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Surface modification on Zn-based biodegradable metals: recent advances and future perspectives

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Abstract

Zinc (Zn) and its alloys have been extensively investigated for orthopedic implants, cardiovascular stents, and gastrointestinal devices due to their favorable biocompatibility, moderate degradation rate, and ability to overcome the limitations of magnesium and iron-based biodegradable metals. Current research on Zn predominantly focuses on alloy design, mechanical enhancement, degradation regulation, and biocompatibility evaluation. However, the clinical translation of Zn remains constrained by adverse reactions such as cytotoxicity, inflammatory response, and bacterial infection. Surface modification is one of the pivotal strategies to address these limitations. Guided by the limitations of Zn alloy substrates, this review comprehensively summarizes the recent advances in surface modification technologies regarding degradation modulation, biocompatibility enhancement, and multifunctional coatings, and sorts out representative achievements and innovative breakthroughs. Based on this, the review discusses and anticipates the limitations and future challenges of current degradable Zn alloys and their surface modification technologies from multiple perspectives, such as multifunctional coatings, mechanical properties, structural design, and the establishment of a standardized evaluation system. It is expected that through reasonable and innovative surface design, the broader development direction and application prospects of zinc alloys in the biomedical field can be explored. This work will help researchers better understand the current research status, opportunities, and challenges of surface modification of degradable Zn alloys, and provide references for the design of personalized medical Zn devices.

Keywords Zn-based biodegradable metals, Surface modification, Degradation behavior, Biocompatibility, Multifunctional coatings

1 Current research status on biomedical Zn-based biodegradable metals

The rapid evolution of medical technologies and materials science has spurred transformative developments in biomedical metallic materials. Traditional non-degradable medical metallic materials, including stainless steel, nickel-titanium, and cobalt-chromium alloys, remain prevalent in clinical practice for tissue repair and structural support [1–3]. Nevertheless, these materials present intrinsic limitations, including stress-shielding effects, suppressed neo-tissue regeneration, and sometimes a second surgery for extraction [4, 5]. In contrast, biodegradable materials can gradually degrade in the physiological environment through controlled host response, and their degradation products can safely participate in the metabolic process [4, 6, 7]. Moreover, during service, the mechanical load transfer from degrading implants to regenerating tissues occurs in concert with healing, thus obviating the need for removal. Biodegradable metals (BMs), represented by magnesium (Mg)- and iron

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(Fe)-based alloys have garnered significant attention in the application of cardiovascular and orthopedic fields. However, they face challenges pertaining to degradation kinetics modulation and hydrogen gas evolution control [8–10]. The standard electrode potential of Zinc (Zn) (−0.76 V/SCE) is between those of Mg (−2.37 V/SCE) and Fe (−0.44 V/SCE), so the degradation rate of Zn is slower than that of Mg but faster than that of Fe. Biodegradation studies demonstrate that Mg alloys undergo excessively rapid corrosion, whereas Fe-based alloys degrade too slowly. In this respect, the properties of Zn alloys are more in line with clinical demand. Hence, Zn-based alloys have emerged as frontrunners in next-generation biodegradable materials due to their balanced degradation profile and biocompatibility [11–13].

As an indispensable trace element, Zn is associated with the activities of more than 300 enzymes and 1000 transcription factors, in addition to being involved in immune regulation, gene transcription, central nervous system function, metabolic homeostasis, nucleic acid metabolism, and DNA synthesis [14–16]. The adult human body contains 2 ~ 3 g of Zn, with serum and urinary concentrations maintained at 800 ± 200 μg/dL and 109 ~ 130 μg/dL, respectively [17–20]. According to the dietary recommendations for Zn in different countries, the recommended daily intake for adults is 7.4 ~ 25 mg. Zn ions exhibit a wide range of antibacterial properties by eliminating bacteria through effects on bacterial adhesion or interaction with DNA [9, 21]. Furthermore, Zn's osteogenic potential is particularly noteworthy. Zn not only induces activation of multiple osteogenic genes and proteins and stimulates the proliferation, differentiation, and mineralization of osteoblasts to promote bone tissue development and formation, but also inhibits bone resorption and inflammation-related pathologies by suppressing osteoclastic activity [22, 23]. Zn supplementation has been demonstrated to enhance osteogenic differentiation through upregulation of bone marrow-associated genes, including osteopontin, osteocalcin, collagen, and alkaline phosphatase [24–26]. Furthermore, Zn ions may stimulate angiogenesis and produce immunomodulatory effects, collectively contributing to neobone formation. These multifunctional properties have earned Zn the distinction of "twenty-first century calcium" in bone tissue engineering [11, 27].

Epidemiological studies have shown that there is a potential link between low serum Zn levels and an increased risk of cardiovascular diseases (CVDs). Imbalances in Zn homeostasis can significantly promote the development of cardiovascular diseases, such as coronary heart disease, congestive heart failure, ischemic cardiomyopathy, myocardial infarction, sudden cardiac death, and cardiovascular disease mortality [28, 29]. Zn ions are involved in the regulation of intracellular molecular mechanisms of cardiovascular functions, including regulating the activity of key proteins in cardiovascular biology such as nitric oxide synthase, phosphodiesterase, and angiotensin-converting enzyme. In addition, owing to the redox properties of Zn, inadequate Zn intake leads to increased oxidative stress, and interruption of nitric oxide (NO) and nuclear factor kappa-light-chain-enhancer of activated B-cell signaling, contributing considerably to endothelial damage and development of arteriosclerosis [29, 30]. Zn also participates in vascular tone regulation. Intracellular Zn chelation results in selected vascular constriction in rats and human beings and depolarizes vascular smooth muscle membrane potentials [31, 32]. The study on thrombosis shows that the Zn-coated stents have anticoagulant properties [33]. These findings collectively suggest that Zn has promising therapeutic potential for cardiovascular disease management [34, 35].

Therefore, Zn has been widely studied and expected to be applied in biomedical field, such as vascular stents [7, 9], bone screws and plates [36–38], bone scaffolds [39], guided bone regeneration (GBR) membranes [40, 41], anastomotic nails, biliary stents [42], biodegradable batteries [43], etc. In addition, Zn-based nanomaterials such as ZIF-8 have broad application prospects as drug carriers in fields such as drug delivery, tumor therapy, and bioimaging [44–46], as shown in Fig. 1.

Despite these advantages, the clinical translation of Zn-based BMs remains constrained by critical challenges:

(i) Pure Zn exhibits limited strength and ductility and fails to meet the requirements for load-bearing implants. Biodegradable metallic materials for orthopedic implants generally require a yield strength (YS) > 230 MPa, ultimate tensile strength (UTS) > 300 MPa, and elongation rate (EL) > 15%, while cardiovascular stents require sufficient radial support to maintain vascular patency [7, 47–49]. Moreover, Zn alloys possess suboptimal creep resistance, with their mechanical stability further compromised by natural aging phenomena [50].

(ii) In vitro immersion experiments have shown that Zn alloys are prone to localized corrosion during degradation, leading to uneven degradation and premature loss of mechanical integrity, thus affecting the performance and service life of implants [51–53].

(iii) Paradoxically, the therapeutic benefits of Zn are counterbalanced by its cytotoxicity at elevated concentrations. Animal models have revealed that a local Zn ion concentration exceeding 100 μM may give rise to apoptosis and inflammatory reactions [54, 55]. Dambatta et al. [56] have investigated the

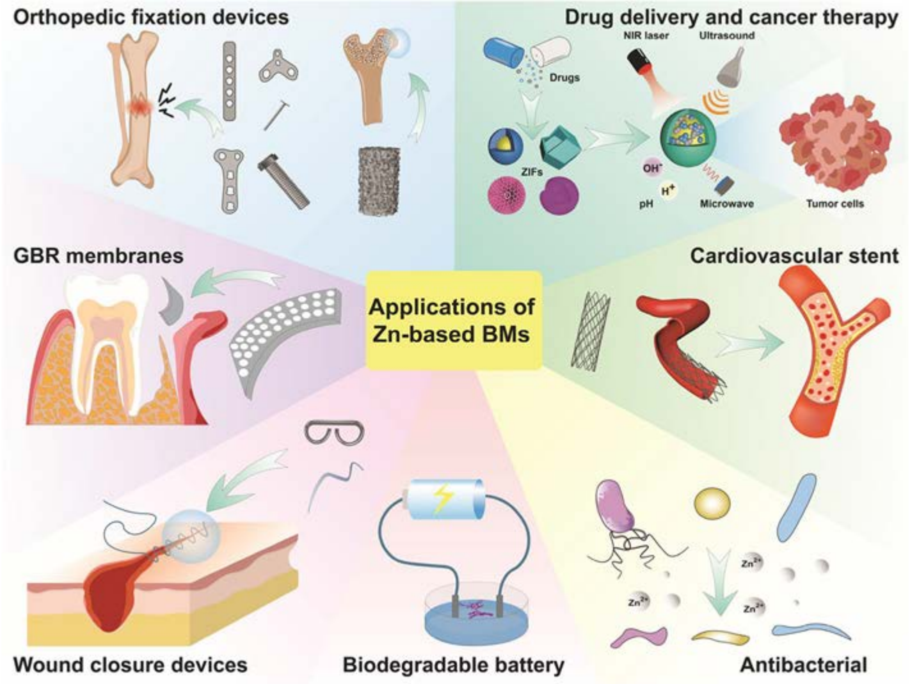

Fig. 1 Medical applications of Zn-based BMs as orthopedic fixation devices, drug delivery and cancer therapy, GBR membranes, cardiovascular stents, wound closure devices, biodegradable batteries, and antibacterial materials

viability of normal human osteoblast cells by culturing in cell culture medium containing extracts of Zn-3 Mg alloy at different concentrations and revealed the cytocompatibility of the alloy extracts at low concentrations (< 0.5 mg/mL), while concentrations of 1 and 2 mg/mL induced significant toxicity. When Zn alloys are implanted into the femoral condyle of rats, poor bone integration is observed due to high local zinc ion concentration [57, 58]. In addition, when the intracellular Zn^{2+} level exceeds the cell's tolerance limit, it can cause damage to intracellular mitochondria and produce a burst of reactive oxygen species (ROS), leading to intense intracellular oxidative stress [59].

To overcome the above challenges and improve the corrosion resistance and biocompatibility of Zn, the following strategies have been proposed:

(i) Alloying and element incorporation: The strategy of adding of alloying elements has been identified as a dual-functional approach to address the mechanical deficiency of Zn and biocompatibility limitation simultaneously [49]. Physically, Zn has a low melting point, can be melted and hot processed in air, has good fluidity and can be easily plastic-processed [60]. Mg, calcium (Ca), strontium (Sr), lithium (Li), silver (Ag), and copper (Cu) have emerged as promising candidates for not only enhancing the tensile strength and elongation but also improving the cytocompatibility, osteogenic differentiation, and bone integration [61–63], as shown in Fig. 2. Furthermore, zinc-germanium (Zn-Ge) alloys exhibit superior hemocompatibility [64]. Table S1 summarizes the relevant research and results on Zn-based alloys in recent years, including mechanical properties, corrosion resistance, and biocompatibility.

