



Research Progress on the Corrosion Behavior of Magnesium–Lithium-Based Alloys: A Review

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Abstract

In this review paper, the research progress on corrosion behavior of hexagonal close-packed (HCP) singular phase, body cubic-centered (BCC) singular phase and (HCP + BCC) duplex-structured Mg–Li alloys has been summarized and reviewed, and the future trend about the studies on corrosion behavior of Mg–Li-based alloys and possible solving methods for the improvement in corrosion resistance are discussed also.

Keywords Magnesium–lithium alloys · Crystallographic structure · Corrosion behavior · Corrosion product film · Corrosion mechanism

1 Introduction

Compared with the other metallic structure materials, magnesium–lithium (Mg–Li) alloys are the lightest and their densities vary from 1.25 to 1.65 g/cm³, which is half of aluminum alloys and three-fourth of traditional Mg alloys. Meanwhile, Mg–Li alloys have high specific strength and stiffness, excellent cold and hot deformability, weak mechanical anisotropy and low-temperature properties [1–12], which have received great attentions worldwide for the applications in the new and high-technology fields such as aerospace, aviation, electronics and military [1–5, 8–12]. Thus, it can be predicted that the research level of Mg–Li alloys is very important for the national development in the future. As for the research of Mg–Li alloys, the USA, the former Soviet Union and some European countries started early. Since the 1940s, the USA

had gradually applied Mg–Li alloys to the manufacture of non-structural and substructural components in armored transport vehicles and aerospace fields [8, 13, 14]. The former Soviet Union used MA18 and MA21 magnesium–lithium alloys to make components such as electrical instrument pieces and shells having excellent properties in terms of high specific strength, good ductility and stable microstructure [8]. Although the research work on Mg–Li alloys was carried out late in Japan, remarkable progress had been made in the development of Mg–Li alloys in recent years. By contrast, universities and research institutes in our country such as Chongqing University, Northeastern University, Shanghai Jiao Tong University, Harbin Engineering University, Changchun Institute of Applied and Chemical Research, Metal Research Institute, Chinese Academy of Sciences and other scientific research institutes have also made great progress in the microstructural optimization and performance enhancement of Mg–Li-based alloys [15–26]. However, compared with the developed countries mentioned above, the actual engineering applications of Mg–Li alloys in China are still in its infancy [1–5, 8–12]. Certainly, Mg–Li alloys also have some shortcomings, such as low absolute engineering strength and poor corrosion resistance [1–5], which seriously restricts their engineering applications and further development.

Regarding their low absolute engineering strength, various processing and treatment methods such as alloying, heat treatment and mechanical deformation can be used to

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solve this issue [27–39]. Therefore, the corrosion issue has become a real bottleneck that restricts the development and engineering application of Mg–Li alloys. It is very difficult to improve their corrosion performance by employing traditional processing methods [40–43]. For Mg–Li binary alloys, when Li content in Mg does not exceed 5.7 wt%, the alloy consists entirely of α -Mg phase and retains the hexagonal close-packed (HCP) structure. When Li content is 5.7–11 wt%, the alloy has a duplex structure and consists of α -Mg phase and body-centered cubic (BCC)-structured β -Li phase. When Li content exceeds 11 wt%, the alloy is entirely composed of β -Li phase [37]. Since Li is more active than Mg, the corrosion resistance of Mg–Li alloys should be closely related to the Li content. In general, Mg–Li alloys are very sensitive to the atmosphere, and the corrosive film formed on the surface is loose, making it difficult to protect the underneath matrix [44]. Under the corrosive environment such as aqueous media, the corrosion damage of Mg–Li alloys was very serious and the anodic dissolution of β -Li phases could occur during the corrosion process [45, 46]. Through investigating and comparing the corrosion behaviors of Mg–4Li, Mg–7.5Li and Mg–14Li (in wt%) alloys, Li et al. [7] reported that the sequence of the corrosion resistance for differently structured Mg–Li alloys was BCC-structured Mg–14Li > HCP-structured Mg–4Li > (HCP + BCC) duplex-structured Mg–7.5Li. In order to further understand their corrosion behavior, the research progress of corrosion behavior of three Mg–Li alloys with different crystallographic structures has been reviewed and systematically summarized, and some existing issues in mechanism and possible solutions for improving the corrosion resistance of Mg–Li alloys in the future have been outlooked, with the aim of providing some guidance for the development and application of newly high-performance Mg–Li alloys.

