K. Chu, A silicate-loaded MgAl LDH self-healing coating on biomedical Mg alloys for corrosion retardation and cytocompatibility enhancement, Surf. Coat. Technol. 439 (2022), 128442.

[68] J. Ouyang, X.Z. Hong, Y. Gao, Retardation and self-repair of erosion pits by a two-stage barrier on bioactive-glass/layered double hydroxide coating of biomedical magnesium alloys, Surf. Coat. Technol. 405 (2021), 126562.

[69] C.Y. Li, L. Gao, X.L. Fan, R.C. Zeng, D.C. Chen, K.Q. Zhi, In vitro degradation and cytocompatibility of a low temperature in-situ grown self-healing Mg-Al LDH coating on MAO-coated magnesium alloy AZ31, Bioact. Mater. 5 (2) (2020) 364–376.

[70] W.J. Li, Y. Su, L. Ma, S.J. Zhu, Y.F. Zheng, S.K. Guan, Sol-gel coating loaded with inhibitor on ZE21B Mg alloy for improving corrosion resistance and endothelialization aiming at potential cardiovascular application, Colloid. Surface. B 207 (2021), 111993.

[71] P. Xiong, J.L. Yan, P. Wang, Z.J. Jia, W.H. Zhou, W. Yuan, Y.Y. Li, Y. Liu, Y. Cheng, D.F. Chen, Y.F. Zheng, A pH-sensitive self-healing coating for biodegradable magnesium implants, Acta Biomater. 98 (2019) 160–173.

[72] Z.Q. Zhang, P.D. Tong, L. Wang, Z.H. Qiu, J.A. Li, H. Li, S.K. Guan, C.G. Lin, H. Y. Wang, One-step fabrication of self-healing poly(thioctic acid) coatings on ZE21B Mg alloys for enhancing corrosion resistance, anti-bacterial/oxidation, hemocompatibility and promoting re-endothelialization, Chem. Eng. J. 451 (2023), 139096.

[73] X.J. Ji, G.F. Luan, J.C. Lyu, L.Y. Cui, S.Q. Li, R.C. Zeng, Z.L. Wang, Corrosion resistance and tunable release of ciprofloxacin-loaded multilayers on magnesium alloy: effects of SiO2 nanoparticles, Appl. Surf. Sci. 508 (2020), 145240.

[74] Y.B. Zhao, Z. Zhang, L.Q. Shi, F. Zhang, S.Q. Li, R.C. Zeng, Corrosion resistance of a self-healing multilayer film based on SiO2 and CeO2 nanoparticles layer-by-layer assembly on Mg alloys, Mater. Lett. 237 (2019) 14–18.

[75] P. Xiong, Z.J. Jia, W.H. Zhou, J.L. Yan, P. Wang, W. Yuan, Y.Y. Li, Y. Cheng, Z. P. Guan, Y.F. Zheng, Osteogenic and pH stimuli-responsive self-healing coating on biomedical Mg-1Ca alloy, Acta Biomater. 94 (2019) 336–350.

[76] B. Li, R. Huang, J. Ye, L. Liu, L. Qin, J.H. Zhou, Y.F. Zheng, S.L. Wu, Y. Han, A self-healing coating containing curcumin for osteoimmunomodulation to ameliorate osseointegration, Chem. Eng. J. 403 (2020), 126323.

[77] Z.H. Zhang, X.Y. Wang, H.T. Xia, H.Y. Li, Z.H. Chen, W.Z. Yang, Electrical-responsive biocompatible coatings for highly corrosion-resistance and self-healing performance on AZ31 Mg alloy, J. Ind. Eng. Chem. 121 (2023) 179–189.

[78] D.D. Xia, Z.J. Jia, Y.N. Shen, Y.F. Zheng, Y. Cheng, P. Xiong, S.K. Guan, Y.Q. Xu, F. Yang, Y.S. Liu, Y.S. Zhou, pH stimuli-responsive, rapidly self-healable coatings enhanced the corrosion resistance and osteogenic differentiation of Mg-1Ca osteoimplant, Small 18 (36) (2022), 2106056.

[79] Y.B. Zhao, L.Q. Shi, X.J. Ji, J.C. Li, Z.Z. Han, S.Q. Li, R.C. Zeng, F. Zhang, Z. L. Wang, Corrosion resistance and antibacterial properties of polysiloxane modified layer-by-layer assembled self-healing coating on magnesium alloy, J. Colloid Interf. Sci. 526 (2018) 43–50.