(ii) Regulation of alloy microstructure: Strategic microstructural control through mechanical processing and compositional design enables regulation of the degradation kinetics and mechanical properties of Zn alloys, and even play a positive role in the growth and proliferation of cells [92]. As illustrated in Fig. 3, the strength properties of Zn-Li alloys can be significantly improved by grain refinement via extrusion [93]. The grain boundary density can be optimized by high-strain plastic deformation (refining grain structure and eliminating compositional segregation) and second-phase redistribution through thermomechanical processing [94]. These coordinated modification strategies synergistically enhance the mechanical strength while concurrently mitigating localized corrosion susceptibility, consequently improving the strength

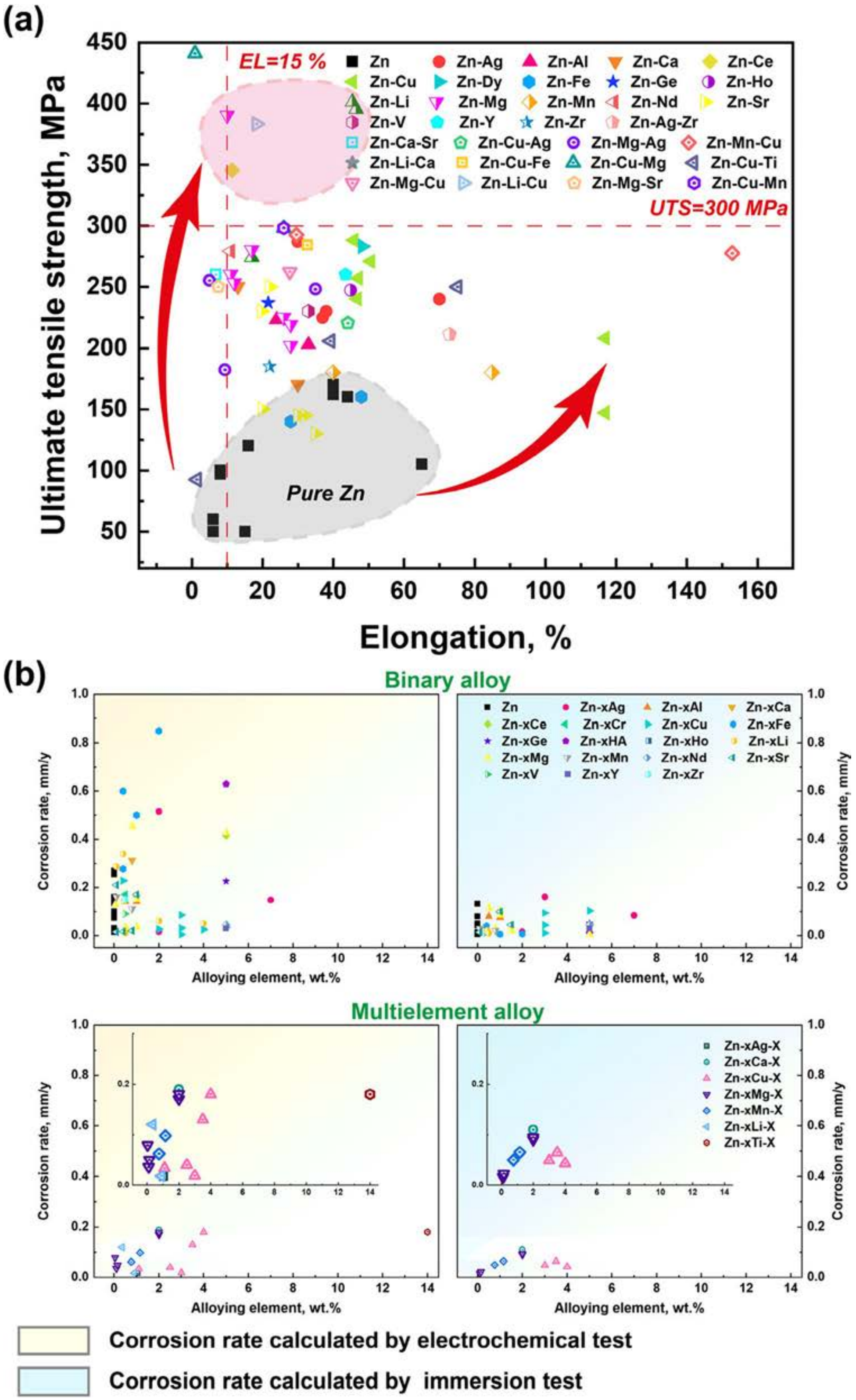

Fig. 2 **a** Summary of ultimate tensile strength and elongation for Zn alloys. **b** Corrosion rate of Zn alloys obtained by different testing methods [15, 26, 36, 38, 40, 58, 61, 62, 64–91]. Table S1 for specific details

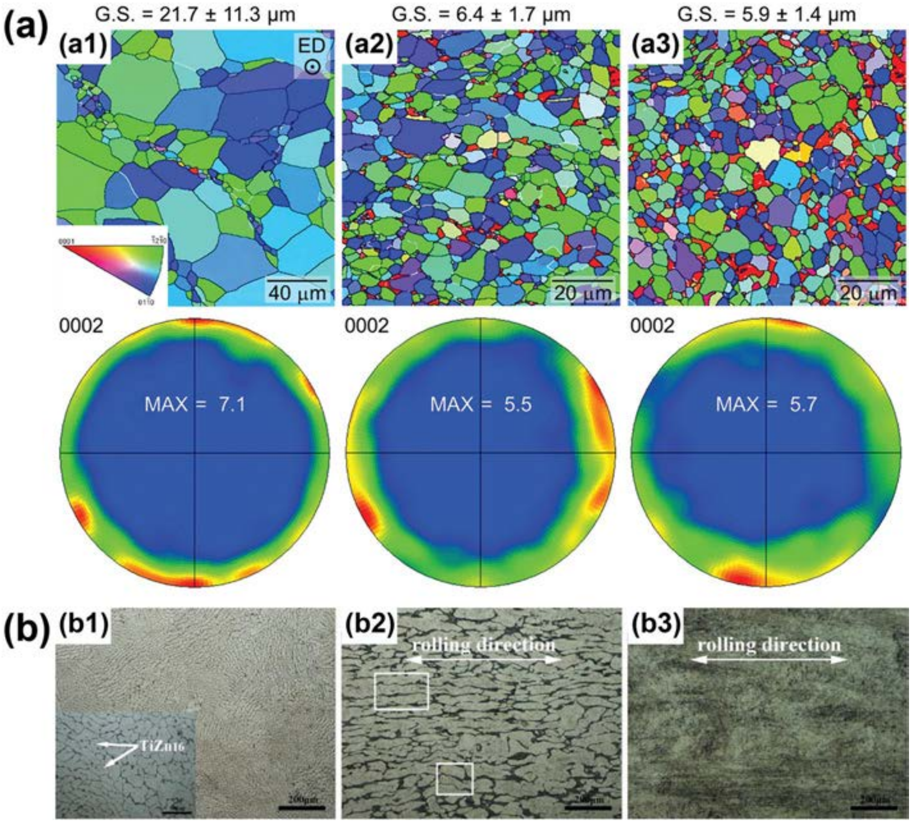

Fig. 3 Adjustment of the microstructure of Zn alloy by alloying and processing: **a** EBSD orientation maps and inverse pole figures perpendicular to ED for the (a1) Zn-0.1Li, (a2) Zn-0.3Li and (a3) Zn-0.4Li alloys. (Reprinted with permission from Ref. [93]. Copyright 2020, Elsevier.). **b** Microstructure of Zn-1Cu-0.1Ti alloy after various plastic deformation processes, (b1) As-cast, (b2) Hot-rolled, and (b3) Hot-rolled plus cold-rolled. (Reprinted with permission from Ref. [94]. Copyright 2020, Elsevier.)

properties of pure Zn and Zn alloys that otherwise do not meet the requirements of biodegradable applications such as vascular stents [95–97]. In addition, laser powder bed fusion is a widely used method for additive manufacturing of Zn-based BMs and is expected to solve the dual challenges of customized structure and biodegradable properties of implants [98–101].

(iii) Surface engineering: While surface modification and coating techniques for industrial Zn alloys (e.g., chemical conversion, anodic oxidation, ion implantation, electropolishing, and composite coatings) have been extensively investigated, their biomedical applications are constrained by the stringent biocompatibility requirements [13, 39, 102–104]. Current research of biomedical Zn alloys focuses not only on degradation rate regulation but also on biocompatibility and multiple functions through advanced surface treatments and coatings [105]. Surface modification methods for biomedical Zn-based degradable materials are diverse. The core principle involves altering the physical structure, chemical composition, crystal structure, or energy state of the material's surface layer, thereby changing or endowing it specific properties. According to the principles of common surface modification techniques, this paper roughly categorizes them into three groups, as illustrated in Fig. 4. The first type is to form a coating on the Zn alloy substrate. Methods such as oxidation, phosphating, or coating application generate a dense, continuous, and strongly adherent layer on the alloy surface, which can isolate corrosive media and enhance surface wear resistance and other properties [106–108]. On the other hand, design biocompatible coatings to enhance or endow the substrate with antibacterial, anti-inflammatory, anticoagulant, osteogenic properties and other properties [109, 110]. The second category focuses on altering surface morphology and roughness through mechanical treatment (such as polishing, sandblasting, etc.), chemical/electrochemical treatment, and other methods. Performance modulation is achieved by controlling surface wettability, increasing surface area, modify-

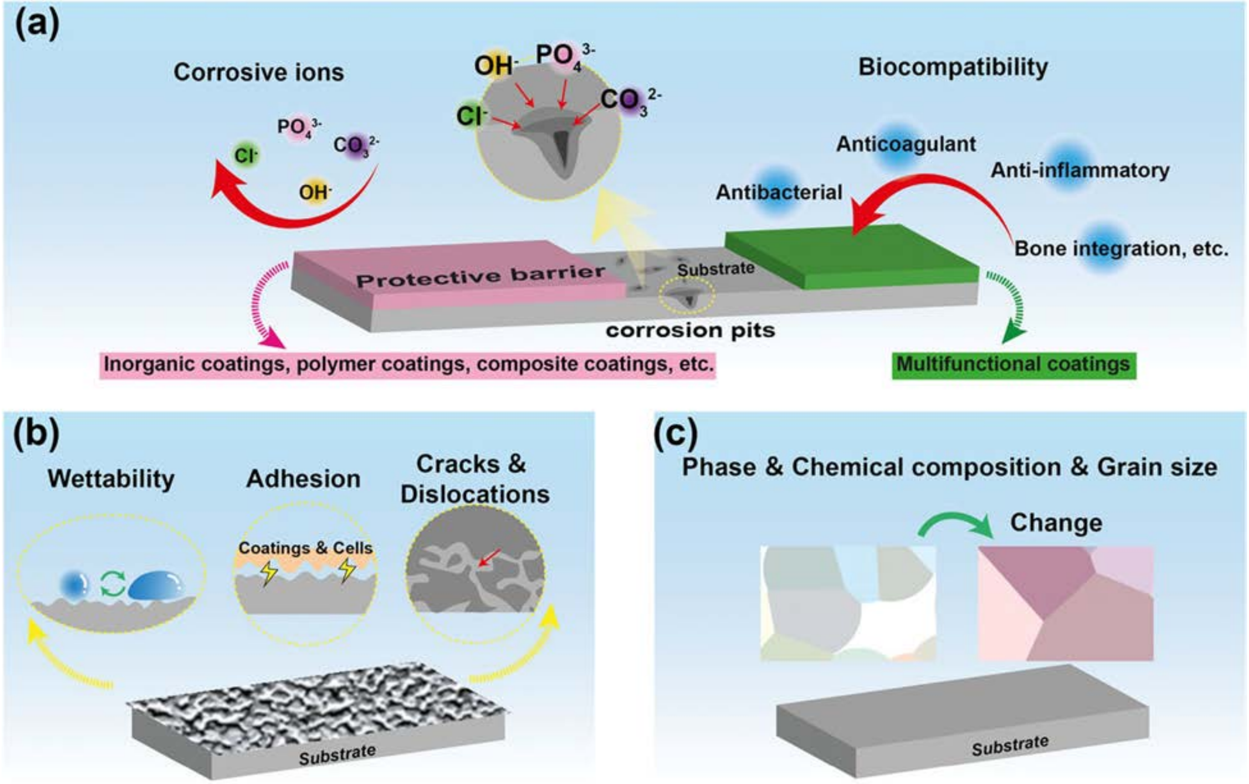

Fig. 4 Schematic diagram of surface modification mechanism: **a** Coating as a physical barrier or biologically active surface. **b** Change the surface morphology and roughness of the substrate. **c** Change the surface microstructure/chemical composition of the substrate

ing surface energy, and adjusting dislocation density. Furthermore, a roughened surface may serve as an anchoring site for subsequent coatings or cell adhesion [111–113], as shown in Fig. 4(b). The third category modifies the surface microstructure, energy sources (such as laser, electron beam, ion beam, heat, etc.) induce surface phenomena such as melting, solidification, phase transformation, grain refinement, solid solution strengthening, dispersion strengthening, or the formation of amorphous/nanocrystalline structures [104, 114]. These microstructural changes significantly alter the surface properties of the substrate. In conclusion, surface modification is a comprehensive process involving the synergistic effect of multiple mechanisms. A single method typically involves the above-mentioned multiple mechanisms. For instance, micro-arc oxidation simultaneously forms a protective coating and modifies the surface morphology and microstructure of the Zn alloy substrate [115]. The working principles of specific surface modification techniques will be elucidated subsequently.