2 Corrosion Behavior of HCP-Structured Single-Phase Mg–Li Alloys

Since the HCP-structured Mg–Li alloys have the same crystal structure as that of traditional Mg alloys, their corrosion behavior has similar characteristics. At present, lots of reports about the corrosion behavior of traditional Mg alloys can be referred [47–62], but there are relatively few literatures on the corrosion behavior of HCP-structured Mg–Li alloys. Meanwhile, researchers mainly used the corrosion mechanism of the traditional Mg alloys to explain that of Mg–Li alloys. Thus, it severely lacks deep understanding about the underneath corrosion mechanism of Mg–Li alloys.

Through investigating the influence of different rolling ratios on the corrosion behavior of Mg–5Li–1Al (LA51)

alloy, Xiang et al. [63] reported that the intensity of crystallographic texture and the density of activated twins increased with the increase in rolling ratios, which finally improved the corrosion resistance of the alloy. This phenomenon is similar to that of the traditional wrought Mg alloys with strong texture, and the main mechanism is ascribed to the weakened corrosion couple between grains with same crystallographic orientations [56, 62]. Through comparing the corrosion behavior of Mg_{94-x}Zn₂Y₄Li_x (at.%, $x = 0, 1, 5, 9, 13$) alloys, Zong et al. reported that the Mg₈₉Zn₂Y₄Li₅ alloy had the lowest corrosion rate, which was mainly due to the formation of long period stacked ordered (LPSO) phases and their barrier effect for the development of corrosion [64]. Zeng et al. [65] reported that the pitting of Mg–1.21Li–1.12Ca–1Y alloy predominately occurred in the α -Mg matrix around the big Mg₂Ca phase particles and gradually developed into filament corrosion, whereas the microstructural refinement (grain size and size of phase particles) by extrusion deformation delayed the pitting initiation and enhanced the corrosion resistance of the alloy. Zhou et al. [66] reported that the formation of extra secondary phases due to the addition of Al and rare earth elements to Mg–3.5Li-based alloys could reduce its corrosion resistance. In the investigation about the effect of hot rolling on the bio-corrosion behavior of Mg–4Li(–1Ca) alloys, Nene et al. [67] disclosed that the refined microstructure significantly improved their corrosion resistance. Based on the description mentioned above, it can be seen that for the HCP-structured Mg–Li alloys, the sensitivity of their corrosion behavior to the microstructure (such as texture, existing phase particles and grain structure) is basically the same to that of traditional Mg alloys. In previous work, it was interesting to be found that for the LA51 alloy, Li₂CO₃/LiOH protective film could be formed on the surface when the rolling ratios were 15% and 20% [63]. Since the surface film of HCP-structured Mg–Li alloys could be composed of different constituents, their corrosion behavior should be different from that of traditional Mg alloys. However, the main mechanism for the formation of Li₂CO₃/LiOH protective film on the surface of HCP-structured Mg–Li alloy with a special rolling ratio is still unknown.

3 Corrosion Behavior of (HCP + BCC) Duplex-Structured Mg–Li Alloys

For the duplex-structured Mg–Li alloys, there exists a potential difference between α -Mg and β -Li phases, which will lead to the formation of α -Mg/ β -Li corrosion couple in the corrosive solution and therefore accelerate the corrosion of alloys. Dobkowska et al. [68] reported that the corrosion behavior of Mg–Li alloys was closely related to

the content of lithium. Compared with the HCP-structured Mg–5Li and BCC-structured Mg–11Li alloys, the corrosion resistance of duplex-structured Mg–7.5Li (in wt%) alloy was obviously lower [68]. Song et al. [69] reported that the pitting of Mg–8Li alloy tended to start at α -Mg/ β -Li interfaces and then preferentially propagated into the interior of β -Li phase to form a typical filament corrosion. Recently, Xu et al. [70] reported the in situ formation of quasi-crystalline phase (I-phase) was very effective for improving the corrosion resistance of Mg–Li-based alloys. The main mechanism was that the continuously distributed I-phase eutectic pockets could serve as physical barriers to the corrosion attack and then significantly improved the corrosion resistance of the duplex-structured Mg–6Li–6Zn–1.2Y (in wt%) alloy [70]. Figure 1 shows the typical microstructure of as-cast Mg–6Li and Mg–6Li–6Zn–1.2Y alloys [70].

