Materials Today Communications 50 (2026) 114278

Contents lists available at ScienceDirect

Materials Today Communications

journal homepage: www.elsevier.com/locate/mtcomm

Toward optimized cracking and corrosion resistance in Zn-Al-Mg coatings

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ARTICLE INFO

Keywords:
Zn-Al-Mg coatings
Microstructure
Cracking resistance
Corrosion resistance

ABSTRACT

Zn-Al-Mg coatings remain at the forefront of anti-degradation coatings for protecting various steel substrates. Despite extensive literature on the microstructure and properties of these coatings, covering a wide compositional spectrum, engineering an optimized coating with simultaneously superior cracking and corrosion resistance (under harsh environmental conditions) remains a challenge. This perspective article provides innovative insights into the materials physics underlying such an optimized Zn-Al-Mg coating. In particular, this work presents a framework for microstructural design based on phase optimization, quaternary alloying elements, multi-scale defects, crystallographic texture, strengthening mechanisms, and anti-corrosive solutions, among other factors. The collective insights offered here aim to stimulate the development of more sustainable Zn-Al-Mg coatings.

1. Introduction

Zn-Al-Mg coatings exhibit dormant and new possibilities due to the complex and diverse microstructural features appearing because of non-equilibrium solidification conditions within a broad compositional spectrum. These alloys have been shown to deliver significantly higher corrosion resistance compared to classical pure zinc coatings. Many steel applications benefit from the unique qualities of these coatings, such as excellent friction resistance, anti-galling and anti-corrosive properties [1–5]. However, an optimized zinc alloy coating delivering sustainable and durable properties remains a continuing challenge for the academic and industrial sectors [6–10]. The sustainability of these materials is negatively affected by the severe cracking (during plastic deformation) and corrosion degradation, demanding responsible surface engineering approaches [11,12].

About half of the global consumption of zinc is allocated to galvanizing steel for corrosion prevention. The usage of Zn, Al and Mg resources in nature can be reduced by engineering more durable and sustainable coatings [13,14]. Hot-dip galvanized Zn-Al-Mg coated steels often undergo plastic deformation to form a desired geometry of the products. However, severe cracking limits their formability performance. Accordingly, comprehensive and in-depth investigations on the mechanical properties, cracking tendency and formability of Zn-Al-Mg coatings were launched by the author and team [15–21]. Consequently, numerous valuable publications have emerged to evaluate the cracking susceptibility and mechanical performance of the coating family, with different composition ranges. A crack-resistant Zn-Al-Mg coating inhibits the penetration of corrosive media and therefore enhances the corrosion protection by sustaining a physical barrier. Besides, the intrinsic corrosion performance of these coatings has been the subject of numerous studies for further developments that will be discussed. Due to the intricate microstructural features and the variety of possible solidification paths during the hot dip galvanizing process, it is challenging to fabricate a coating with concurrent corrosion and cracking resistance, as the two most critical properties.

Hence, there are two key categories of properties that must be jointly considered in future design of the optimal Zn-Al-Mg coatings, namely category 1: mechanical properties, category 2: corrosion performance. In this perspective letter, the author presents innovative insights into these aspects from the intrinsic nano/microstructural perspective. Finally, this article presents the theoretical foundation for engineering a durable Zn-Al-Mg microstructure with optimized properties using fundamental concepts. It is expected that such an optimized coating design can deliver concurrently improved cracking resistance, formability, and anti-corrosive performance, leading to enhanced sustainability.

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https://doi.org/10.1016/j.mtcomm.2025.114278
Received 23 September 2025; Received in revised form 3 November 2025; Accepted 6 November 2025
Available online 7 November 2025

2. Existing microstructures of the Zn-Al-Mg coatings

Owing to the non-equilibrium solidification routes in hot-dip galvanizing, Zn-Al-Mg alloy coatings exhibit heterogeneous and multi-phase microstructures [22]. Depending on the hot-dip galvanizing process parameters—such as strip entry temperature, bath temperature, cooling rate, etc.—and the compositional spectrum varying Al and Mg elements, one can attain multiple types of microstructures within these coatings [8]. Many research groups have fabricated and studied different microstructural types of Zn-Al-Mg coatings via various characterization techniques [22–27]. Some selected Zn-Al-Mg coating microstructures are depicted in Fig. 1, both in surface and cross-sectional views, which will be discussed. Note that the aim in Sections 2, 3, and 4 is not to provide an exhaustive literature review but to give a brief impression on the status of these coatings relevant to the subject of this short perspective article.