Figure 5(a) displays the keywords co-occurrence network derived from Web of Science Core Collection publications (2015–2024) using the search topic "biodegradable Zn/Zn alloys/Zn-based alloys/absorbable Zn/biomedical Zn". Bibliometric analysis reveals five predominant research domains in Zn-based BMs: compositional design and microstructural engineering, mechanical performance enhancement, antibacterial functionality development, advanced manufacturing technologies, and surface modification strategies. Previous studies have mostly focused on the optimization of the properties of Zn and its alloy matrix. In recent years, research activities on the surface modification of Zn-based degradable materials have increased year by year (Fig. 5(b)). Surface modification appears to be one of the effective methods to overcome the inherent limitations of Zn alloys in biomedical applications, by precisely modulating the degradation kinetics and enhancing the biocompatibility without compromising the intrinsic bulk properties of the materials. In this review, common surface modification strategies and related advances of biomedical Zn-based metal materials are summarized according to functional classification, and the future

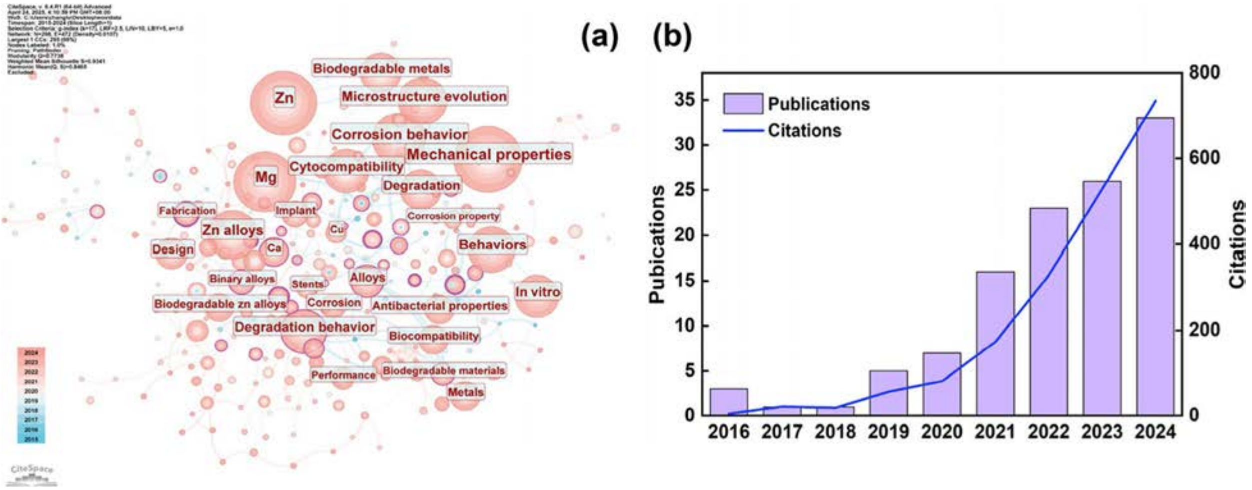

Fig. 5 **a** Keywords co-occurrence network. **b** Publication and citation of literature related to surface modification of biodegradable Zn alloys over time

challenges and directions of surface modification of Zn-based degradable materials are discussed. This objective is to help researchers gain a multidimensional perspective on the current research status in this field and provide theoretical guidance for the design of advanced medical Zn devices.

2 Surface modification of Zn-based BMs for biomedical applications

2.1 Controlling biodegradation property by surface modification

In general, the degradation kinetics of ideal biodegradable metallic implants is expected to synchronize with the progression of tissue regeneration, that is, maintaining the mechanical functionality while degrading. This requires controlling the corrosion rate of Zn-based BMs according to different application scenarios.

In orthopedic applications, clinical requirements specify that bone fixation devices must retain structural integrity for > 6 months post-implantation while achieving complete degradation within 24 months, corresponding to an optimal degradation rate of 0.5 mm/year. However, previous in vivo studies disclose that the degradation rate range of Zn alloys is 0.14 ~ 0.52 mm/year in rat femoral condyle defect models [36, 62]. Notably, this translates into a complete degradation period exceeding 10 years for a standard 4 mm-diameter bone screw, meaning that Zn implants may persist long after bone tissue have healed [114]. The slow degradation process may interfere with the new bone rebuilding process and impair the healing or growth of bone tissue, especially in pediatric patients [116]. In the event of prolonged retention complications, accelerating the rate of corrosion may be beneficial. However, vascular stents need to degrade at a rate of less than 0.02 mm/year, while maintaining mechanical integrity for 3 ~ 6 months and avoiding cytotoxicity to delay reendothelialization and inflammation in vivo caused by excessive release of Zn ions in the early stage of implantation [47, 117]. It is necessary to adjust the degradation rate of Zn-based materials for specific applications. Researchers have used different surface modification methods to regulate the degradation rate of biodegradable Zn, including acid etching, micro arc oxidation, anodizing, chemical conversion, polymer coatings, and so on.

2.1.1 Chemical conversion coatings

Deposition of chemical conversion coatings, a widely utilized surface engineering technique for metallic biomaterials, involves the formation of engineered interfaces by chemical/electrochemical reactions. These conversion coatings modify the corrosion and surface characteristics to enable functionalization while maintaining the bulk attributes. Current advanced coatings for Zn alloys are primarily phosphate-based coatings and oxalate-based.

Ca-P coatings As the main chemical component of bone, calcium-phosphorus (Ca-P) have attracted much attention in Zn-based implants due to their dual functions of corrosion regulation and osteogenic enhancement. In order to improve the biocompatibility and reduce the toxicity of Zn implants, Jablonska et al. [15] have immersed the Zn-1.5 Mg alloy in simulated body fluids for pre-incubation to form a protective layer rich in calcium phosphate, which decreases the initial corrosion rate and improves the cytocompatibility of the alloy.

Li et al. [107] have prepared Ca-P coated Zn-Mg alloy, and electrochemical tests show that the Ca-P coating improves the hydrophilicity and corrosion resistance of the Zn alloy, consequently inhibiting the explosive release of Zn^{2+} in the initial stage of implantation.

Zinc phosphate coatings Inspired by the bone mineral component calcium phosphate, researchers have proposed a coating composed of zinc phosphate (ZnP), which has stable chemical properties and biocompatibility, in order to avoid the potentially adverse effects of calcium phosphate on vascular calcification in vascular stents. It has also been reported that zinc phosphate can promote the growth of hydroxyapatite (HA) and enhance the cell adhesion of fibroblasts on Ti [117, 118]. Su et al. [117] have used a chemical conversion technique using 0.15 M H_3PO_4 and 0.07 M $Zn(NO_3)_2$ solution to fabricate uniform micro-/nano-structured ZnP coatings on pure Zn. The ZnP coatings reduce the release of Zn ions from the substrate during degradation and pH change in the surroundings. Zhang et al. [119] have improved the phosphating process by incorporating graphene oxide (GO) to create dense ZnP coatings with enhanced corrosion resistance. Optimal GO doping (1 g/L) accelerates the phosphating kinetics, refines the morphology of the ZnP coating, and achieves a corrosion rate of 68.5 μm/year for improved corrosion resistance. Mo et al. [106] have synthesized alendronate (AL)-embedded ZnP coatings by alternating immersion in alendronate and acidic phosphate solutions, as shown in Fig. 6(a). The compact, dense, and high-quality ZnP&AL hybrid coating reduces the degradation rate of the Zn substrate and local corrosion and improves the corrosion protection and degradation properties of the Zn substrate. Du et al. [120] have investigated the effects of different reaction times on the chemical conversion film of hopeite (HP, $Zn_3(PO_4)_2 \cdot 4H_2O$) prepared on the microstructure and corrosion behavior of Zn alloys. They found that there was the formation and peeling of the coatings during the preparation process. The microstructure and corrosion resistance of the coating can be optimized by adjusting the processing parameters.

Notably, both Ca-P and Zn-P coatings are able to effectively improve the hydrophilicity of Zn alloys, regulate the release of Zn^{2+}, and reduce the corrosion rate of the Zn alloy substrate [107]. However, compared to the Ca-P coatings, the Zn-P coatings have a better improvement in substrate corrosion resistance. This is attributed to the greater surface roughness of the Ca-P coatings, which increases the contact area with corrosive media. Furthermore, the primary component of the Ca-P coatings consists of soluble CaP phases. Additionally, both coatings effectively promote the activity and proliferation of osteoblasts, this aspect will be discussed in detail subsequently.

Zinc oxalate coatings Carboxylation is an environmentally friendly Zn surface treatment for metallic substrates. Although saturated linear aliphatic monocarboxylic acids have been widely investigated for magnesium alloy corrosion protection, their application to Zn surface engineering is relatively underexplored [125, 126]. As a dicarboxylic acid containing two carboxyl groups, oxalic acid ($C_2H_2O_4$) has unique coordination capabilities with Zn ions. Ferreira Jr et al. [127] have studied the interactions between oxalic acid and Zn substrates through the deposition of zinc oxalate (ZnC_2O_4) coatings. The coatings showed effective protection against corrosion and this is mainly due to the formation of simonkolleite from the base of the pores until their completion. It has been proposed that the corrosion products can form a barrier to protect the substrate from being exposed to chloride solutions.

2.1.2 Electrochemical method

Anodic oxidation As a conventional surface treatment technique, anodization offers operational simplicity and enables rapid formation of stable, uniform, and thickness-controllable oxide layers on metallic substrates [128, 129]. This process is well-established for industrial Zn alloy treatment, and zinc oxide (ZnO) is the primary reaction product [130]. ZnO has good biocompatibility, intrinsic antibacterial properties, and osteogenic potential, enabling accelerated bone tissue regeneration [131, 132]. Hence, anodized Zn alloys have promising potential in biomedical applications. Furthermore, the construction of surface nanostructures by anodization enhances the stability of Zn in biological environments and reduces the susceptibility to non-uniform permeation caused by localized physiological fluctuations.

Dong et al. [133] have fabricated large-scale flower-like ZnO nanotube layers on Zn substrates by anodic oxidation to accelerate corrosion degradation. The Zn/ZnO nanotube composites exhibit better suitability in bone fixation than untreated Zn, which may be related to the enhanced electrochemical activity on its surface. In addition, it also promotes the precipitation of Ca-phosphate on Zn. Zhong et al. [134] have used different anodization durations (1 ~ 20 min) to prepare ZnO nanowire coatings on the Zn-0.4Li alloy. The modified surfaces exhibit enhanced hydrophilicity but reduced corrosion resistance, as the corrosion rates progressively increase from 0.28 mm/year (1 min) to 0.57 mm/year (7 min).