The corrosion morphologies of two alloys are shown in Fig. 2 [70]. Since the corrosion resistance of I-phase is very superior [71], the formed I-phase pockets acted as the effective barriers for suppressing the growth of pits and the occurrence of filiform corrosion from the pits. Thus, the corrosion attack occurred in the I-phase-containing Mg–6Li–6Zn–1.2Y alloy was homogeneous and no severe pits

and filiform corrosion were observed, whereas severely localized pits and filiform corrosion occurred on the surfaces of Mg–6Li alloy [70].

Besides the formed I-phase, the addition of rare earth elements (RE) can also have the beneficial effect on the improved corrosion resistance of Mg–Li-based alloys [72–74]. Gu et al. [72] reported that the addition of Nd in Mg–8Li–3Al–2Zn alloy could purify the matrix, induce the formation Nd₂O₃-rich protective film on surface, optimize the structure of film layer and finally improve the corrosion resistance of the alloy. However, the addition of Nd cannot change the corrosion mechanism of Mg–8Li–3Al–2Zn alloy and the corrosion attack still preferentially occurred at α -Mg/ β -Li interfaces [72]. In the investigation about the effect of element Y on the corrosion resistance of duplex-structured Mg–8Li–3Al–2Zn alloy, Gu et al. [73] reported that the potential of α -Mg phases was higher than that of β -Li phases, but the precipitated Al₂Y particles at grain boundaries weakened the corrosion attack at α -Mg/ β -Li interfaces. Therefore, the addition of element Y can improve the corrosion resistance of Mg–8Li–3Al–2Zn alloy. Similarly, Lv et al. [74] reported that the corrosion resistance of Mg–8Li–1Y alloy was higher than that of Mg–8Li–0.5Y alloy, while the improved corrosion