The typical microstructural component in Zn-Al-Mg coatings is the primary zinc (Zn) grains or dendrites which generally appear lighter in scanning electron microscopy (SEM) images as shown in Fig. 1. The Zn grains are usually surrounded by a continuous eutectic region. These eutectic structures are typically classified into two types: binary eutectic (BE) and ternary eutectic (TE). The BE is made of the coarse platelets of Zn and Mg-Zn intermetallic compounds. On the other hand, TE possesses a fine granular structure of Zn, Mg-Zn intermetallic and pure aluminum

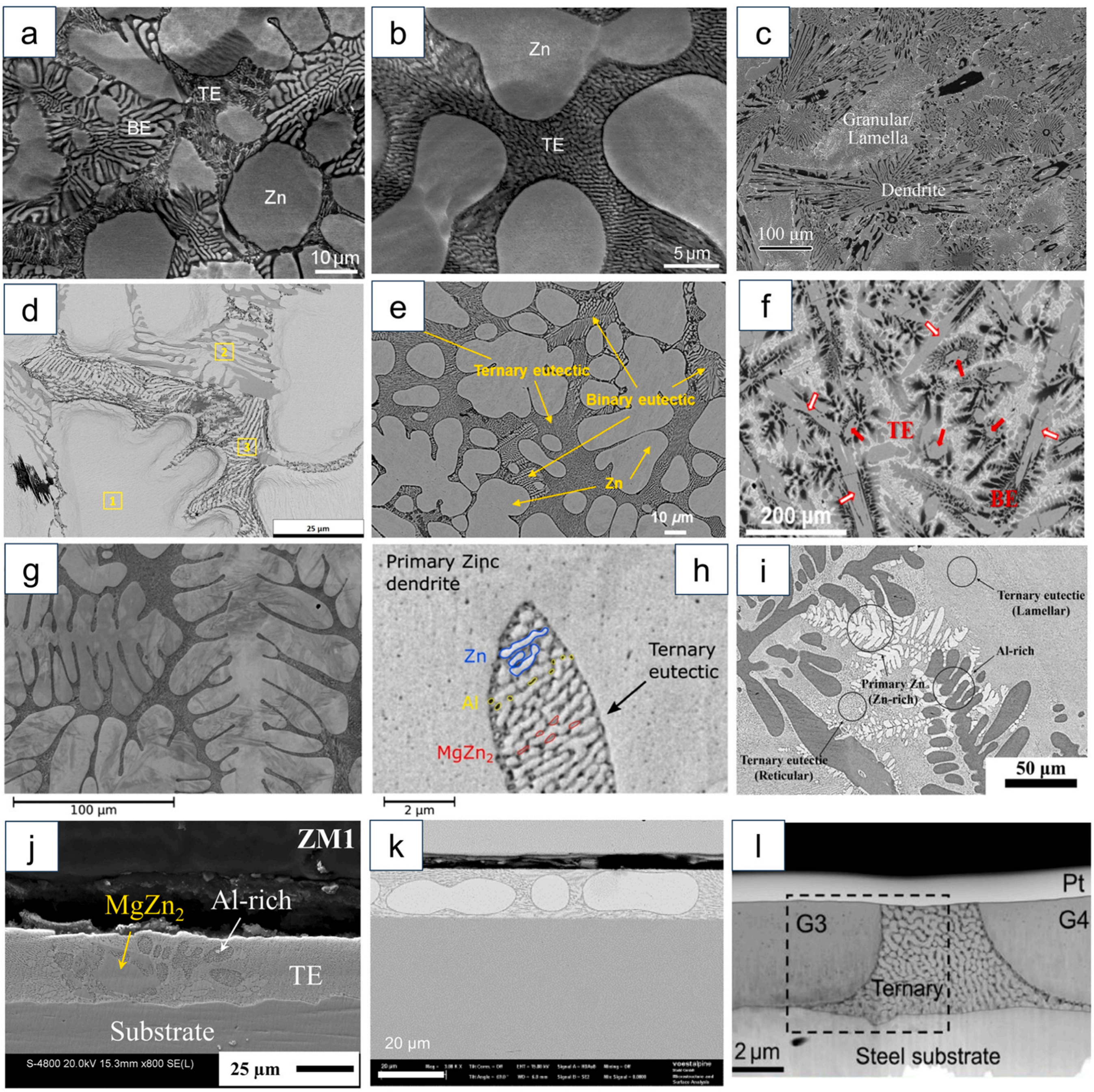

Fig. 1. The representative micrographs of existing Zn-Al-Mg coatings microstructures in the literature. (a-b) Zn-2.9Al-1.9Mg [19], (c) Zn-2.3Al-2.7Mg [28], (d) Zn-1.2Al-1.2Mg [29], (e) Zn-1.6Al-1.0Mg [30], (f) Zn-12Al-5Mg [31], (g-h) Zn-2.5Al-1.5Mg [32], (i) Zn–6Al–3Mg [33], (j) Zn-6Al-3Mg [34], (k) Zn-2.7Al-1.5Mg [35] and (l) Zn-2.9Al-1.9Mg [17]. These coating microstructures are explained within the text. Zn, Al, BE, TE refer to zinc, aluminum, binary eutectic and ternary eutectic components respectively. (Adapted with permission, Elsevier).