[80] Y.K. Kim, S.Y. Kim, Y.S. Jang, I.S. Park, M.H. Lee, Bio-corrosion behaviours of hyaluronic acid and cerium multi-layer films on degradable implant, Appl. Surf. Sci. 515 (2020), 146070.

[81] S. Farshid, M. Kharaziha, M. Atapour, A self-healing and bioactive coating based on duplex plasma electrolytic oxidation/polydopamine on AZ91 alloy for bone implants, J. Magnes. Alloy. 11 (2) (2023) 592–606.

[82] Y.C. Xu, Y.T. Guo, G.Y. Li, J.S. Lian, Biodegradable phytic acid conversion coatings on magnesium alloy for temporary orthopedic implant: a review, Prog. Org. Coat. 169 (2022), 106920.

[83] Y. Wang, Z.P. You, K. Ma, C.N. Dai, D.Q. Wang, J.F. Wang, Corrosion resistance of a superhydrophobic calcium carbonate coating on magnesium alloy by ultrasonic cavitation-assisted chemical conversion, Corros. Sci. 211 (2023), 110841.

[84] Z.Y. Zhang, Y.L. An, X.S. Wang, L.Y. Cui, S.Q. Li, C.B. Liu, Y.H. Zou, F. Zhang, R. C. Zeng, In vitro degradation, photo-dynamic and thermal antibacterial activities of Cu-bearing chlorophyllin-induced Ca-P coating on magnesium alloy AZ31, Bioact. Mater. 18 (2022) 284–299.

[85] B. Pillado, B. Mingo, R.D. Olmo, E. Matykina, A.M. Kooijman, Y. Gonzalezgarcia, R. Arrabal, M. Mohedano, LDH conversion films for active protection of AZ31 Mg alloy, J. Magnes. Alloy. 11 (1) (2023) 201–216.

[86] N. Singh, U. Batra, K. Kumar, N. Ahuja, A. Mahapatro, Progress in bioactive surface coatings on biodegradable Mg alloys:A critical review towards clinical translation, Bioact. Mater. 19 (2023) 717–757.

[87] J.X. Yang, X.D. Wang, Y.R. Cai, X.Y. Yang, Corrosion resistance and electrical conductivity of V/Ce conversion coating on magnesium alloy AZ31B, Int. J. Min. Met. Mater. 30 (4) (2023) 653–659.

[88] Y. Zhang, Y.J. Guo, J.Y. Dai, L. Zhao, L.P. Wu, Fabrication of hazenite conversion coating on AZ31 Mg alloy, Surf. Coat. Technol. 435 (2022), 128249.

[89] Z.W. Song, Z.H. Xie, L.F. Ding, Y.J. Zhang, X.Y. Hu, Preparation of corrosion-resistant MgAl-LDH/Ni composite coating on Mg alloy AZ31B, Colloid. Surface. A 632 (2022), 127699.

[90] T. Li, Z.J. Leng, S.F. Wang, X.T. Wang, R. Ghomashchi, Y.S. Yang, J.X. Zhou, Comparison of the effects of pre-activators on morphology and corrosion resistance of phosphate conversion coating on magnesium alloy, J. Magnes. Alloy. 10 (12) (2022) 3485–3494.

[91] A.C. Bouali, M. Serdechnova, K.A. Yasakau, A. Lutz, G. Wiese, H. Terryn, M.G. S. Ferreira, M.L. Zheludkevich, The role of Cu-based intermetallic on the direct growth of a ZnAl LDH film on AA2024, J. Electrochem. Soc. 169 (8) (2022), 081501.

[92] J. Dziková, S. Fintová, D. Kajánek, Z. Florková, P. Doleal, Characterization and corrosion properties of fluoride conversion coating prepared on AZ31 magnesium alloy, Coatings 11 (6) (2021) 675.

[93] D. Saran, A. Kumar, S. Bathula, D. Klaumünzer, K.K. Sahu, Review on the phosphate-based conversion coatings of magnesium and its alloys, Int. J. Min. Met. Mater. 29 (7) (2022) 1435–1452.

[94] G.S. Hikku, C. Arthi, R.B.J. Robert, K. Jeyasubramanian, R. Murugesan, Calcium phosphate conversion technique: a versatile route to develop corrosion resistant hydroxyapatite coating over Mg/Mg alloys based implants, J. Magnes. Alloy. 10 (7) (2022) 1959–1986.

[95] P.F. Yu, Y. Lei, Z.L. Luan, Y.G. Zhao, H.P. Peng, Effect on the surface anticorrosion and corrosion protection mechanism of integrated rust conversion coating for enhanced corrosion protection, ACS Omega 7 (10) (2022) 8995–9003.