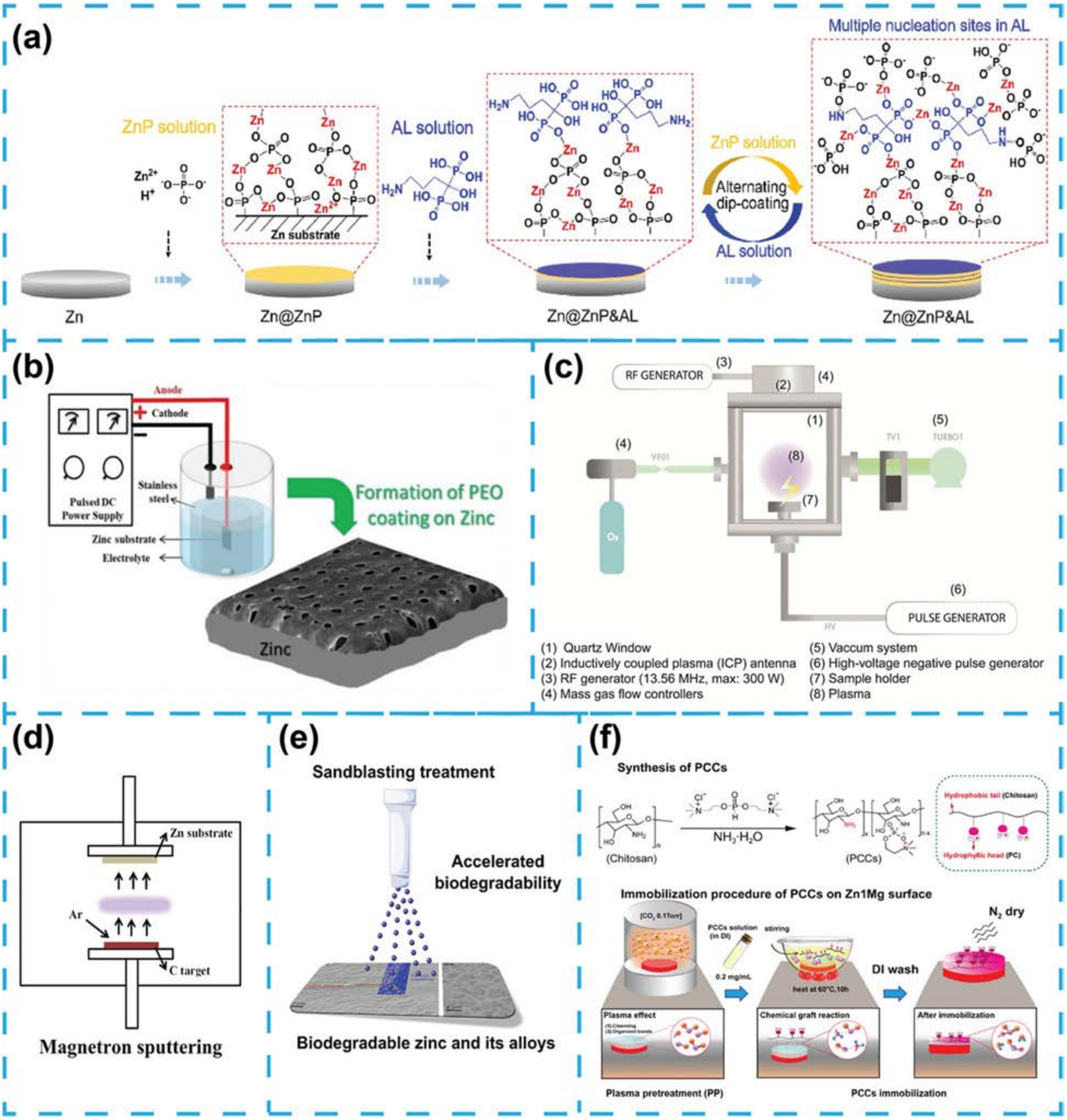

Fig. 6 Schematic diagram of different surface modification methods. **a** Schematic illustration for the construction of AL-mediated bioactive ZnP hybrid coating on the Zn substrate by the alternating dip-coating method (Reprinted with permission from Ref. [106]. Copyright 2021, Elsevier.). **b** Schematic diagram of the *in-situ* PEO process forming a coating embedded with calcium phosphate-based compounds on pure Zn (Reprinted with permission from Ref. [121]. Copyright 2020, Elsevier.). **c** Schematic representation of the plasma immersion ion implantation system (PBII-300) (Reprinted with permission from Ref. [122]. Copyright 2025, Springer Nature.). **d** Schematic diagram of the deposition of diamond-like carbon films on the Zn substrate by magnetron sputtering (Reprinted with permission from Ref. [123]. Copyright 2021, American Chemical Society.). **e** Schematic diagram showing the sandblasting surface treatment of Zn and its alloys. (Reprinted with permission from Ref.[114]. Copyright 2021, Elsevier.). **f** Schematic illustration of the synthesis of PCCs and the immobilization procedure of PCCs on the Zn1Mg surface. (Reprinted with permission from Ref. [124]. Copyright 2020, American Chemical Society.)

Interestingly, extended processing (20 min) decreases the corrosion rate to 0.24 mm/year, indicating a nonlinear relationship between anodization duration and degradation. Guillory II et al. [135] have achieved corrosion inhibition with anodized dense coatings on pure Zn in an oxalate electrolyte. It is attributed to the formation of compact oxalate films as diffusion barriers and subsequent restriction of electrolyte penetration to the substrate. These findings collectively demonstrate that rational surface nanostructuring enables precise control of the Zn degradation kinetics.

Plasma electrolytic oxidation Plasma electrolytic oxidation (PEO) or microarc oxidation (MAO) is an economical, effective, and environmentally friendly surface treatment method for valve metals and their alloys, which are difficult to anodize by conventional anodizing procedures. PEO allows the formation of a thick, uniform, long-lasting, and adhesive coating on metal parts with complex forms [136, 137]. One of the effective techniques for corrosion inhibition of Mg alloys is PEO, which produces coatings with a porous outer layer and a compact inner layer. Mg alloys treated with PEO exhibit excellent biocompatibility and corrosion resistance in simulated bodily fluids (SBF) as the internal dense barrier limits electrolyte penetration [138–141]. In contrast, PEO-treated Zn substrates exhibit paradoxical corrosion acceleration due to the formation of thermal stress-induced cracks caused by the high voltage discharge. These cracks serve as the preferential corrosion pathways to allow the medium to invade and accelerate Zn corrosion through chemical dissolution and galvanic corrosion [136, 142, 143]. Chen et al. [115] have fabricated $ZnO/Zn(OH)_2$ hybrid coatings on pure Zn by PEO and observed that higher applied voltages accelerated long-term degradation of Zn. The electrolyte composition is optimized to produce in situ PEO coatings with enhanced thickness, as shown in Fig. 6(b) [108, 121]. These thick coatings exhibit outstanding corrosion resistance and scratch resistance while maintaining excellent interfacial adhesion without observable delamination.

Electrodeposition Electrodeposition is an effective strategy to fabricate coatings because of its environmental benignity, energy efficiency, and exceptional adaptability for conformal coatings on substrates with complex geometries [144]. Wang et al. [145] have used a polydopamine (PDA)-assisted electrodeposition technique to prepare a strontium-doped HA/polydopamine (SrHA/PDA) composite coating on alkaline-pretreated Zn. This approach achieves controlled Zn^{2+} release from the Zn matrix while preserving its inherent antibacterial properties, while Sr^{2+} ions are simultaneously incorporated to mitigate the cytotoxic effects associated with Zn^{2+} release. Systematic immersion tests demonstrate that the SrHA/PDA composite coating has remarkable corrosion inhibition characteristics and mitigates Zn degradation.

2.1.3 Plasma immersion ion implantation

As an effective and reliable strategy to modify the surface properties of biomaterials, plasma immersion ion implantation enables selective enhancement of surface characteristics and biocompatibility while preserving the bulk attributes [146, 147]. Gambaro et al. [122] have conducted low-energy oxygen implantation (1 ~ 5 kV) on mechanically polished industrial pure Zn with varying durations. The sample implanted at 1 kV for 60 min shows a corrosion rate of 78 ± 26 μm/year, which is significantly less than that of the untreated specimen (135 ± 9 μm/year). This improvement stems from the protection rendered by the denser oxygen-rich surface films formed by implantation.

2.1.4 Acid etching

The degradation of BMs originates from the permeation and interfacial interaction between corrosive physiological media and metallic surfaces. Chemical treatment by strong acid etching (e.g., concentrated H_2SO_4 and HCl solutions) has been employed as a subtractive surface engineering technique for metallic implants for controlled surface morphology modulation via micro-pit/cavity formation with tunable roughness parameters [148–151] as cells tend to attach to rougher surfaces [152]. Xiang et al. [153, 154] have performed acid etching on Zn alloys in $H_2SO_4/HCl/H_2O$ (1:1:1) and nitric acid. The process adjusts the surface microstructures by controlled corrosion patterning and modulates the surface roughness to accelerate degradation. In addition, the acid-etched pure Zn metal has good cell compatibility and antibacterial properties, boding well for applications to orthopedic implants and bioresorbable surgical instruments.

2.1.5 Atomic Layer Deposition

The Atomic Layer Deposition (ALD) is a layer growth process for very thin thickness having self-limiting feature which is done by repetitive exposure of growing thin film with precursors causing successive deposition of layers [155, 156]. This technique has the exceptional capabilities of preserving the substrate topography and accomplishing atomic-level precision in film thickness and compositional control, making it particularly valuable for biomedical surface engineering [157–160]. Yuan et al. [86] have prepared a dual layer on the Zn-0.1Li alloy by ALD. Comprising a ZrO_2 nanofilm and a thicker ZnO layer, this structure decelerates degradation by establishing a robust physical barrier, and corrosion rate modulation is achieved by the precise adjustment of the ALD cycles and parameters. The sustained corrosion protection mechanism gives rise to a stable surface microenvironment that enhances cellular adhesion and proliferation and in vitro cytocompatibility compared to the untreated control.

2.1.6 Sandblasting

Sandblasting is a surface treatment technique for cleaning, smoothing, or etching surfaces by spraying fine particles at a high speed. It has been used in surface treatment and finishing in the industry. Mg/Fe alloys treated with sandblasting exhibit higher degradation rates, while titanium alloys exhibit lower degradation rates. However, studies on Zn alloys have been limited [161–164]. Li et al. [114] have evaluated the effect of sandblasting with two different particle sizes on the degradation of Zn alloys in vitro. Sandblasting accelerates the degradation of Zn, primarily because of the increased surface roughness, elevated defect density, and microcrack formation. However, there is no significant difference in the effect of different particle sizes on the degradation rate of Zn alloys. The in vitro degradation rate after sandblasting is 0.059 ± 0.002 mm/year, which is substantially lower than the clinically required in vivo degradation threshold (0.5 mm/year). In addition, sandblasting increases the degradation rate of Zn and produces adverse effects on cytocompatibility.

2.1.7 Ultrafast pulsed laser surface modification

Ultrafast pulsed laser surface modification is a suitable candidate method to control metal degradation with high precision and good repeatability [165]. This technique enables significant modulation of degradation rates of biodegradable magnesium alloys through the creation of laser-induced micro/nano-structured surfaces [166]. In a systematic investigation [167] comparing nanosecond and femtosecond laser irradiation on biodegradable Zn, distinct surface morphologies are produced with different parameters. Comprehensive characterization reveals accelerated corrosion on the laser-treated specimens compared to the polished control. This phenomenon is mechanistically attributed to the laser-generated structures, which promote stress concentration sites for crack initiation and produce localized corrosion microenvironments, inducing corrosion and accelerating the degradation rate.

2.1.8 Magnetron sputtering

Magnetron sputtering is an advanced physical vapor deposition technique utilizing magnetic field confinement to control ion bombardment of the target materials and enable the ejection and subsequent deposition of target atoms onto substrates to form thin films [168–170]. For example, Peng et al. [123] have employed this method to deposit diamond-like carbon (DLC) films on Zn. The DLC films do not change the surface morphology of Zn but alter the hydrophobic properties. In addition, the corrosion resistance of the DLC-coated Zn decreases unexpectedly, which is possibly due to galvanic corrosion between the DLC film and Zn substrate.