(Al). Depending on the applied process parameters and the composition of the coatings, Mg-Zn intermetallic is usually found to be either $MgZn_2$ or Mg_2Zn_{11} compounds which appear darker in SEM imaging using secondary electron mode. Additionally, at higher Al contents, this element can form a distinct phase within the microstructure. For instance, coatings containing approximately 1.7–2 wt% Al and 1.7–2 wt % Mg have shown to deliver Zn, BE and TE as depicted in Fig. 1a. With an increase of Al content to a medium range (2.9 wt%) and 1.9 wt% Mg, the microstructure predominantly exhibits Zn and TE (Fig. 1b) [19]. Within the range of 2.7 wt% Mg- 2.3 wt% Al, a predominant dendritic structure composed of coarse Zn grains and $MgZn_2$ phases, and the granular/lamellar area with a complex ternary eutectic was reported [28] shown in Fig. 1c. Coarse Zn grains with a balanced fraction of BE and TE were attained by low contents of Al and Mg in a Zn-1.2Al-1.2Mg coating [29] depicted in Fig. 1d. In a Zn-1.6Al-1.0Mg coating, similar result of the three typical microstructural components was observed (see Fig. 1e) [30]. At a relatively high content of Al in Zn-12Al-5Mg coatings, a different microstructure was fabricated (see Fig. 1f), comprising of primary Al dendrites, primary $MgZn_2$ grains, a coarse BE of Al-$MgZn_2$, and a fine TE of Al-$MgZn_2$-Zn [31]. As demonstrated in Fig. 1g-h, a microstructure exhibited relatively coarse Zn dendrites with multiple branches, and a TE region in a hot-dip galvanized Zn-Al-Mg coating with 2.5 wt% Al and 1.5 wt% Mg [32]. As shown in Fig. 1i, four microstructural constituents, i.e. primary Zn, Al-rich, BE and TE were characterized in a Zn–6Al–3Mg coated steel [33]. As Fig. 1j-l display, the primary Zn grains typically grow in a columnar manner across the thickness of the coating which is normally in a range of 10–20 μm in the as-produced format [17,34,35]. The addition of a sufficient amount of Al and Mg has been shown to inhibit the formation of Fe-Zn intermetallic layers between the coating and steel substrate, resulting in good adhesion properties [36]. Hence, the coating microstructure can be tuned by modifying Al/Mg contents, adjusting the process parameters such as cooling rate, or by the addition of quaternary alloying elements [37–41]. Nevertheless, there remains a lack of systematic microstructural fine-tuning aimed at optimizing the coating for both high crack resistance and enhanced corrosion resistance.