[96] Y.H. Wang, L.S. Guo, P. Zhou, T. Zhang, F.H. Wang, Corrosion resistance of Ni- P composite coating on Mg alloy:critical role of the intermediate phosphate conversion coating layer for suppressing galvanic couple formation, Aerospance SH 39 (1) (2022) 102–110.

[97] X.X. Zhang, Y.P. Zhang, Y. Lv, Z.H. Dong, L. Yang, E.L. Zhang, T. Hashimoto, X. R. Zhou, PEO coating on Mg-Ag alloy: the incorporation and release of Ag species, J. Magnes. Alloy. 11 (6) (2023) 2182–2195.

[98] J. Park, S.H. Um, Y. Seo, J. Lee, Y.C. Kim, M.R. Ok, S.W. Hwang, J.Y. Sun, H. S. Han, H. Jeon, Improving hydroxyapatite coating ability on biodegradable metal through laser-induced hydrothermal coating in liquid precursor: Application in orthopedic implants, Bioact. Mater. 25 (2023) 796–806.

[99] P. Zhou, B.X. Yu, Y.J. Hou, G.Q. Duan, L.X. Yang, B. Zhang, T. Zhang, F.H. Wang, Revisiting the cracking of chemical conversion coating on magnesium alloys, Corros. Sci. 178 (2021), 109069.

[100] X.Y. Liu, D. Vonk, K. Kisslinger, X. Tong, G. Halada, S. Petrash, K. Foster, Y.C. K. Chen-Wiegart, Unraveling the formation mechanism of a hybrid Zr-based chemical conversion coating with organic and copper compounds for corrosion inhibition, ACS Appl. Mater. Interfaces 13 (4) (2021) 5518–5528.

[101] J. Yang, P. Zhang, L.H. Su, F.A. Yan, Z. Yong, The one-step preparation and corrosion resistance of a Ca-Mn-Zn ternary phosphate conversion coating on magnesium alloy AZ91D, Anti-Corros. Method. M. 69 (4) (2022) 341–350.

[102] X.B. Chen, N. Birbilis, T.B. Abbott, Review of corrosion-resistant conversion coatings for magnesium and its alloys, Corrosion 67 (3) (2011), 035005.

[103] R.G. Buchheit, S.B. Mamidipally, P. Schmutz, H. Guan, Active corrosion protection in chromate and chromate-free conversion coatings, Coatings 58 (1) (2002) 3–14.

[104] M. Kim, L.N. Brewer, G.W. Kubacki, Microstructure and corrosion resistance of chromate conversion coating on cold sprayed aluminum alloy 2024, Surf. Coat. Technol. 460 (2023), 129423.

[105] X.Y. Li, Y.J. Su, X.S. Lang, L. Li, C.G. Yao, K.D. Cai, High-performance surface optimized Mg-doped V2O5 (Mg@V2O5) cathode material via a surfactant-assisted hydrothermal technology for lithium-ion and lithium-sulfur batteries, Ionics 28 (4) (2022) 1511–1521.

[106] Y.B. Luo, W. Zhu, Z.J. Su, W.F. Li, A.H. Yi, K. Li, K. Chen, J.H. Song, Y.X. Duan, Y. M. Li, Formation and characterization analysis of a chromium-free conversion coating on Mg alloy prepared in a Mo-Mn-V based weak acid solution, Surf. Coat. Technol. 458 (2023), 129313.

[107] S.A. Salman, M.K. Gouda, Characterization and corrosion behaviour of vanadium-based conversion coating on AZ31 magnesium alloy, Mater. Today Proc. 62 (2022) 611–614.

[108] X.Y. Lu, J. Hu, Y. Zhao, M. Wang, X. Wang, Z.H. Ye, Z.H. Zheng, J.J. Hu, X. G. Feng, Corrosion behaviour of stannate conversion coatings on AZ31B alloys and their initial discharge performance as anodes for seawater batteries, J. Electrochem. Soc. 168 (8) (2021), 080513.

[109] H.D. Zhu, X.R. Guan, Z.C. Shao, Effect of EDTA on corrosion resistance of stannate conversion coatings on magnesium alloy, Electroplat. Pollut. Control 39 (5) (2019) 45–47.

[110] Y.R. Cai, X.D. Wang, L.L. Wan, J.X. Yang, Y.F. Gao, Preparation of antimony-doped stannate chemical conversion coating on AZ31B Mg alloy, T. Indian I. Metals 73 (7) (2020) 1891–1898.