2.1.9 Polymer coatings

Surface modification of Zn alloys by the deposition of polymer coatings has received less research attention than Mg and Fe alloys. Oriňaková et al. [84] have prepared hydrophilic and biocompatible polyethylene glycol (PEG) coatings on Zn-Fe alloy substrates and evaluated the surface morphology, microstructure, in vitro degradation behavior, cytotoxicity, and mechanical properties. The PEG-coated specimens have smoother surfaces than the uncoated counterparts. However, the local pH alteration caused by the dissolution of PEG unexpectedly accelerates corrosion. Notably, the PEG-coated surface-modified Zn substrates show enhanced cellular viability. Shomali et al. [171] have observed improved electrochemical impedance characteristics after poly-L-lactic acid film deposition on Zn wires. A composite coating consisting of the CaHPO4 conversion coating and polylactic acid (PLA) decorated with Li-octacalcium phosphate particles (PLA/Li-OCP) has been prepared on pure Zn [172]. Immersion tests show that the Ca-P coating and PLA/Li-OCP coating reduce the pH. Compared with the Ca-P coating, the PLA/Li-OCP film on the Ca-P-coated samples enhances the corrosion resistance, as shown by a one order of magnitude decrease in the corrosion current density.

Inspired by the cell membrane structure, Sheng et al. [124] have synthesized a biomimetic zwitterionic copolymer, phosphorylcholine chitosan (PCCs), by modifying chitosan with zwitterionic phosphorylcholine groups based on the Atherton-Todd reaction and then depositing it on the Zn-1 Mg alloy. The synthesis and immobilization process of PCCs is shown in Fig. 6(f). This coating enhances the corrosion resistance of the alloy, although the protective efficacy diminishes after 12 h and completely fails in 24 h.

2.2 Improving biological performances by surface modification

As a new generation of biodegradable metallic materials, Zn alloys have large potential in orthopedic fixation and cardiovascular stent applications. However, their clinical translation has been hindered by two critical surface-related limitations: (i) Localized microenvironmental alterations (e.g., elevated pH levels and excessive Zn^{2+} release) during alloy degradation may induce cytotoxic effects or inflammatory responses, consequently compromising biocompatibility [173, 174]. (ii) The inherent bioinert surface fails to provide essential bioactive functionalities such as endothelialization-promoting capacity, antibacterial efficacy, or controlled drug release, thereby failing to meet complex physiological demands [102, 175–178]. Surface modification techniques address these challenges by using different coatings that enhance the substrate bioactivity and confer multifunctionalities, including antimicrobial, anti-inflammatory, antithrombotic, and osteointegration-promoting properties.

2.2.1 Orthopedic applications

In orthopedic applications, traditional biodegradable metallic implants remain confined to passive structural supporting roles due to the limited capacity to orchestrate bone regeneration actively. Inadequate bone mass can complicate the placement of bone implants and may jeopardize the long-term therapeutic effect of the implants due to associated trauma, infection, periodontal disease, and systemic metabolic disorders [179–181].

CaP-based biomaterials have become common synthetic alternatives for autologous bone as their chemical composition is similar to that of the inorganic component in bone tissues [182, 183]. Boasting high biological activity and bone conduction ability, it has been demonstrated to enhance cell proliferation, adhesion, and differentiation [184]. Extensive research has been conducted on Ca-P coatings for Mg alloy substrates [10, 185, 186]. Zhuang et al. [187] have developed a Ca-P coating on porous Zn alloy scaffolds by chemical deposition. This coating regulates Zn^{2+} release and also upregulates osteogenic and angiogenic gene expressions. In vivo studies reveal that the coated alloy scaffold possesses excellent osteoinductivity and osteoconductivity. It also enables bone tissues to grow in the porous scaffolds and osteointegration. Notably, HA is a predominant inorganic component of human bones and teeth [188]. Zhang et al. [109] have performed HA deposition on polydopamine-modified Zn-1 Mg. AS shown in Fig. 7(a), the morphology of the PDA coating loaded with vancomycin after the modification of HA is presented, the Zn-1 Mg scaffold showed a grayish-black appearance, and the typical HA petal-shaped crystals were observed adhering to the surface of PDA microspheres. After implantation into the femur of rats, new bone formation and good bone integration are observed, as indicated by increased osteogenic differentiation and mineralization levels.

Significant advancements have been made in developing calcium phosphate-based osteogenic coatings on Zn alloys, including the Azadirachta indica (neem)-assisted brushite-hydroxyapatite coatings [190] and the nano-hydroxyapatite coatings containing chitosan [191]. The rat calvaria defect model confirms the biosafety and osteogenesis of ZnP-coated Zn alloys in vivo [192]. Alendronate (AL) is a potent bone resorption inhibitor and is used as a drug to treat osteoporosis. An AL-embedded ZnP coating is synthesized on biodegradable Zn to control its corrosion and long-term degradation while promoting the adhesion and proliferation of osteoblasts and inhibiting the excessive growth of osteoclasts [106]. This hybrid coating offers insights into the development of functional Zn-based materials for promoting bone fracture remodeling, especially for osteoporosis, as shown in Fig. 7(c). Qian et al. [189] have introduced three different bioactive molecules, namely cysteine (Cys), phenylalanine (Phe), and bovine serum albumin (BSA), into inorganic ZnP by metal organic/inorganic coordination. Figure 7(d) shows the surface and cross-sectional SEM images of the hybrid coating on Zn surface, when incorporating Phe or BSA with the ZnP the hybrid coatings exhibited distinctly different surface morphologies. In addition, the coatings have a more compact and well-integrated morphology compared with the ZnP coating in Fig. 7(c). The modified Zn exhibits enhanced biocompatibility and significant promotion of proliferation and adhesion of pre-osteoblastic MC3T3-E1 cells and rat bone marrow mesenchymal stem cells (Fig. 8(a)). Furthermore, they have synthesized Zn^{2+} coordinated zoledronic acid and 1-hydroxyethylene-1,1-diphosphonic acid nanorods with a ZnP solution to form a metal organic/inorganic hybrid coating with bone compatibility and bone-promoting properties on Zn [193]. The hybrid coated Zn intramedullary nail promotes bone fracture healing and inhibits bone resorption in the femur fracture OVX rats. Zhao et al. [112] have constructed a calcium zinc phosphate (Ca-Zn-P) coating on Zn alloy implants by a phosphate chemical conversion method, Fig. 7(b) shows the surface topographies of the Ca-Zn-P coating. Systematic evaluations employing both subcutaneous implantation and bone defect models conclusively verify the osteogenic superiority of this bioactive coating.

Beyond conventional calcium phosphate-based coatings, degradable polymeric coatings have emerged as a promising choice for orthopedic implants. Xu et al. [181] have fabricated a composite membrane comprising polycaprolactone and chitosan layers (designated DSZM) on

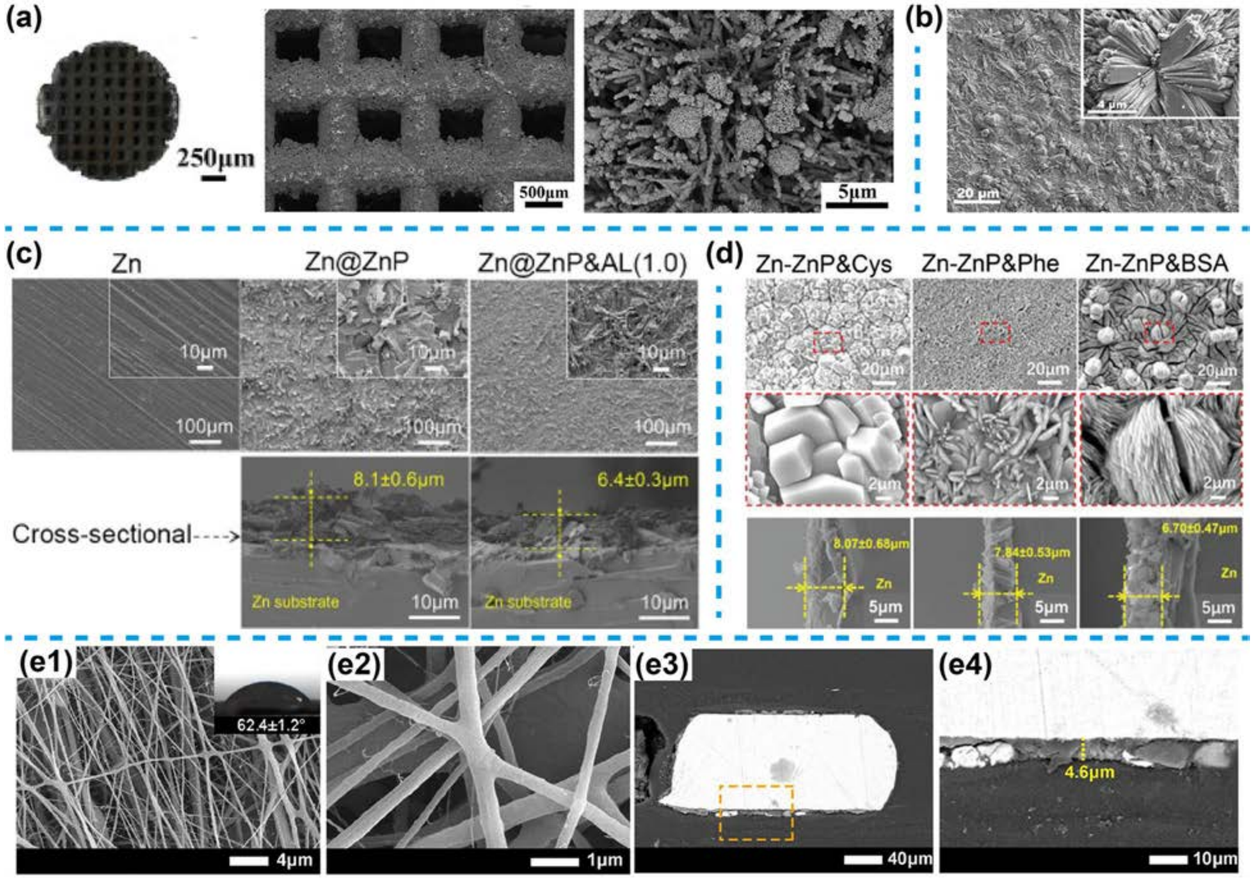

Fig. 7 **a** Morphology of surface of the HA/PDA composite coating loaded with bioactive factor BMP2 and an antibacterial drug vancomycin. (Reprinted with permission from Ref [109]. Copyright 2023, KeAi.). **b** The surface morphology of the calcium-zinc phosphate (Ca-Zn-P) coating prepared on the surface of Zn alloy implants by the PCC method. (Reprinted with permission from Ref [112]. Copyright 2022, Wiley.). **c** Surface SEM images of uncoated Zn, ZnP layer on the Zn (Zn@ZnP), 1.0 mM AL-embedded ZnP coating on Zn samples (Zn@ZnP&AL(1.0)), and cross-sectional SEM images of Zn@ZnP, Zn@ZnP&AL(1.0) coated Zn samples. (Reprinted with permission from Ref [106]. Copyright 2021, Elsevier.). **d** Representative surface and cross-sectional morphology of pure Zn, Zn-ZnP, Zn-ZnP&Cys, Zn-ZnP&Phe, and Zn-ZnP&BSA samples. (Reprinted with permission from Ref [189]. Copyright 2022, Wiley.). (e1-e4) The surface morphology of electrospun polycaprolactone—chitosan nanofibers on zinc mesh: Surface morphology of the double-sided electrospinning of polycaprolactone and chitosan layers (DSZM) on a Zn mesh surface (e1), locally enlarged SEM images of nanofibers (e2), cross-sectional and locally enlarged cross-sectional SEM images (e3, e4). (Reprinted with permission from Ref [181]. Copyright 2024, Elsevier.)