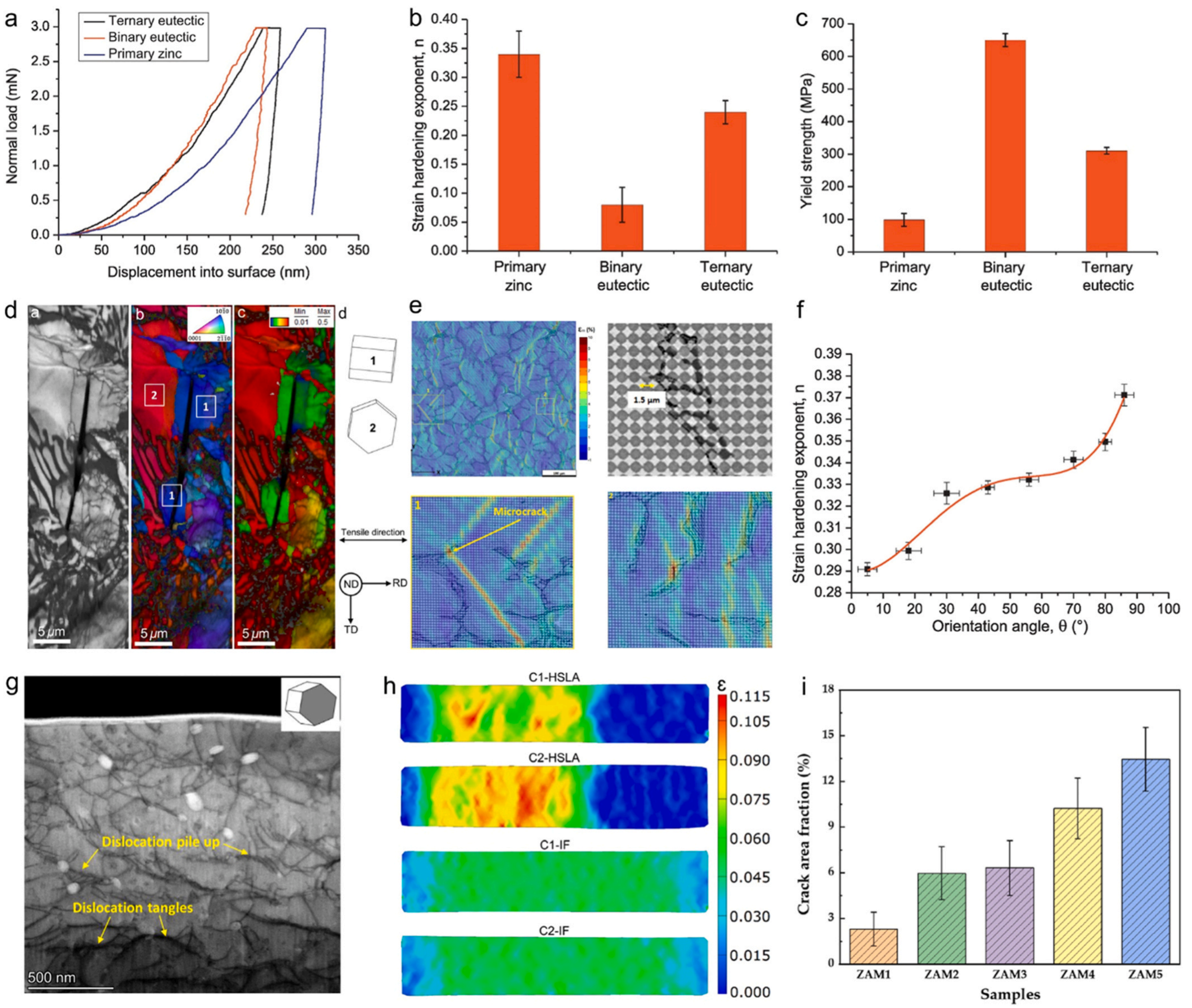

Fig. 2. Multi-aspect mechanical properties of Zn-Al-Mg coatings. (a) nanoindentation curves of the constituent phases [18], (b-c) micromechanical properties of the phases [18], (d) orientation-dependent cracking analyzed by EBSD [18], (e) in-situ DIC analysis on the deformed Zn-Al-Mg coating [29], (f) correlation of the crystal orientation with strain hardening exponent of Zn grains [18], (g) dislocation imaging in the deformed coating, revealing plasticity mechanism [17], (h) macro-DIC on the influence of steel substrate [19], (i) the effect of Mg content on the cracking behavior of the coating [30]. (Adapted with permission, Elsevier).

3. Mechanical properties

Mechanical integrity of the coatings is critical for avoiding white rust formation, and sustaining the anti-degradation performance [42,43]. Recently, numerous studies have emerged on the mechanical properties of Zn-Al-Mg coatings uncovering important aspects, such as hardness [21,44,45], damage evolution [18,29], deformation mechanisms [17], cracking behavior [16,19,20,30,31,34,46–48], formability [16,19], stress-strain localization [18,29], bending deformation [17,46], twinning [49], and liquid metal embrittlement [50–52], among others. Indeed, it is important to elaborate on the anisotropic mechanical properties and cracking behavior, arising due to the various microstructural and deformation modes. The main source of microstructural cracking within these coatings is associated with hard yet brittle intermetallic compounds (i.e. $MgZn_2$) in the binary eutectic component [18]. Nanoindentation results (see Fig. 2a-c) indicate that Zn grains exhibit higher local ductility and plasticity compared to BE and TE. Although BE has the highest local strength due to the abundant $MgZn_2$, TE is found to be more formable and effective for crack suppression [17,18]. Additionally, it was revealed that (see Fig. 2d) some crystal orientations with low Schmid factor (c-axis aligned with tensile direction) are more susceptible to cracking in tensile and bending deformations, utilizing detailed EBSD analysis [18]. One of the first attempts to gain a more crack-resistant and ductile Zn-Al-Mg coating was to reduce the binary eutectic volume fraction within the coatings. This approach resulted in an enhanced plasticity in the coating via activating abundant deformation mechanisms as profoundly studied via correlative microscopy [17]. It was revealed that in Zn-Al-Mg coatings with tailored basal (0001) fiber texture, transgranular crack propagation in primary zinc grains was nearly prevented. Furthermore, simultaneous texture and microstructure control offered a highly crack-resistant and formable Zn-Al-Mg coating during plastic deformation [16]. The damage evolution in tensile deformation was evaluated by in-situ testing and digital image correlation techniques [29] as depicted in Fig. 2e. It was found that twinning creates pronounced damage incidents within the deformed coating, leading to micro-cracking. Zinc crystal orientations were correlated with the local micromechanical properties, namely strain hardening exponent, as a predictive methodology for cracking tendency [18]. Favorable Zn orientations have created large dislocation networks [17], promoting the overall plasticity of the coating as shown in Fig. 2g. In a separate study, it was revealed that the mechanical properties of the steel substrates influence the extent of cracking within the coatings [19]. The substrate with more uniform ductility (i.e. the IF steel) provided a more compatible deformation within the coatings and resulted in a better cracking performance compared to the HSLA substrate exhibiting Lüders banding (discontinuous yielding), as illustrated by DIC maps in Fig. 2h. It has been reported that the increase in Mg content worsens the cracking resistance in these coatings as indicated in Fig. 2i [30]. Another study [31] showed that the grain refinement can lead to poor cracking resistance if $MgZn_2$ phase (as crack initiation site) solidifies into primary phase grains in a coating with 12 wt% Al-5 wt% Mg. However, if the primary Zn grains get refined down to 10 μm, a higher ductility and cracking resistance can be obtained in a coating with 1.7 wt% Al and 1.5 wt% Mg [20]. Whereas the coating with a coarse Zn grain microstructure triggered more transgranular cracking during plastic deformation [20]. Nonetheless, several open questions remain regarding the mechanical performance of these coatings under combined exposure to harsh environmental conditions, such as highly corrosive media and elevated temperatures.