[111] M. Zhao, J.G. Li, G.P. He, H.L. Xie, Y.A. Fu, Nano Al2O3/phosphate composite conversion coating formed on magnesium alloy for enhancing corrosion resistance, J. Electrochem. Soc. 160 (11) (2013) 553–559.

[112] L.T. Trang, N.Q. Cao, S. Hiromoto, O. Minho, E. Kobayashi, Formation and corrosion behaviour of calcium phosphate coating layers on ZK60 alloy coated at various pH conditions by chemical conversion method, Surf. Coat. Technol. 444 (2022), 128639.

[113] D.W. Zhao, C.M. Du, K.Q. Zuo, Y.X. Zhao, X.Q. Xu, Y.B. Li, S. Tian, H.R. Yang, Y. P. Lu, L. Cheng, G.Y. Xiao, Calcium-Zinc phosphate chemical conversion coating facilitates the osteointegration of biodegradable zinc alloy implants by orchestrating macrophage phenotype, Adv. Healthc. Mater. 12 (9) (2023), 2202537.

[114] H. Zhang, K. Liu, M.M. Lu, L. Liu, Y.Z. Yan, Z.Z. Chu, Y.R. Ge, T. Wang, J. Qiu, S. S. Bu, C.B. Tang, Micro/nanostructured calcium phytate coating on titanium fabricated by chemical conversion deposition for biomedical application, Mater. Sci. Eng. C 118 (2018), 111402, 2020.

[115] Q. Li, P.B. Liang, Y.Q. Li, Z.P. Ye, B. Zhang, C.Y. Ji, The use of phytic acid conversion coating to enhance the corrosion resistance of AZ91D magnesium alloy, Int. J. Electrochem. Sci. 17 (9) (2022), 22095.

[116] Y. Wu, R. Yan, Y.Y. Duan, J. Qiu, T. Chen, H.Y. Ma, An environmental-friendly tannic acid/Zn conversion film with a good corrosion protection for iron, Surf. Interf. 24 (2021), 101078.

[117] W. Zhu, F.R. Chen, Y.B. Luo, Z.J. Su, W.F. Li, A.H. Yi, Z.M. Liao, K. Li, K. Chen, Y. W. Hu, Y.S. Xu, S.A. Guo, Vanadium and tannic acid-based composite conversion coating for 6063 aluminum alloy, Front. Mater. 8 (2021), 802468.

[118] J.Q. Feng, Y.B. Wang, X.L. Lin, M.H. Bian, Y.Z. Wei, SECM in situ investigation of corrosion and self-healing behaviour of trivalent chromium conversion coating on the zinc, Surf. Coat. Technol. 459 (2023), 129411.

[119] R.X. Sun, S.K. Yang, T. Lv, Corrosion behaviour of AZ91D magnesium alloy with a calcium-phosphate-vanadium composite conversion coating, Coatings 9 (6) (2019) 379.

[120] B. Vaghefinazari, E. Wierzbicka, P. Visser, R. Posner, R. Arrabal, E. Matykina, M. Mohedano, C. Blawert, M. Zheludkevich, S. Lamaka, Chromate-free corrosion protection strategies for magnesium alloys-A review: PART I-Pre-Treatment and conversion coating, Materials 15 (23) (2022) 8676.

[121] G.S. Pereira, O.M. Prada Ramirez, P.R.T. Avila, J.A. Avila, H.C. Pinto, M. H. Miyazaki, H.G. de Melo, W.W. Bose Filho, Cerium conversion coating and sol-gel coating for corrosion protection of the WE43 Mg alloy, Corros. Sci. 206 (2022), 110527.

[122] L. Liu, S. Dong, F.H. Wang, X.B. Chen, J. Dong, Fabrication of uniform and anti-corrosion layered double hydroxides film on Mg-Gd-Y-Zn-Zr alloy through solution pH tailoring, Electrochim. Acta 411 (2022), 140057.

[123] J.R. Culham, P. Niessen, Low cost solar selective chromate conversion coatings applied to stainless steel substrates, J. Sol. Energy Eng. 102 (3) (1980) 188–191.

[124] S. Pommiers-Belin, J. Frayret, A. Uhart, J. Ledeuil, J.C. Dupin, A. Castetbon, M. Potin-Gautier, Determination of the chemical mechanism of chromate conversion coating on magnesium alloys EV31A, Appl. Surf. Sci. 298 (2014) 199–207.

[125] J.L. Zhang, C.D. Gu, Y.Y. Tong, W. Yan, J.P. Tu, A smart superhydrophobic coating on AZ31B magnesium alloy with self-healing effect, Adv. Mater. Interfaces 3 (14) (2016), 1500694.