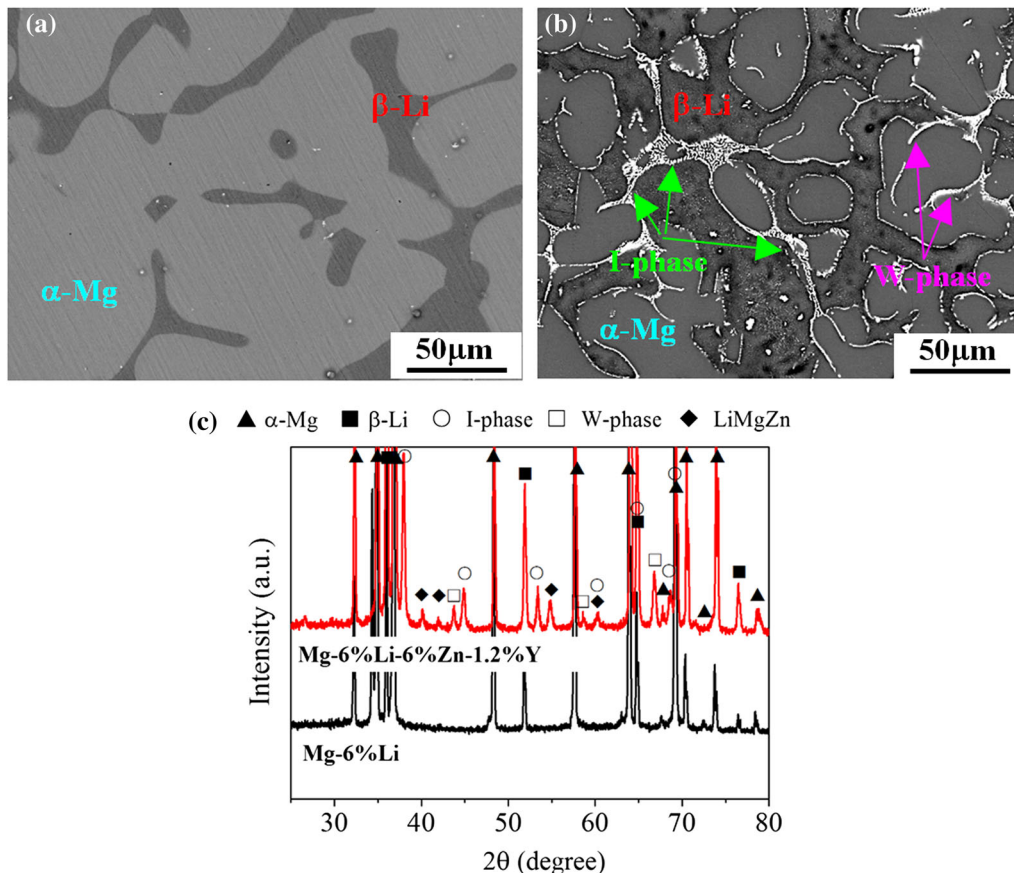


Fig. 1 Microstructures of the as-cast alloys: **a** Mg–6Li, **b** Mg–6Li–6Zn–1.2Y, **c** X-ray diffraction patterns of the two alloys [70]

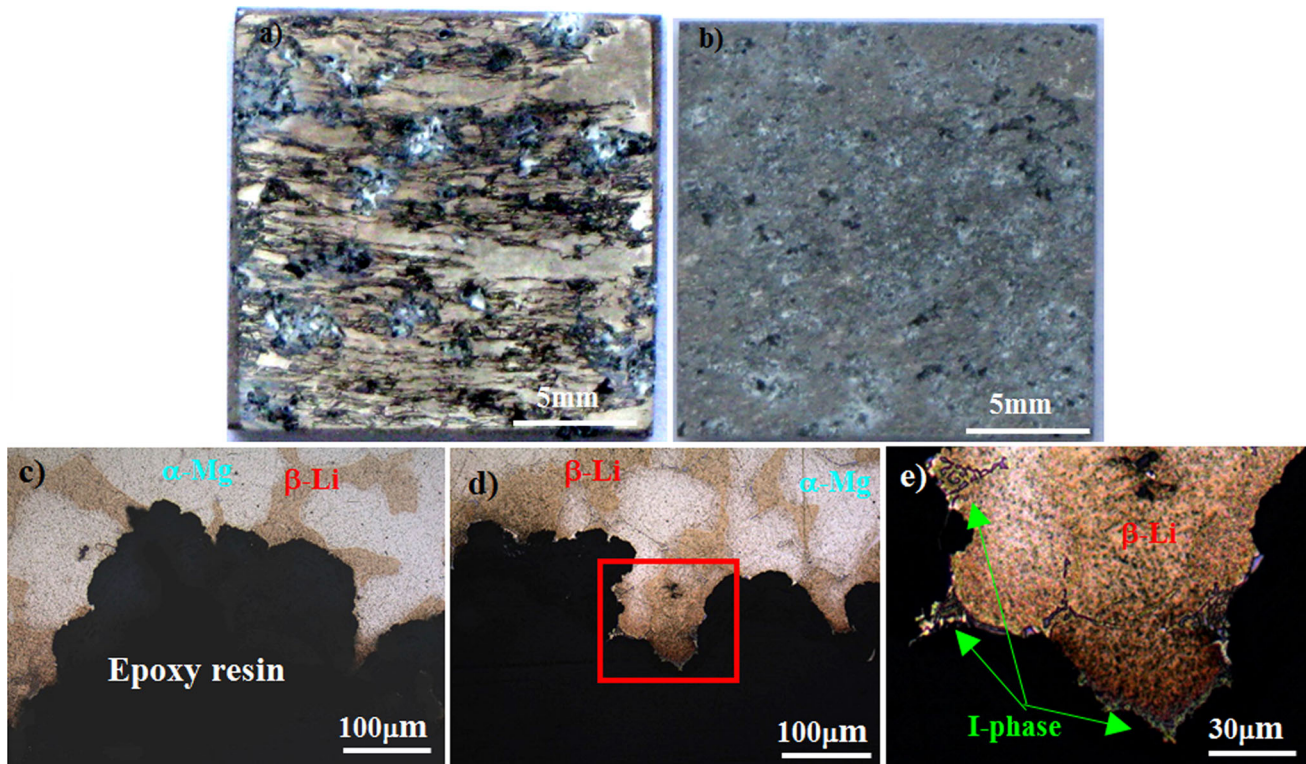


Fig. 2 Observation to the samples after immersion in 0.1 M NaCl for 7 days: **a, b** overall surfaces, **c, d** cross sections of the as-cast Mg-6Li and Mg-6Li-6Zn-1.2Y alloys, **e** high-magnification observation to the squared area in image **d**. Bright holes and black filaments in image **a** are the deep pits and the filiform corrosion, respectively. [70]