4. Corrosion behavior

Numerous research works have been performed to understand the corrosion properties as a function of internal material factors (such as composition and microstructure) or external environmental conditions and corrosive media [53–57]. Here, the author showcases some selected studies highlighting the significant relevant findings (as this article does not aim at providing an exhaustive literature review). Accordingly, the corrosion protection of Zn-Al-Mg coatings has been identified mainly through the following mechanisms. These coatings form shielding corrosion product films on the surface, including simonkolleite and layered double hydroxides. The resultant films form effective barriers, inhibiting direct connection between the corrosive media and the underlying steel substrate, consequently restricting corrosion to the surface. In addition, the conversion of basic zinc salts into oxides and carbonates is actively avoided by sustaining the pH of the local electrolyte at roughly 10.2, via Mg-based corrosion product [58–61]. Nonetheless, due to the environmental sensitivity of Zn-Al-Mg alloys, the actual corrosion behavior of these coatings remains an active area of research across varying atmospheric conditions. In terms of microstructure (focus of this perspective letter), the eutectic regions, consisting of a lamellar mixture of Zn and $MgZn_2$, exhibit higher corrosion resistance compared to coarse primary Zn grains. Although the $MgZn_2$ phase within the eutectic is more active and dissolves preferentially, its dissolution facilitates the formation of protective Mg-Zn corrosion products (e.g. $Mg(OH)_2$ and Zn–Mg layered double hydroxides), which contribute to the enhanced corrosion resistance of the eutectic microstructure as a whole [36,62,63]. Besides, due to the coarser lamellar $MgZn_2$ structure in BE, this eutectic type outperforms TE in terms of corrosion resistance [64]. Incorporating an additional element such as Si, Ge or Re [65] into the Zn-Al-Mg system shows a promising potential to improve the corrosion properties.

For instance, the corrosion behavior of Zn-Al-Mg coatings was investigated by the electrochemical impedance spectroscopy (EIS) technique, varying the Al and Mg contents in different samples [66]. As Fig. 3a-b illustrate, it was found that the corrosion product formed on a coating with 5 wt% Al and 1.5 wt% Mg has the highest corrosion resistance among all tested specimens, indicating an optimum amount of Mg for corrosion prevention. This is in line with the findings of [67] highlighting that exceeding 3 % Mg content may result in gradual deterioration of corrosion resistance. Regarding Al content, it was reported that Al addition significantly improves the corrosion resistance of these coatings due to the higher phase fractions of α-Al [44]. The addition of Sn into the Zn-3.5Al-1.8Mg system further enhances the corrosion resistance of the coating as depicted in Fig. 3c [44]. In another study [68] it was shown that the individual phases exhibit different corrosion current and potentials, resulting in a combined protection of the Zn-6Al-3Mg coated steel (see Fig. 3d). The mechanism of corrosion resistance (see Fig. 3e) at micro level was demonstrated in Zn-Al-Mg and Zn-Al-Mg-Si alloys [69]. It was found that combined layered structures containing an inner film of ZnAl-LDH and outer layers of Zn-based compounds (e.g. $Zn_5(OH)_8Cl\cdot H_2O$) offer long-term anti-corrosive properties [65]. The intergranular and pitting corrosion were prevented by addition of RE and Si to the Zn-Al-Mg system as reported in [70]. The surface morphologies of the corroded coatings in Si and RE modified coatings are shown in Fig. 3f-h. Another important challenge is the filiform corrosion occurring in Zn-Al-Mg coatings with low Al-content, which needs to be considered for an efficient coating design [71]. Nevertheless, corrosion behavior has not been the subject of thorough investigation, where the coating microstructure has been deformed beforehand. In this context, it is important to know how crack-resistant coating material can also endure a corrosive environment once severely deformed.

5. Theoretical foundation for future microstructural developments

As described in the previous sections, there is a lack of systematic research where concomitant mechanical properties (i.e. ductility, formability and cracking-resistance), and corrosion behavior are improved in Zn-Al-Mg coatings. The direction for obtaining a microstructure delivering concurrent superior cracking and corrosion