[126] J.J. Xu, S.A. Xu, Enhancing the interfacial bond strength of aluminum/polymer laminated film of the soft package lithium-ion battery through polydopamine surface modification, J. Appl. Polym. Sci. 140 (8) (2022), 53530.

[127] C.W. Chen, S.L. Aktug, C.J. Chang, Y.L. Lee, M.D. Ger, S.Y. Jian, A chromate- free and convenient route to fabricate thin and compact conversion coating for corrosion protection on LZ91 magnesium alloy, Nanomaterials 12 (9) (2022) 1614.

[128] M.D. Helal Hossain, N. Hossain, M. Asaduzzaman Chowdhury, M. Arefin Kowser, M. Masud Rana, Surface synthesization of magnesium alloys for improving corrosion resistance and implant applications, Arab. J. Chem. 16 (2) (2023), 104465.

[129] I. Cacciotti, Cationic and anionic substitutions in hydroxyapatite. Handbook of Bioceramics and Biocomposites 46, 2016, pp. 145–211.

[130] M. Catauro, F. Bollino, R. Giovanardi, P. Veronesi, Modification of Ti6A14V implant surfaces by biocompatible TiO2/PCL hybrid layers prepared via sol-gel dip coating: structural characterization, mechanical and corrosion behaviour, Mater. Sci. Eng. C. 74 (2017) 501–507.

[131] H.R. Bakhsheshi-Rad, E. Hamzah, M.R. Abdul-Kadir, S.N. Saud, M. Kasiri-Asgarani, R. Ebrahimi-Kahrizsangi, The mechanical properties and corrosion behaviour of double-layered nano hydroxyapatite-polymer coating on Mg-Ca alloy, J. Mater. Eng. Perform. 24 (10) (2015) 4010–4021.

[132] M. Rahman, Y.C. Li, C.E. Wen, Realization and characterization of double-layer Ca-P coating on WE43 Mg alloy for biomedical applications, Surf. Coat. Technol. 398 (1) (2020), 126901.

[133] K.L. Lin, C.T. Wu, J. Chang, Advances in synthesis of calcium phosphate crystals with controlled size and shape, Acta Biomater. 10 (10) (2014) 4071–4102.

[134] Y.C. Xu, Y.T. Guo, G.Y. Li, J.S. Lian, Biodegradable phytic acid conversion coatings on magnesium alloy for temporary orthopedic implant: a review, Prog. Org. Coat. 169 (2022), 106920.

[135] N. Singh, U. Batra, K. Kumar, N. Ahuja, A. Mahapatro, Progress in bioactive surface coatings on biodegradable Mg alloys: a critical review towards clinical translation, Bioact. Mater. 19 (2023) 717–757.

[136] T.F. da Conceicao, N. Scharnagl, W. Dietzel, D. Hoeche, K.U. Kainer, Study on the interface of PVDF coatings and HF-treated AZ31 magnesium alloy: determination