resistance reached the best when the Y content was 1.5 wt%. However, in the investigation about the long-term corrosion behavior of duplex-structured Mg-Li-Al-(RE) alloys in simulated body fluids, Lee et al. [75] reported that the corrosion rate of LA92 alloys was lower than that of rare earth-containing LAE912 and LAE922 alloys, and even lower than the WE series Mg alloys without lithium. Therefore, for the duplex-structured Mg-Li alloys, the controlling of the localized corrosion due to the corrosion couple between α -Mg and β -Li phases plays the key role in determining their corrosion resistance. The addition of rare earth elements for the formation of continuously distributed phase pockets or finely precipitates at α -Mg/ β -Li interfaces is an effective way for weakening the corrosion attack of the duplex-structured Mg-Li alloys. Due to the difference in crystallographic structure between α -Mg and β -Li phases, the main composed constituents on the surface films of two phases are different, and then, their corrosion protection to the underneath matrix must be different. Moreover, the degradation in their corrosion resistance can hardly be weakened by the formed rare earth-containing phases or precipitates at α -Mg/ β -Li interfaces. However, so far, it greatly lacks investigations about how to improve the protectiveness of surface films

formed on the α -Mg and β -Li phases by adding suitable alloying elements.

4 Corrosion Behavior of BCC-Structured Single-Phase Mg-Li Alloys

Since the content of active element Li in the BCC-structured Mg-Li alloys is high, it is intuitively understood that the corrosion resistance of the alloy should be quite poor. However, early studies demonstrated that the corrosion rate of the binary BCC-structured Mg-12Li alloy was only 0.4 mg/cm²/day in 0.1 M NaCl solution, which was significantly lower than the corrosion rate (142.8 mg/cm²/day) of pure Mg [76]. The main reason was that the presence of lithium element resulted in the high alkalinity at localized areas, which in turn ensured the existence of a stable and intact the Mg(OH)₂ protective film in a long period [76].

Morishige et al. [77] reported that the addition of element Al inhibited the peeling corrosion of Mg-14Li-based alloys, but a high content of Al would reduce their corrosion resistance. The main mechanism was that when the Al content was high, a certain amount of cathode AlLi phases would precipitate in the matrix, causing the formation of

corrosion couples with the matrix, and thus accelerated the anode dissolution of the matrix [77]. With an optimal Al content of 3 wt%, the Mg–14Li-based alloy maintained more solid-soluble Al atoms in the matrix and no AlLi particles were precipitated. In addition, the inevitable introduction of Fe element could significantly reduce the corrosion resistance of Mg–14Li-based alloys, but there was no critical threshold for the content of Fe element [78]. Xu et al. [28] reported that after a series of thermal treatments and processing deformation, the corrosion resistance of BCC-structured Mg–Li alloys in NaCl solution exceeded that of all Mg alloys reported so far, which was mainly due to the formation of dense and evenly distributed Li_2CO_3 protective film having self-repairing function. The schematic illustration about the protective effect of Li_2CO_3 film on corrosion resistance of BCC-structured Mg–Li alloys is shown in Fig. 3.

The main mechanism is that on exposure to the atmospheric (humid) air, Mg and Li will oxidize preferentially to form MgO and Li_2O that subsequently depletes most of the Li adjacent to these layers to generate the underlying Mg-rich zone [28]. Then, the rapidly formed Li_2O layer reacts with atmospheric CO_2 to generate the Li_2CO_3 film. By using ICP-MS online testing technology, Hu et al. quantitatively measured the content of Mg^{2+} and Li^+ dissolved in 0.01 M NaCl solution and the determined $\text{Mg}^{2+}/\text{Li}^+$ ratio was approximately 3.4, which was significantly higher than the atomic ratio of Mg to Li in Mg–33at.%Li alloy. Therefore, the surface of BCC-structured Mg–Li alloys was prone to form an insoluble and Li-rich protective film [79]. However, so far, the good corrosion performance of BCC-structured Mg–Li-based alloys was measured in a short immersion period. As for their long-term corrosion behavior, no further reports can be referred. Meanwhile, the degradation process of protective surface films is still unknown. Therefore, it is hard to have a reasonable comparison in terms of corrosion resistance of BCC-structured Mg–Li-based alloys and traditional Mg alloys.