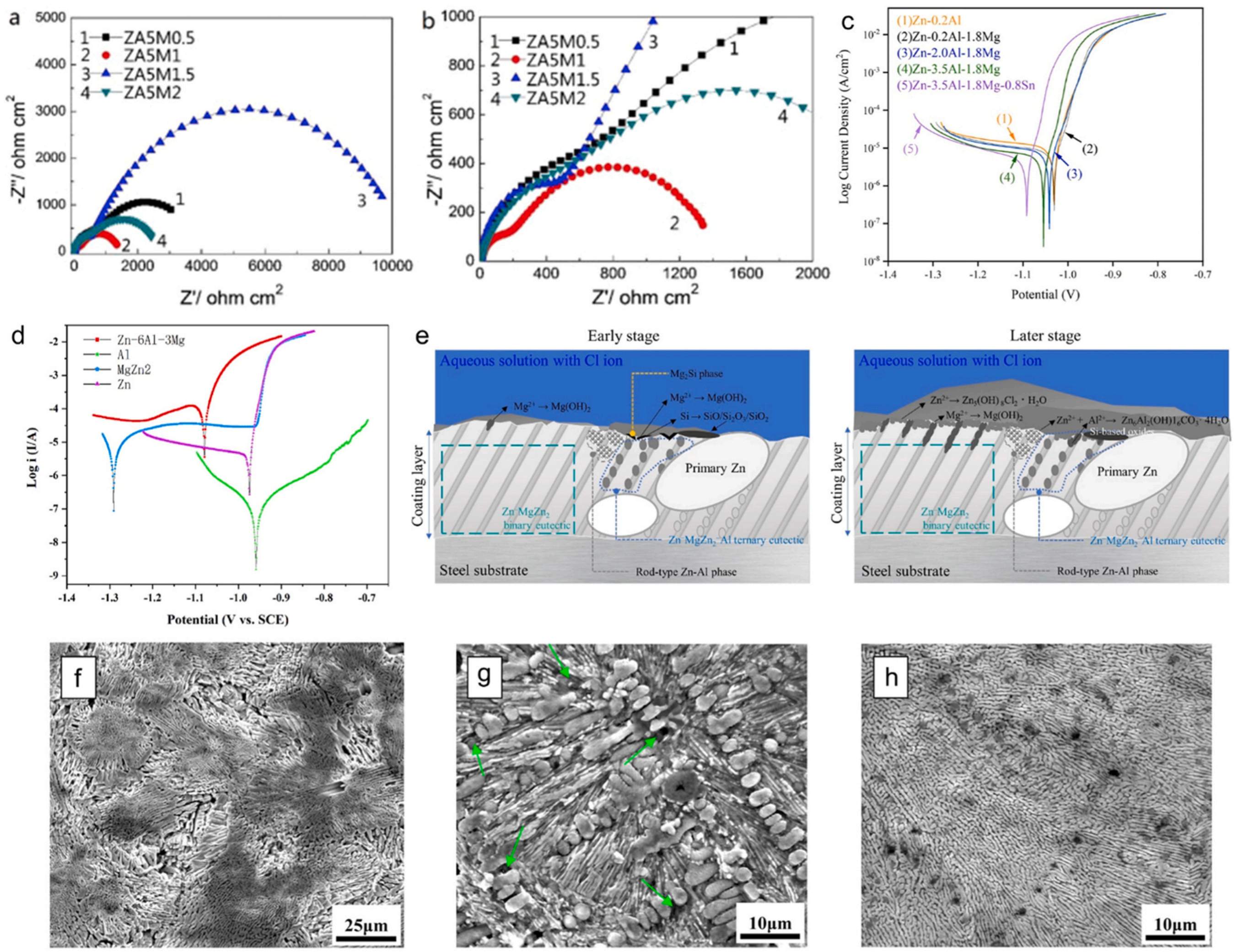

Fig. 3. Some selected corrosion properties and performance of Zn-Al-Mg coatings. (a-b) EIS results on different samples [66], (c) The effects of Sn addition and Al content on the corrosion tendency [44], (d) polarization curves obtained for different phases [68], (e) schematic of the corrosion mechanism and stages [69], (f-h) corroded surface microstructure of Zn-Al-Mg coatings [70]. (Adapted with permission, Elsevier).

resistance remains elusive as an open challenge. Here, the author elaborates on optimized microstructural design strategies aimed at mitigating the drawbacks of existing Zn-Al-Mg coatings and fostering synergistic approaches to achieve this goal. Other possible microstructural designs may exist with varying amounts of Al, Mg, and quaternary alloying elements. Here, the author presents a general guideline applicable to various types of Zn-based coatings. Note that the emphasis is on the collective consideration of the following parameters:

The significant commonality for optimized cracking and corrosion resistance is the refined microstructure. Primary Zn grain refinement down to an average of around 10 μm has been shown to improve the cracking resistance [20]. Such a reduction in grain size has been also correlated with enhanced localized corrosion resistance [63]. However, the grain refinement must be performed in an intelligent and optimized way with effective morphologies and volume fractions of the coexisting phases. Accordingly, it is proposed (see Fig. 4) to eliminate the brittle and detrimental BE, constituting a microstructure comprising Zn grains and TE in a balanced ratio. Although BE helps the anti-corrosive properties of the coating particularly through lamellar $MgZn_2$ phase as discussed earlier, it is quite detrimental in terms of cracking. An alternative way is to benefit from $MgZn_2$ phase in a more ductile form, i.e. in the TE structure to alleviate the trade-off by removing BE. The author has previously shown that nearly an equal volume fraction of Zn and TE can significantly enhance the transgranular cracking resistance and formability of these coatings [17]. As a step forward, the author proposes the dominance of ternary eutectic (~70 % or more) to further mitigate the adverse effect of removing BE on the corrosion resistance. An additional factor is the geometrical shape of the existing phases. Since coarse Zn dendrites exhibit multiple arms, they can trigger high local stress and damage, whereas granular Zn grains can homogeneously deform. A combination of spinodal TE and refined granular Zn grains (instead of coarse dendrites) can deliver synergic ductility-strength improvement within the coating. Because the spinodal nanostructure of the TE provides pronounced grain boundary sliding, whereas the granular Zn grains (preferably < 5 μm) generate noticeable in-grain slip to accommodate the deformation. The proposed Zn grains < 5 μm are well below the thickness size of Zn-Al-Mg coatings (i.e. around 10–20 μm) which can enhance the shear deformation response. As Fig. 4a depicts, the large grain sizes create coarse twins serving as damage and cracking incidents during plastic deformation [20]. Coarse deformation twinning can be largely suppressed by refining the primary Zn grains to approximately 5 μm. Additionally, the presence of the TE component has been shown to inhibit twin formation within the Zn grains [72]. Nano-precipitation can be an additional effective way to inhibit deformation twinning in HCP metals [73]. The targeted ideal future microstructures should also exhibit a good level of active strengthening mechanisms that can contribute to the overall formability of the coating, specifically once exposed to extreme loading conditions.