of interfacial interactions and reactions with self-healing properties, Corros. Sci. 53 (2) (2011) 712–719.
[137] Q.F. Zong, L.D. Wang, W. Sun, G.C. Liu, Active deposition of bis (8-hydroxyquinoline) magnesium coating for enhanced corrosion resistance of AZ91D alloy, Corros. Sci. 89 (2014) 127–136.
[138] X. Hou, Y.X. Sang, L.H. Dong, The improved effect and its mechanism of phytic acid on DSS-induced UC mice, Life Sci. 311 (2022), 121139.
[139] A. Kumar, B. Singh, P. Raigond, C. Sahu, U.N. Mishra, S. Sharma, M.K. Lal, Phytic acid: blessing in disguise, a prime compound required for both plant and human nutrition, Food Res. Int. 142 (2021), 110193.
[140] F. Brouns, Phytic acid and whole grains for health controversy, Nutrients 14 (1) (2021), 14010025.
[141] R.Y. Zhang, S. Cai, G.H. Xu, H. Zhao, Y. Li, X.X. Wang, K. Huang, M.G. Ren, X. D. Wu, Crack self-healing of phytic acid conversion coating on AZ31 magnesium alloy by heat treatment and the corrosion resistance, Appl. Surf. Sci. 313 (2014) 896–904.
[142] L. Liu, Q.Y. Yang, L. Huang, X.M. Liu, Y.Q. Liang, Z.D. Cui, X.J. Yang, S.L. Zhu, Z. Y. Li, Y.F. Zheng, K.W.W. Yeung, S.L. Wu, The effects of a phytic acid/calcium ion conversion coating on the corrosion behaviour and osteoinductivity of a magnesium- strontium alloy, Appl. Surf. Sci. 484 (2019) 511–523.
[143] S.Q. Liu, G.H. Li, Y.M. Qi, Z.J. Peng, Y.P. Ye, J. Liang, Corrosion and tribocorrosion resistance of MAO-based composite coating on AZ31 magnesium alloy, J. Magnes. Alloy. 10 (12) (2022) 3406–3417.
[144] B.L. Jiang, Y.F. Ge, Micro-arc oxidation (MAO) to improve the corrosion resistance of magnesium (Mg) alloys, Corros. Prev. Magnes. Alloy (2013) 163–196.
[145] L. Zhang, J.Q. Zhang, C.F. Chen, Y.H. Gu, Advances in microarc oxidation coated AZ31 Mg alloys for biomedical applications, Corros. Sci. 91 (2015) 7–28.
[146] Y. Chen, J.H. Dou, Z.F. Pang, Z.Q. Zheng, H.J. Yu, C.Z. Chen, Ag-containing antibacterial self-healing micro-arc oxidation coatings on Mg–Zn–Sr alloys, Surf. Eng. 37 (7) (2020) 926–941.
[147] B. Li, R. Huang, J. Ye, L. Liu, L. Qin, J.H. Zhou, Y.F. Zheng, S.L. Wu, Y. Han, A self-healing coating containing curcumin for osteoimmunomodulation to ameliorate osseointegration, Chem. Eng. J. 403 (2020), 126323.
[148] L.M. Calado, M.G. Taryba, Y. Morozov, M.J. Carmezim, M.F. Montemor, Novel smart and self-healing cerium phosphate-based corrosion inhibitor for AZ31 magnesium alloy, Corros. Sci. 170 (2020), 108648.
[149] S.Q. Liu, Z.X. Li, Q.L. Yu, Y.M. Qi, Z.J. Peng, J. Liang, Dual self-healing composite coating on magnesium alloys for corrosion protection, Chem. Eng. J. 424 (2021), 130551.
[150] Z. Gu, J.J. Atherton, Z.P. Xu, Hierarchical layered double hydroxide nanocomposites: structure, synthesis and applications, Chem. Commun. 51 (15) (2015) 3024–3036.
[151] M.Q. Zhao, Q. Zhang, J.Q. Huang, F. Wei, Hierarchical nanocomposites derived from nanocarbons and layered double hydroxides - properties, synthesis, and applications, Adv. Funct. Mater. 22 (4) (2012) 675–694.
[152] R.Z. Ma, Z.P. Liu, L. Li, N. Iyi, T. Sasaki, Exfoliating layered double hydroxides in formamide: a method to obtain positively charged nanosheets, J. Mater. Chem. 16 (39) (2006) 3809–3813.
[153] J.F. Yu, Q. Wang, D. O'Hare, L.Y. Sun, Preparation of two dimensional layered double hydroxide nanosheets and their applications, Chem. Soc. Rev. 46 (19) (2017) 5950–5974.
[154] R.C. Zeng, Z.G. Liu, F. Zhang, S.Q. Li, Q.K. He, H.Z. Cui, E.H. Han, Corrosion resistance of in-situ Mg–Al hydrotalcite conversion film on AZ31 magnesium alloy by one-step formation, T. Nonferr. Metal. Soc. 25 (6) (2015) 1917–1925.