Zn mesh, excepting to achieve better mechanical strength and the capability of guided GBR. DSZM shows both direct and indirect cytocompatibility toward MC3T3-E1 cells. The electrospun polycaprolactone—chitosan nanofiber layer showed an average thickness of 4.6 µm, and the nanofibers were randomly deposited on the surface of the Zn mesh, with a smooth surface, no bead-like structure, and an average diameter of 331.4 ± 63.7 nm. This diameter is conducive to replicating the structure of the extracellular matrix and provides the necessary environmental conditions for cell growth and proliferation. In rat critical-sized maxillary bone defect models, significant enhancement in osteogenic differentiation and calcium deposition is observed, suggesting osteogenic potential for alveolar bone regeneration applications, as shown in Fig. 8(d).

2.2.2 *Antibacterial properties*

It is well known that Zn has antimicrobial properties by multiple mechanisms, including membrane disruption, enzymatic activity inhibition, and reactive oxygen species (ROS) generation [194–197]. Recent investigations reveal that *Escherichia coli* (*E. coli*) can take advantage of low-concentration Zn^{2+} as nutrients to boost growth [198]. This dual function emphasizes that the antibacterial threshold of Zn^{2+} is concentration-dependent [199]. The antimicrobial properties of Zn-based implants can prevent postoperative infections and facilitate wound

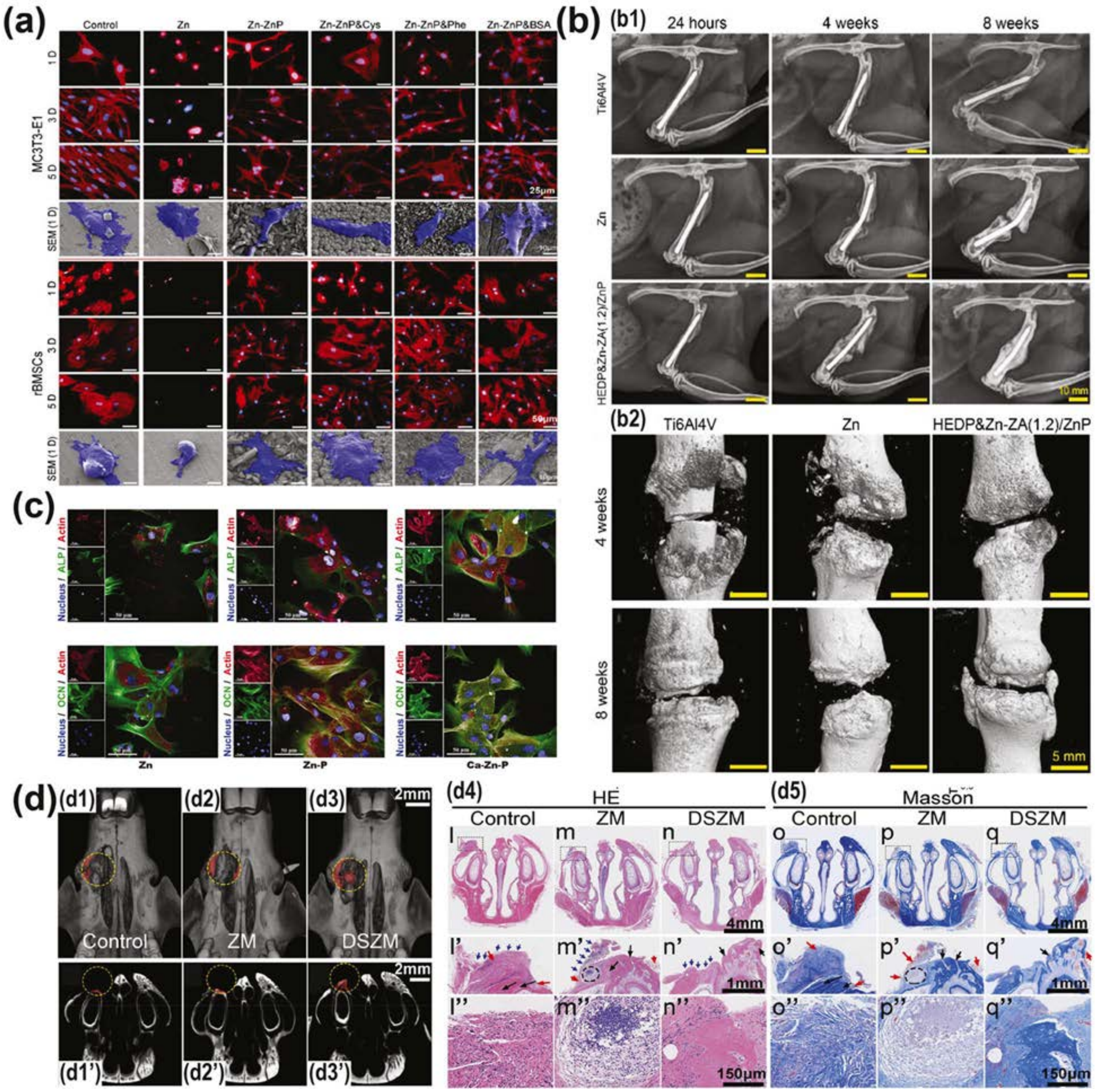

Fig. 8 Representative surface-modified Zn -based alloys in the literature with enhanced bone repair performance. **a** Cell morphology and cytoskeleton of osteoblast MC3T3-E1 cultured on various surfaces for 1, 3, 5 days, and SEM images cells on samples' surface after 1-day culturing (Reprinted with permission from Ref. [189]. Copyright 2022, Wiley.) **b** Multiple surface-treated intramedullary nail implants in rat femoral shaft fracture model: representative radiographs scanning images (b1) and micro-CT 3D reconstruction images (b2) (Reprinted with permission from Ref. [193]. Copyright 2023, Elsevier.). **c** PCC-coated Zn implants influence the osteogenesis of rat BMMSCs, expression of ALP and OCN in rat BMMSCs on implants on day seven or day 14 by immunofluorescence staining (Reprinted with permission from Ref. [112]. Copyright 2022, Wiley.). **d** Osteogenic analysis of ZM and DSZM samples in the rat maxillary defect model: 3D micro-CT images (d1-d3), cross-section micro-CT images (d1'-d3'), and maxillary section staining with H&E (d5) and Masson (d6) (Reprinted with permission from Ref [181]. Copyright 2024, Elsevier.)

healing. Current surface engineering strategies focus on two principal approaches:

(i) Physical or chemical modifications to optimize surface characteristics (hydrophilicity, morphology, and surface charge) for bacterial adhesion resistance. Distinct surface morphologies exhibit differential antibacterial properties. For Zn alloy substrates, a high surface roughness ($R_a \sim 30$ nm) accelerates Zn^{2+} release, which has been observed to inhibit bacterial proliferation and biofilm formation effectively [154, 200]. In contrast, a smoother surface layer characteristic of hopeite coatings has superior antibacterial efficacy by inhibiting the bacterial viability. Generally, increased surface roughness provides more opportunities for bacterial attachment, as surface irregularities offer crevices and niches for bacteria to anchor themselves. Rough surfaces can promote initial bacterial attachment and enhance the formation of biofilm, which can result in persistent colonization and increased resistance to antimicrobial agents. Consequently, reducing surface roughness produces the opposite effect. [117, 200].

(ii) Incorporation of bioactive components (antibiotics, Ag nanoparticles, or quaternary ammonium compounds) using coatings to achieve either sustained release or stimuli-responsive antibacterial activation. Incorporation of antimicrobial metal ions (Ag, Cu, Mn) into calcium phosphate coatings combines biocompatibility and enhanced antibacterial functionality. Rau et al. [113] have developed manganese-strontium co-doped tricalcium phosphate coatings on Zn-Li substrates. Microbial tests demonstrate the inhibition of *Staphylococcus aureus*, *Enterococcus faecalis*, *Pseudomonas aeruginosa*, and *E. coli*. The bactericidal properties of the coating of Ag-incorporated HA nanoparticles on the Zn-1 Mg alloy also produce satisfactory cellular activity and reasonable corrosion and abrasion resistance [201].

The method of loading antibacterial drugs has been extensively studied. Zhang et al. [202] have prepared a new degradable drug-eluting coating of aliphatic polycarbonate on the surface of Zn-Li alloy to reduce the cytotoxicity of Zn alloys, produce antibacterial properties against *Staphylococcus aureus* and *E. coli*, and enhance the osteogenic and angiogenic ability of Zn alloys. Researchers have integrated vancomycin [109, 203], lysozyme [204], and γ-polyglutamic acid [205] into polydopamine to prepare coatings with enhanced antibacterial performance.

2.2.3 Anti-inflammatory properties

Biomedical implants are generally recognized as "foreign bodies" by the host immune system, consequently triggering inflammatory responses. Following surgical implantation, the immune cascade initiates through sequential processes including protein adsorption, cellular infiltration, and adhesion. Macrophages play pivotal roles in this phase and exhibit distinct polarization states: proinflammatory M1 and anti-inflammatory M2 phenotypes. While the transient presence of M1 macrophages facilitates essential inflammatory responses, their prolonged activation gives rise to severe foreign body reactions and chronic inflammation, ultimately compromising tissue regeneration and leading to the clinical failure of biomedical devices [124, 206]. Therefore, creating coatings with anti-inflammatory properties is critical.

Recent studies have demonstrated that regulating the morphological architecture of zinc surface phosphate conversion coatings into micro-nano lamellar crystalline structures facilitates M2 polarization, thereby enhancing anti-inflammatory cytokine production [112]. Qian et al. [207] immobilized a carboxymethyl chitosan/gelatin coating on Zn pretreated with zinc phosphate, followed by aspirin loading to construct a drug-loaded organometallic hydrogel composite coating. This coating protects the Zn substrate from pitting/localized corrosion while demonstrating superior antibacterial efficacy. Furthermore, sustained an acetylsalicylic acid release regulates macrophage phenotypic differentiation toward M2 polarization and upregulates anti-inflammatory factor expression, collectively producing anti-inflammatory effects. Sheng et al. [124] have immobilized phosphorylcholine chitosan, a biomimetic zwitterionic polymer layer, onto plasma-treated Zn-1 Mg surfaces. Although bulk Zn-1 Mg triggered more aggressive macrophage activation, the modified surface and its corrosion products ($Zn_3(PO_4)_2$) preferentially promote macrophage differentiation into M2 phenotypes, which is beneficial for implant applications.

2.2.4 Anticoagulation properties

Given the high mortality rates associated with stent thrombosis, integrating anticoagulant functionality remains a critical prerequisite for the clinical translation of vascular stent coatings [208–210]. While bare Zn alloys have acceptable hemocompatibility in preliminary animal trials, targeted surface engineering strategies are imperative to addressing thrombogenic risks during the acute (0 ~ 24 h post-implantation) and subacute (1 ~ 30 days) phases [211, 212].

Poly (trimethylene carbonate) (PTMC), a biodegradable polymer extensively employed in vascular grafts and tissue engineering scaffolds, and heparin are anticoagulants [213, 214]. Pan et al. [209] have developed a zinc ion-crosslinked PTMC/heparin composite coating on Zn alloy stents by electrophoretic deposition. The coating exhibits enhanced adhesion and proliferation of endothelial cells. Moreover, surface erosion of the composite coating leads to the uniform and long-term release of heparin, which significantly inhibits platelet adhesion and activation and endows the coated Zn alloy stent with long-term anticoagulant functions. Huang et al. [215] have prepared a multifunctional coating system using metal-catechol coordination chemistry, by which copper ions are chelated with PDA. With increasing copper content, platelet adhesion decreases by > 78%. The blood coagulation index shows substantial improvement, with the maximum increase nearly 10%, as shown in Fig. 9(a-b). Fang et al. [110] have prepared a UV PDA/polylactic acid-glycolic acid coating, which establishes a robust binding interface with the Zn substrate for sufficient adhesion strength to maintain the mechanical integrity under corrosive conditions. In addition, the coating inhibits hemolysis and shows smooth muscle cell activity.