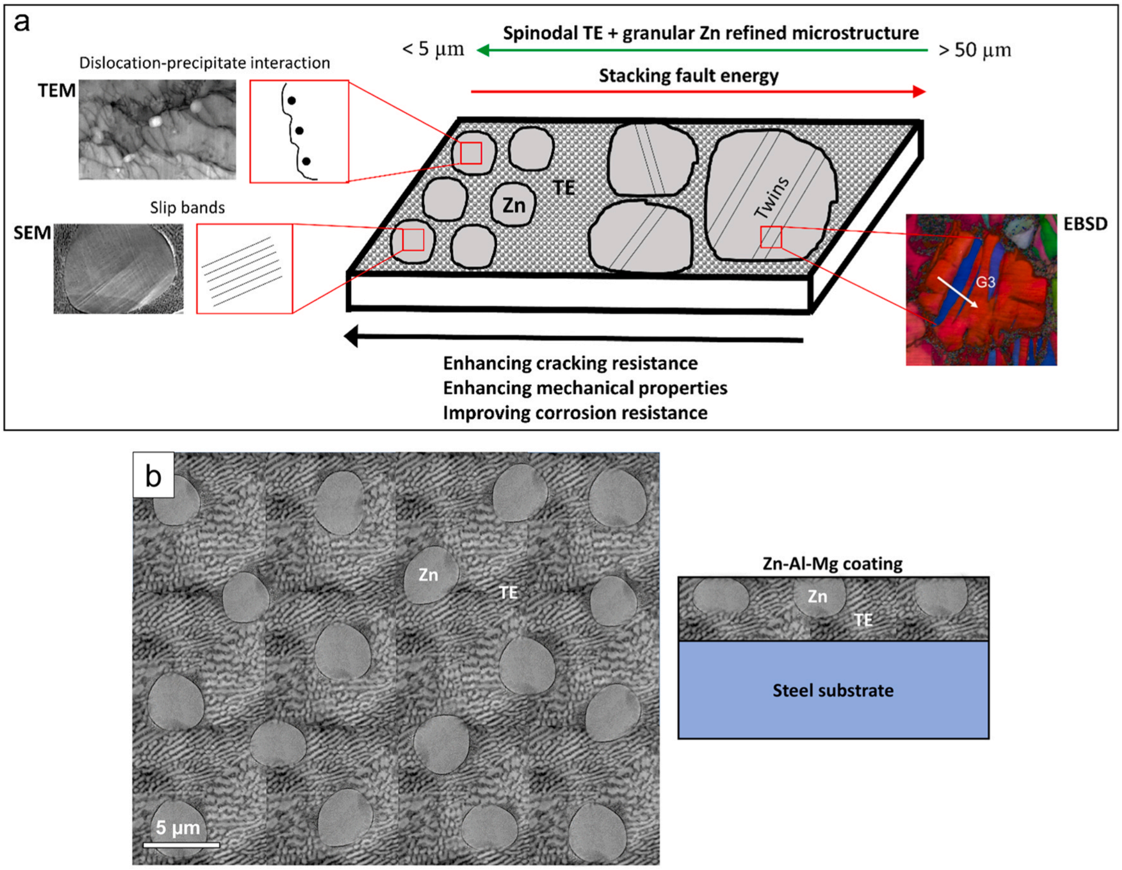

Fig. 4. (a) Schematic illustration of the proposed microstructural design based on the theoretical concepts for durable Zn-Al-Mg coatings with optimized properties, (b) reconstructed surface and cross-section views of the microstructurally engineered Zn-Al-Mg coating targeting optimized properties.

Such strengthening mechanisms have been mainly quantified in our previous study [17] as follows:

$$\sigma_{\varepsilon} = \sigma_0 + \Delta\sigma_{GB} + \Delta\sigma_{dis} + \Delta\sigma_{pr} + \Delta\sigma_{ss} \quad (1)$$

where σ_{ε}, σ_0, $\Delta\sigma_{GB}$, $\Delta\sigma_{dis}$, $\Delta\sigma_{pr}$, $\Delta\sigma_{ss}$ denote the overall flow stress, friction stress, grain boundary strengthening, dislocation (forest) strengthening, precipitation strengthening and solid solution contributions, respectively. Dislocation strengthening can be realized by inhibiting the twin formation and by fostering desirable crystallographic texture. Precipitation strengthening can be nurtured by tuning the solidification parameters during the hot-dip galvanizing process, allowing supersaturation of Mg solute within Zn grains forming $MgZn_2$ or Mg_2Zn_{11} intermetallic, which also potentially can help the localized in-grain corrosion resistance. The design presented in Fig. 4 promotes a significant level of grain boundary strengthening due to the refined microstructure. A reconstructed SEM image of such optimal microstructural design is presented in Fig. 4b. Through a controlled solidification sequence, it is proposed that the ternary eutectic be positioned at the interface between the coating and the steel substrate, thereby preventing single grain contact and enhancing interfacial plasticity.