[155] F.Z. Zhang, M. Sun, S.L. Xu, L.L. Zhao, B.W. Zhang, Fabrication of oriented layered double hydroxide films by spin coating and their use in corrosion protection, Chem. Eng. J. 141 (2008) 362–367.
[156] J.L. Chen, L. Fang, F. Wu, J. Xie, J. Hu, B. Jiang, H.J. Luo, Corrosion resistance of a self-healing rose-like MgAl-LDH coating intercalated with aspartic acid on AZ31 Mg alloy, Prog. Org. Coat. 136 (2019), 105234.
[157] C.D. Ding, Y. Tai, D. Wang, L.H. Tan, J.J. Fu, Superhydrophobic composite coating with active corrosion resistance for AZ31B magnesium alloy protection, Chem. Eng. J. 357 (2019) 518–532.
[158] Y.N. Chen, L. Wu, W.H. Yao, Y.H. Chen, Z.Y. Zhong, W.J. Ci, J.H. Wu, Z.H. Xie, Y. Yuan, F.S. Pan, A self-healing corrosion protection coating with graphene oxide carrying 8-hydroxyquinoline doped in layered double hydroxide on a micro-arc oxidation coating, Corros. Sci. 194 (2021), 109941.
[159] L.X. Li, Z.H. Xie, C. Fernandez, L. Wu, D. Cheng, X.H. Jiang, C.J. Zhong, Development of a thiophene derivative modified LDH coating for Mg alloy corrosion protection, Electrochim. Acta 330 (2020), 135186.
[160] N.A. Johari, J. Alias, A. Zanurin, N.S. Mohamed, N.A. Alang, M.Z.M. Zain, Recent progress of self-healing coatings for magnesium alloys protection, J. Coat. Technol. Res. 19 (3) (2022) 757–774.
[161] N.S. Mohamed, J. Alias, N.A. Johari, N.A. Alang, A.H. Ahmad, Development of smart self-healing coating for the corrosion protection of magnesium alloys: a brief review, J. Adhes. Sci. Technol. 1 (2022) 1–19.
[162] P. Xiong, Z.J. Jia, W.H. Zhou, J.L. Yan, P. Wang, W. Yuan, Y.Y. Li, Y. Cheng, Z. P. Guan, Y.F. Zheng, Osteogenic and pH stimuli-responsive self-healing coating on biomedical Mg-1Ca alloy, Acta Biomater. 94 (2019) 336–350.
[163] D. Liu, E.H. Han, Y.W. Song, D.Y. Shan, Enhancing the self-healing property by adding the synergetic corrosion inhibitors of Na3PO4 and 2-mercaptobenzothiazole into the coating of Mg alloy, Electrochim. Acta 323 (2019), 134796.
[164] J.L. Liu, D.S. Yan, Z.H. Zhang, Y.L. Wang, D.L. Song, T. Zhang, J.Y. Liu, F. He, M. Zhang, J. Wang, Eco-friendly silane as corrosion inhibitor for dual self-healing anticorrosion coatings, J. Coat. Technol. Res. 19 (5) (2022) 1381–1391.
[165] J.H. Song, X.F. Cui, J. Guo, Z.B. Cai, E.B. Liu, X.L. Li, Y.B. Chen, B.W. Lu, Self-healing conversion coating with gelatin–chitosan microcapsules containing inhibitor on AZ91D alloy, Surf. Eng. 34 (1) (2018) 79–84.
[166] Y. Li, S. Cai, S.B. Shen, G.H. Xu, F.Y. Zhang, F.W. Wang, Self-healing hybrid coating of phytic acid/silane for improving the corrosion resistance of magnesium alloy, J. Coat. Technol. Res. 15 (3) (2018) 571–581.
[167] H.C. Qian, D.K. Xu, C.W. Du, D.W. Zhang, X.G. Li, L.Y. Huang, L.P. Deng, Y.C. Tu, J.M.C. Mol, H.A. Terryn, Dual-action smart coatings with a self-healing superhydrophobic surface and anti-corrosion properties, J. Mater. Chem. A 5 (5) (2017) 2355–2364.
[168] L.M. Calado, M.G. Taryba, M.J. Carmezim, F. Montemor, Self-healing ceria-modified coating for corrosion protection of AZ31 magnesium alloy, Corros. Sci. 142 (2018) 12–21.
[169] A. Yabuki, M. Sakai, Self-healing coatings of inorganic particles using a pH-sensitive organic agent, Corros. Sci. 53 (2) (2011) 829–833.
[170] D.S. Yan, Y.L. Wang, J.L. Liu, D.L. Song, T. Zhang, J.Y. Liu, F. He, M. Zhang, J. Wang, Self-healing system adapted to different pH environments for active corrosion protection of magnesium alloy, J. Alloy. Compd. 824 (2020), 153918.
[171] G. Zhang, L. Wu, A.T. Tang, X.X. Ding, B. Jiang, A. Atrens, F.S. Pan, Smart epoxy coating containing zeolites loaded with Ce on a plasma electrolytic oxidation coating on Mg alloy AZ31 for active corrosion protection, Prog. Org. Coat. 132 (2019) 144–147.