2.2.5 *Endothelialization and angiogenesis properties*

NO, a signaling molecule continuously secreted by natural endothelial cells (ECs) in blood vessels, plays crucial physiological roles in maintaining cardiovascular homeostasis, regulating vasodilation, inhibiting smooth muscle cell proliferation, and preventing atherosclerosis [111, 216–218]. Copper ions (Cu^{2+}) have the catalytic capacity to trigger NO release from endogenous S-nitrosothiols in blood, a mechanism that has been strategically employed to create an endothelium-mimetic surface [219–221]. Pan et al. [222] have engineered a biodegradable phenolic-copper–amine crosslinked coating on Zn alloy stents by polycarbonate, tannic acid, and Cu^{2+} co-assembly. The coating exhibits exceptional long-term stability and corrosion resistance, and NO generation enhances endothelial cell adhesion and proliferation rates, suggesting the potential to promote post-implantation re-endothelialization of Zn alloy scaffolds. Chen et al. [223] have fabricated magnesium/copper metal–organic framework (Mg/Cu-MOF) coatings with tunable Cu^{2+} doping concentrations on pure Zn. The coating shows good cytocompatibility with human umbilical vein endothelial cells (HUVECs) and higher expression of angiogenic genes *eNOs* and *VEGF*. The images of the wounding regions in Fig. 9(c) show the angiogenesis capacity.

Collagen, a bioactive extracellular matrix molecule, can interact with α1β1 and α2β1 integrin receptors in ECs to assemble into cords and endows the CL I with angiogenic promotion [224, 225]. Qian et al. [224] have developed an organic–inorganic collagen-entrapped calcium/zinc phosphates coating, which not only regulates Zn^{2+} release within cytocompatible thresholds but also enhances the angiogenic capability, as evidenced by improved HUVECs viability, accelerated migration rates, and enhanced tube formation capacity. Mao et al. [226] have produced a bone-mimetic extracellular matrix-like surface on Zn-1Ca implants consisting of Zn_2SiO_4 nanorods layered with collagen I. The coating enhances the differentiation of HUVECs, upregulates angiogenesis-related gene expression, and vascularizes endothelial cells. Zhang et al. [202] have prepared a multifunctional drug-eluting coating on the Zn-Li alloy by co-assembly of simvastatin (SIM) with an imidazole- and phenylazide-functionalized aliphatic polycarbonate. Owing to the controlled release of Zn^{2+} and SIM, the coating promotes the spontaneous migration and aggregation of EA. hy926 to form a capillary network conducive to angiogenesis. As shown in Fig. 9(e), the SIM-coated Zn alloy shows the richest vascular networks and most intersections of the neo-vessels, indicative of better angiogenesis.

2.2.6 *Against infection and encrustation properties*

The excellent biocompatibility, antibacterial properties, and degradation characteristics of Zn alloys render them suitable for ureteral stents. In this application, their anti-infection and antiencrustation properties reduce the occurrence of crystal aggregation in the presence of excessive urine. The anionic charge of the sulfate group in heparin molecules interacts with Ca^{2+} to inhibit the nucleation, growth, and aggregation processes of crystals [227]. Awonusi et al. [228] have evaluated the potential applications of heparin-coated Zn-Cu alloys to biodegradable ureteral stents. The degradation and biological properties are investigated by immersion tests in artificial urine and in vivo rat bladder implantation models. The heparin-functionalized surface has significant encrustation inhibitory properties and bactericidal activity, and no tissue damage or pathological changes occur during degradation. The modified alloy thus has large potential in anti-encrustation biodegradable ureteral stents.

(See figure on next page.)

Fig. 9 **a** Red blood cell image of hemolysis test with Cu(II)-incorporated PDA coating on Zn (Reprinted with permission from Ref. [215]. Copyright 2025, Elsevier.). **b** SEM images of platelet adhesion on Cu(II)-incorporated PDA coatings (Reprinted with permission from Ref. [215]. Copyright 2025, Elsevier.). **c** Optical pictures showing the effect of different alloy extracts on the cell migration rate of HUVECs (Reprinted with permission from Ref.[223]. Copyright 2024, American Chemical Society.). **d** Optical images of the HUVECs tube formation capability of the different samples (Reprinted with permission from Ref.[224]. Copyright 2022, Elsevier.). **e** A modified chicken chorioallantoic membrane (CAM) method to evaluate the angiogenesis of different samples in vivo (Reprinted with permission from Ref [202]. Copyright 2024, KeAi.)

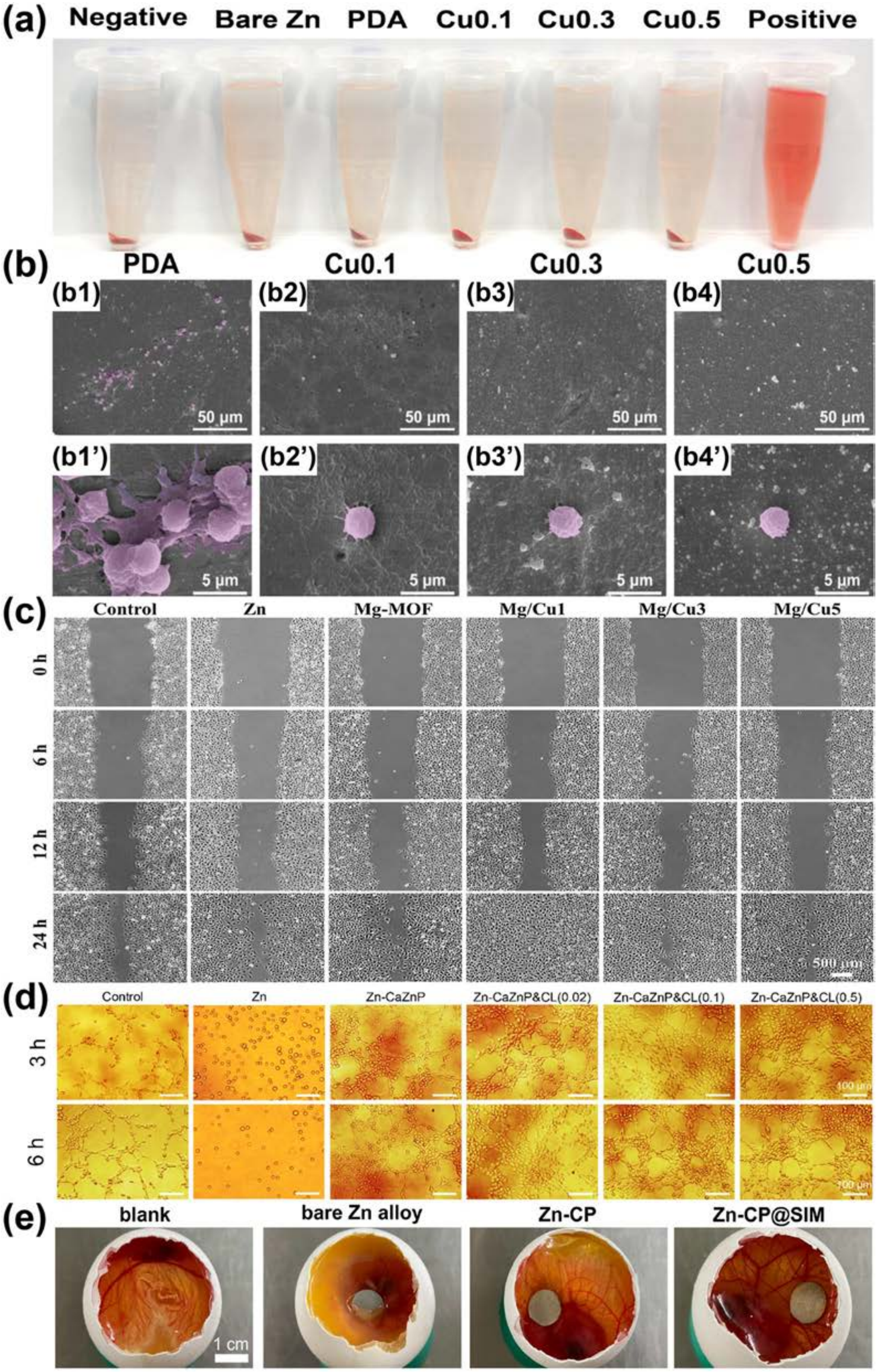

Fig. 9 (See legend on previous page.)

3 Perspectives and challenges of surface-modified Zn-based BMs

Significant progress has been made in addressing the mechanical deficiencies and biocompatibility limitations of pure Zn by means of alloying, processing optimization, and surface modification. However, existing technologies still fail to meet the stringent clinical requirements for implantable devices in various sites. Here, we discuss the challenges and potential solutions of Zn-based biomaterials.

3.1 Mechanical challenges

The mechanical properties of pure Zn are not sufficient. For instance, cardiovascular stents demand a high Young's modulus, low YS (200 ~ 300 MPa), high UTS (UTS > 300 MPa), a large work hardening rate, adequate ductility (EL > 20%), and controllable degradation profiles (12 ~ 24 months) [7, 47, 48]. Orthopedic fixtures require improved strength and ductility. Alloying changes the mechanical/degradation properties of Zn (Sect. 1), and it is necessary to continue to optimize the design to develop Zn alloys that meet the various application requirements. In addition, the undesirable thermal stability of Zn leads to work-softening and aging. With extended storage time, the tensile strength of Zn-Mg alloys increases while the elongation at break decreases [57, 90]. The insufficient mechanical properties may cause uneven deformation or sudden fracture of implants, thus seriously affecting the service life and patients [229, 230].

The aging of Zn alloys stems from unstable mechanical properties, the materials will become brittle after storage at room temperature for a long period of time. Recent research on magnesium alloys has provided valuable insights into overcoming aging-related challenges for biodegradable Zn alloys [231]. Building upon the structural relaxation mechanism observed in Mg-based amorphous systems, Yuan et al. [232] have developed the Zn-Cu binary alloy with tensile YS of 230 MPa, EL of 58%, and a degradation rate of only 10 μm/y. After storing for 20 months at room temperature, the mechanical strength and tensile plasticity are almost unchanged, verifying the excellent aging resistance and excellent plastic deformation ability.

The low melting point of Zn leads to dynamic recrystallization at room temperature and impairs work-hardening capacity. The insufficient work hardening ability of Zn alloys gives rise to uneven deformation and even excessive local deformation and fracture. The strategies to solve this problem include inhibiting recrystallization during the deformation process of Zn alloys at room temperature, promoting the initiation of non-base slip, and controlling the grain size to form a bimodal structure [233–235]. The recrystallization temperature of the materials can be increased by adding trace solutes or second-phase particles to the grain boundaries to prevent the slip and climb of dislocations and the migration of the grain boundaries for the inhibition of nucleation and recrystallization. Yuan et al. have applied the Nabarro-Herring model to predict the trend of thermostability and creep resistance of Zn alloys at ambient temperature and found that solutes such as Cu and Mn improve the creep resistance of Zn alloys, because these elements hinder directional lattice diffusion of Zn atoms [50, 236]. The ideal vascular scaffold should have a bimodal microstructure with a small grain size to improve the strength of the materials and a large grain size to provide the necessary work hardening rate and uniform elongation [237–239]. Using Mg-0.43Ce-0.17Y-0.17Zn-0.12Zr-0.06Ca alloy as an example [240], the bimodal structure increases the yield strength to 348 MPa while maintaining 10.3% plasticity.

Furthermore, surface modification can also influence the mechanical properties of Zn-based materials by altering their surface characteristics, such as structure, morphology, composition, hardness, and microstructure distribution, providing more possibilities for their application. Therefore, future research should not only focus on designing Zn alloys with enhanced aging resistance, tunable degradation kinetics, and clinically relevant mechanical properties, but also investigate the effects of various surface modification techniques on the mechanical performance of these alloys.