As Zn alloys exhibit an HCP lattice with a high c/a ratio and large stacking fault energy, competitive and anisotropic deformation mechanisms occur during their deformation [74]. These coatings often should tolerate high local stress due to Lüders banding of the substrate [19]. Lowering the stacking fault energy using a quaternary alloying element such as Ca solutes [75,76] not only can inhibit the formation of twins and enhance the mechanical properties, but also can contribute to improvements in corrosion resistance [76]. The effective quaternary alloying element can be chosen among the following candidates: Re, Ca, Li, Ge, Cu and Bi for improved properties. Additionally, promoting multiple dislocation slip systems [17] by a suitable texture control can trigger an enhanced plasticity. Strong fiber texture along (0001) with appropriate microstructure leads to improved cracking resistance [16], and anti-corrosive properties [77,78]. Nevertheless, such basal fiber texture prompts the formation of abundant deformation twins [29]. This can be mitigated by reducing the Zn grain size well below 5 μm to increase the chance of slip deformation instead of twinning occurrence. An alternative way is to promote the activation of pyramidal or close-packed systems instead of basal systems by crystallographic texture control away from fiber (0001) that can be challenging to obtain during the hot-dip galvanizing process. On the other hand, the latter solution may weaken the corrosion performance of the coatings, as it has been demonstrated that Zn-based coatings with basal fiber texture outperform ones with non-basal texture [77]. Hence, an effective crystallographic texture (both of Zn and TE) requires further in-depth analysis, as its impact has been proven on the metallic materials' corrosion behavior [79].

Considering the corrosion specifically, such durable and highly formable coating design proposed in Fig. 4 can potentially deliver an enhanced corrosion resistance by delaying the degradation mechanisms. Note that the extrinsic effect of corrosive environment is not covered here, as the focus is on the intrinsic microstructural aspect. Firstly, the superior mechanical properties increase the lifetime of the coating. It is expected that such microstructures can resist fatigue and wear well due to the excellent combination of strength and ductility. A crack-resistant Zn-Al-Mg coating inhibits the penetration of corrosive media and therefore enhances the corrosion protection by sustaining a sacrificial barrier. As a result, the coating can provide prolonged sacrificial protection of the steel substrate through its mechanical durability and structural integrity. Secondly, factors such as the presence of optimum amount of Mg, the dominance of TE volume fraction, the refined Zn microstructure, the existence of the spinodal $MgZn_2$ in TE (by the effective suppression of galvanic corrosion), potential quaternary alloying elements such as Ca, Ge, Bi or Re [39,65,69,80], and a tailored crystallographic orientation/texture can promote the localized and intrinsic corrosion resistance of the Zn-Al-Mg coatings—without compromising the concomitant cracking resistance and formability properties. It is worth noting that this microstructure design presupposes the formation of Zn and/or ternary eutectic components as the initial solidification phases, rather than the Al phase, which may lead to different resulting properties.

6. Summary and outlook

Thus, by engineering the microstructural heterogeneities of Zn-Al-Mg coatings through compositional alteration and defect distribution, it is possible to develop crack-resistant, highly formable, and corrosion-resistant coatings simultaneously. Such an optimized coating can deliver a longer lifetime, reduced thickness and enhanced reliability, which in turn contributes to the overall sustainability of the hot-dip galvanized coatings. In light of the theoretical basis offered in this article, a pathway for designing durable and sustainable Zn-Al-Mg coatings on steel substrates is available. The prevailing challenges in the future of this field will not lie in understanding and improving an isolated property, but in unraveling multi-aspect properties. Exciting possibilities for investigating simultaneous mechanical properties and corrosion performance demand further in-depth investigations using both experimental techniques, including in-situ microscopy, novel testing methodologies, and computational approaches such as machine learning. The proposals given in this study can also be instrumental for the technological advancement of sustainable Zn-Al-Mg coatings with a broad range of material compositions in the steel industry.

CRediT authorship contribution statement

Masoud Ahmadi: Writing – review & editing, Writing – original draft, Visualization, Validation, Software, Methodology, Investigation, Funding acquisition, Formal analysis, Data curation, Conceptualization.

Declaration of Competing Interest

The author declares that he has no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgments

This perspective article has been prepared based on the author's PhD thesis previously published at the University of Groningen (https://doi.org/10.33612/diss.202021172). The author gratefully acknowledges Prof. Yutao Pei and Prof. Bart J. Kooi for their mentorship during the period 2018–2022.

Data Availability

Data will be made available on request.

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