[172] J.K. Wang, W.M. Tan, H. Yang, X.X. Rao, X.L. Luo, L.W. Ma, C.H. Ren, A. Mol, D. W. Zhang, Towards weathering and corrosion resistant, self-warning and self-healing epoxy coatings with tannic acid loaded nanocontainers, npj Mater, Degrad. 7 (1) (2023) 39.
[173] Z.H. Chen, W.Z. Yang, Y. Chen, X.S. Yin, Y. Liu, Smart coatings embedded with polydopamine-decorated layer-by-layer assembled SnO(2) nanocontainers for the corrosion protection of 304 stainless steels, J. Colloid Interf. Sci. 579 (2020) 741–753.
[174] Y.J. Ouyang, L.X. Li, Z.H. Xie, L.L. Tang, F.H. Wang, C.J. Zhong, A self-healing coating based on facile pH-responsive nanocontainers for corrosion protection of magnesium alloy, J. Magnes. Alloy. 10 (3) (2022) 836–849.
[175] B. Qian, Z.L. Zheng, M. Michailids, N. Fleck, M. Bilton, Y. Song, G.L. Li, D. Shchukin, Mussel-inspired self-healing coatings based on polydopamine-coated nanocontainers for corrosion protection, ACS Appl. Mater. Interfaces 11 (10) (2019) 10283–10291.
[176] J.B. Xu, Y.Q. Cao, L. Fang, J.M. Hu, A one-step preparation of inhibitor-loaded silica nanocontainers for self-healing coatings, Corros. Sci. 140 (2018) 349–362.
[177] Z.H. Xie, D. Li, Z. Skeete, A. Sharma, C.J. Zhong, Nanocontainer-enhanced self-healing for corrosion-resistant Ni coating on Mg alloy, ACS Appl. Mater. Interfaces 9 (41) (2017) 36247–36260.
[178] C.D. Ding, J.H. Xu, L. Tong, G.C. Gong, W. Jiang, J.J. Fu, Design and fabrication of a novel stimulus-feedback anticorrosion coating featured by rapid self-healing functionality for the protection of magnesium alloy, ACS Appl. Mater. Interfaces 9 (24) (2017) 21034–21047.
[179] F. Fan, C.Y. Zhou, X. Wang, J. Szpunar, Layer-by-Layer assembly of a self- healing anticorrosion coating on magnesium alloys, ACS Appl. Mater. Interfaces 7 (49) (2015) 27271–27278.
[180] J.J. Richardson, M. Bjornmalm, F. Caruso, Multilayer assembly. Technology-driven layer-by-layer assembly of nanofilms, Science 348 (6233) (2015) 2491.
[181] L.Y. Cui, L. Gao, J.C. Zhang, Z. Tang, X.L. Fan, J.C. Liu, D.C. Chen, R.C. Zeng, S. Q. Li, K.Q. Zhi, In vitro corrosion resistance,antibacterial activity and cytocompatibility of a layer-by-layer assembled DNA coating on magnesium alloy, J. Magnes. Alloy. 9 (1) (2021) 266–280.
[182] N.A. Ahmad, P.S. Goh, K.C. Wong, S.C. Mamah, A.F. Ismail, A.K. Zulhairun, Accelerated spraying-assisted layer by layer assembly of polyethyleneimine/ titania nanosheet on thin film composite membrane for reverse osmosis desalination, Desalination 529 (2022), 115645.
[183] H. Xu, W.P. Huang, K.F. Ren, Y.M. Tang, Spraying layer-by-layer assembly of tannin-Fe3+ and polyethyleneimine for antibacterial coating, Colloid Interfac. Sci. 42 (2021), 100422.
[184] D.D. Chen, M.D. Wu, B.C. Li, K.F. Ren, Z.K. Cheng, J. Ji, Y. Li, J.Q. Sun, Layer- by-layer-assembled healable antifouling films, Adv. Mater. 27 (39) (2015) 5882–5888.
[185] Y.B. Zhao, H.P. Liu, C.Y. Li, Y. Chen, S.Q. Li, R.C. Zeng, Z.L. Wang, Corrosion resistance and adhesion strength of a spin-assisted layer-by-layer assembled coating on AZ31 magnesium alloy, Appl. Surf. Sci. 434 (2018) 787–795.
[186] L.Y. Cui, P.H. Qin, X.L. Huang, Z.Z. Yin, R.C. Zeng, S.Q. Li, E.H. Han, Z.L. Wang, Electrodeposition of TiO2 layer-by-layer assembled composite coating and silane treatment on Mg alloy for corrosion resistance, Surf. Coat. Technol. 324 (2017) 560–568.
[187] Y.J. Shi, L. Zhang, J.H. Chen, J. Zhang, F. Yuan, L. Shen, C.X. Chen, J. Pei, Z.H. Li, J.Y. Tan, G.Y. Yuan, In vitro and in vivo degradation of rapamycin-eluting Mg-Nd-Zn-Zr alloy stents in porcine coronary arteries, Mater. Sci. Eng. C 80 (2017) 1–6.