3.2 Geometric design and structural optimization

The clinical success of Zn-based implants relies on the coordinated regulation of their macroscopic geometry (shape/structure/size) and microstructure (surface morphology), which collectively determine the key properties of the implants such as mechanical and biological performance. The matrix structure serves as the foundation and carrier of modification technology. Macroscopic/mesoscopic structures (such as porous design) provide the platform for microscopic/nanoscale surface modification and also constitute its constraints. Although their action scales are different, they synergistically affect the overall performance of the implant. For instance, substrate characteristics such as surface area, morphology, and curvature directly govern the feasibility and uniformity of surface modification, the resultant surface properties, and the interactions between the environment and the surface (e.g., cellular responses). Given their interdependence, achieving high-performance, long-life implantable devices necessitates viewing structural optimization and surface modification as an integrated system for co-design and concurrent implementation.

3.2.1 Surface roughness optimization challenges

Rough and porous surfaces enhance biomechanical responses and cellular adhesion compared to smoother ones [241–243]. While a higher surface roughness on Ti alloys generally enhances osseointegration as a result of improved protein adsorption and osteoblast attachment density, excessively high roughness can reduce the mechanical properties of implants [152, 244]. The surface roughness of degradable Zn alloys presents an engineering dilemma compared to titanium alloys [245, 246] as shown in the following

(i) Biosafety limitations: Although existing surface modification strategies (e.g. chemical etching, physical sandblasting) can control the roughness of Zn alloys (Sects. 2.1.4 and 2.1.6 for details) [114, 153, 154], the residual etching medium (e.g. acidic ions) or embedded sandblasting particles (e.g. Al_2O_3) may trigger chronic inflammation or cytotoxicity [247, 248].

(ii) Mechanical-degradation imbalance: The change of surface roughness by mechanical machining inevitably leads to surface plastic deformation and microcracks. The inherent softness of Zn exacerbates surface damage during processing, affects fatigue life, and accelerates the crack propagation rate during degradation.

(iii) Long term stability deficiency: Dynamic surface evolution (such as the formation of ZnO corrosion layer) during degradation undermines initial biofunctionalization.

A multi-scale surface engineering strategy spanning from microscale texture modulation to macroscale design is imperative to circumvent defect generation inherent to conventional machining. Advanced surface patterning techniques must achieve dynamic equilibrium between degradation kinetics and roughness attenuation rates, while meeting biocompatibility requirements.

3.2.2 Porous structure design

Porous materials have been widely studied in orthopedics and other fields because of their excellent features and properties. They can mimic the structure of bone and can provide the space for bone to grow inward to achieve osseointegration. Porous structures with interconnected and suitable pores increase cell adsorption and migration and promote nutrient distribution and osteogenic capacity through newly formed blood vessels [249–251]. Metallic foams have gained increasing attention in biomedical applications due to their unique structural characteristics and functional advantages. They are suitable for bone substitution and dental implants, where tailored porous structures can effectively modulate the mechanical properties to match specific implantation requirements [252–254]. Recent research activities have expanded into biodegradable Zn-based systems, including Zn-derived foams and coatings. Preliminary studies reveal promising properties of Zn-based foams, including appropriate yield strength and good corrosion resistance as well as necessary biological reactions such as biocompatibility and antimicrobial activity [255–257]. However, several challenges in achieving the optimal balance between structural characteristics (pore size distribution, porosity) and functional requirements (degradation kinetics, load-bearing capacity) have hindered the clinical application of Zn-based foams [254, 258, 259].

In addition, additive manufacturing technology is regarded as an ideal approach in the field of medical device manufacturing due to its significant reduction in the difficulty and cost of customized production, especially in biomedical applications that require personalized geometries and biodegradability [100, 260, 261]. Additive manufacturing technology has advanced the precise fabrication of Zn-based scaffolds with complex internal architectures and customized external shapes. Such scaffolds demonstrate strong potential to meet key criteria for ideal bone implants, including bone-matching mechanical properties, interconnected porous structures, controllable degradation rates, and good biocompatibility [262, 263]. Since 2017, laser powder bed fusion technology (L-PBF) has developed rapidly as a leading additive manufacturing technology for Zn-based BMs [99, 101], and expanded applications such as guided bone regeneration membranes, interbody fusion devices, and biliary stents.

Integrating surface modification techniques with the advantages of porous materials not only allows for the regulation of porous architectures but also imparts enhanced multifunctionality to the substrate. While such synergistic strategies have been extensively explored for porous Ti alloys [264–266], their application to BMs Zn-based materials remains limited. Exploring surface modification technologies applicable to complex surfaces and achieving multi-functional and customized implant materials through the synergistic effect of surface modification and porous structures is the future research direction.

3.3 Intelligent coatings with multiple functions

While contemporary surface modification techniques have substantially improved the degradation behavior and biocompatibility of Zn alloys, limitations persist in clinical implementation. For instance, vascular stent implantation inevitably damages arterial tissues, leading to a series of pathological processes, including thrombosis and acute inflammation [267]. In addition, the nanoscale topological heterogeneity of implant surfaces preferentially adsorbs fibrinogen and fibronectin, forms protein coronas, enhances smooth muscle cell migration, and increases the risk of in-stent restenosis [268–270]. Furthermore, there is no permanent blood microbiome in the blood of healthy individuals, and there may be bacterial infection issues after stent implantation [271]. Orthopedic applications are confronted with challenges pertaining to bone integration efficiency and local inflammation.

Inspired by drug-eluting stent technology, loading drugs (rapamycin, paclitaxel, quaternary ammonium salt, dexamethasone, ibuprofen, etc.) onto BMs surfaces is a solution for multifunctional surfaces [272–276]. Zhao et al. have constructed a self-healing polymer coating on the Mg alloy in combination with a stimuli-response drug delivery nanoplatform for promoting cytocompatibility, osteogenesis, angiogenesis, alkaline phosphatase activity, extracellular matrix mineralization, and expression of osteogenesis-related genes [277].

Metal–organic frameworks have emerged as promising candidates for drug-loading applications due to their chemically tunable architectures, multiple active sites, high porosity, and exceptional thermal stability, boding well for anti-corrosion, antibacterial, anticoagulant, and skin repair [278–282]. While significant progress has been made to fabricate MOF coatings on iron, magnesium, and titanium alloys, investigations on Zn substrates remain relatively scarce [283–285]. Nonetheless, Zn is one of the most common cations in MOF design, such as ZIF-8, which has attracted a lot of attention as a drug carrier in tumor treatment and bioimaging. The synthesis of ZIF films on Zn alloys has significant potential in corrosion protection, long storage, and slow release of metal ions to prevent the sudden release of potentially toxic metal ions [46, 286–288]. Yao et al. [289] have fabricated an omniphobic porous hydrogel wound dressing loaded with ZIF-8, which synergistically inhibits bacterial invasion with controlled release of bactericidal, anti-inflammatory, and nontoxic zinc ions. This multifunctional system further accelerates tissue regeneration by stimulating angiogenesis and collagen deposition. Therefore, how to exploit the advantages of MOFs to develop a multi-functional coating with slow release of zinc ions, antibacterial and anti-inflammatory, intelligent response to drug-controlled release, is an attractive research direction.

3.4 Testing and evaluation standards

The establishment of standardized evaluation frameworks for Zn-based biodegradable alloys and associated surface engineering technologies remains a key issue. Although research on biodegradable Zn alloys has been increasing year by year, comparative analysis remains hindered by inconsistent experimental protocols. For example, disparities in in vitro testing parameters (cell lines, immersion medium composition, test durations) and in vivo models (implantation sites, animal species) create substantial challenges in comparability. Different studies have different emphases, making it difficult to form systematic conclusions. Additionally, most of the zinc alloy-related studies have been performed in vitro in simulated body fluid environments. There is the limitation of ignoring the coupled effects of multiple physical fields in physiological environments, such as shear stress from intravascular blood flow and fretting wear in bone implantation areas [290–293]. Last but not least, the research and analysis of the interaction mechanism at the coating-substrate interface and the evolution process of surface coatings are insufficient. All of the above may lead to significant differences between in vitro and in vivo results.

Emerging machine learning algorithms offers a viable solution. As a subfield of artificial intelligence, machine learning combined with the creation of models that can efficiently learn from real data has been advocated to solve complex modeling and optimization problems in fields such as materials science [294] Machine learning can accelerate and improve the development of new coatings, and researchers have experimentally utilized machine learning to predict the effects of different coating parameters like the coating thickness [295]. In addition, the combination of machine learning and in-silico trial can establish the relationship of "in vitro experiment-computational simulation-clinical effect", which can significantly shorten the research and development cycle, reduce the cost and ethical disputes, and accelerate the development of future technologies [296–298].

4 Summary

Biodegradable Zn and Zn alloys are promising new-generation biomedical metallic materials following the footsteps of magnesium- and iron-based materials due to their excellent biocompatibility and controllable degradation properties, especially in the orthopedic and cardiovascular fields. Among the various strategies to optimize the properties of Zn, surface modification is especially attractive, and implementation is increasing due to the various merits, such as selective changes in the surface properties without sacrificing the bulk characteristics of the materials. This review systematically summarizes the research progress of surface modification technologies for biodegradable Zn alloys, focusing on aspects including degradation rate regulation, improvement of biocompatibility, and design of multifunctional coatings. Representative achievements and innovative breakthroughs are summarized, and the mechanisms and differentiated effects of various modification technologies are elucidated. Based on the current research status, the review further summarizes the limitations (such as mechanical properties, structural optimization requirements, etc.) and future development directions of Zn-based biomaterials and their surface modification technologies, including critical issues such as the development of multifunctional coatings and the establishment of standardized evaluation systems. Particular emphasis is placed on discussing the challenges and prospects facing this field. By synthesizing the current research status and core issues of surface modification of degradable Zn alloys, this review aims to provide researchers with a comprehensive understanding of the present situation, opportunities, and challenges, thereby facilitating the advancement of related materials and technologies towards clinical translation and commercialization.

Abbreviations

Ag	Silver
AL	Alendronate
ALD	Atomic layer deposition
AM	Additive manufacturing
BMs	Biodegradable metals
BSA	Bovine serum albumin
Ca	Calcium
Ca-P	Calcium-phosphorus
Cu	Copper
CVDs	Cardiovascular diseases
Cys	Cysteine
DLC	Diamond-like carbon
ECs	Endothelial cells
EL	Elongation rate
Fe	Iron
GBR	Guided bone regeneration
Ge	Germanium
GO	Graphene oxide
HA	Hydroxyapatite
HUVECs	Human umbilical vein endothelial cells
Li	Lithium
MAO	Microarc oxidation
Mg	Magnesium
MOFs	Metal-organic frameworks
NO	Nitric oxide
PCCs	Phosphorylcholine chitosan
PDA	Polydopamine
PEG	Polyethylene glycol
PEO	Plasma electrolytic oxidation
Phe	Phenylalanine
PTMC	Poly (trimethylene carbonate)
ROS	Reactive oxygen species
SBF	Simulated bodily fluids
SEM	Scanning electron microscope
SIM	Simvastatin
Sr	Strontium
UTS	Ultimate tensile strength
YS	Yield strength
Zn	Zinc
ZnO	Zinc oxide
ZnP	Zinc phosphate

Supplementary Information

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Supplementary Material 1.

Authors' contributions

Lu Zhang: Conceptualization, Investigation, Data curation, Software and Writing. Xin Zhang: Investigation. Jianwei Dai: Investigation, Validation, Software. Juyi Yang: Investigation. Zijian Huang: Investigation. Linyuan Han: Conceptualization, Methodology. Cheng Wang: Investigation and Resources. Zhihai Huang: Investigation and Resources. Chao Guo: Investigation and Resources. Jing Bai: Investigation and Resources. Feng Xue: Investigation and Supervision. Paul. K. Chu: Writing – review & editing. Chenglin Chu: Conceptualization, Funding acquisition, Supervision, Project administration, Writing – review & editing.

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Data availability

No datasets were generated or analysed during the current study.

Declarations

Competing interests

The authors declare no competing interests.

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