5 Corrosion Protective Film on the Surfaces of Mg–Li Alloys

In general, Pilling–Bedworth ratio (PBR) is the ratio between the volume of each metallic ion in the oxide film and the volume of each metallic atom in the metal, i.e., the ratio between the volume of formed oxides and the volume of consumed metal, reflecting the coverage and stress status of oxide films [80]. When the PBR value is between 1 and 2, a certain degree of compressive stress will be produced in the surface oxide film. Then, the oxide film layer is relatively dense and the anti-oxidation/anti-corrosion of the

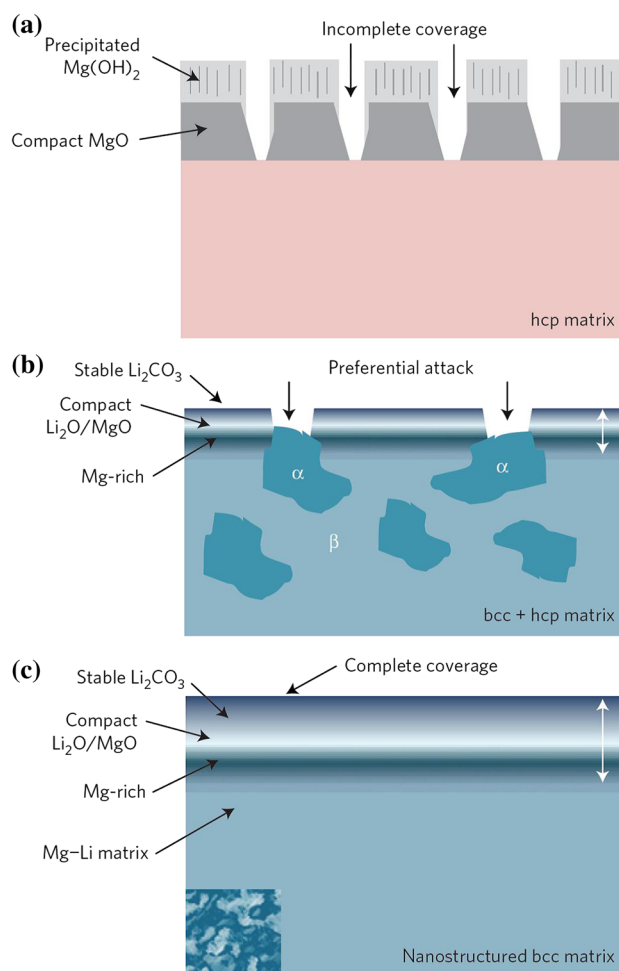


Fig. 3 Surface layer formation on HCP Mg and BCC Mg–Li after exposure to standard atmospheric conditions. Images **a–c**: schematics of the incomplete coverage of the surface film developed on conventional HCP Mg alloys **a**; thin surface film on the extruded Mg–Li alloy and potential reaction sites due to the conventional two-phase structure **b**; and complete coverage of the thicker surface film on the solute nanostructured BCC Mg–Li alloy **c**. The inset in **c** denotes the solute nanostructure. The white arrows in images **b**, **c** represent schematically the difference in thickness of the surface layers between the extruded and solute nanostructured alloys [28]

metal will be higher. When PBR values are less than 1 or greater than 2, tension stress or excessive compressive stress will be generated in oxide films, which results in easy rupture of surface oxide film and the lower anti-oxidation/anti-corrosion of metals [80]. The PBR values of chemical compounds formed on the surface of binary Mg–Li alloys are listed in Table 1.

Table 1 PBR of some chemical compounds [80]

Compound	Li_2O	MgO	LiOH	$\text{Mg}(\text{OH})_2$	Li_2CO_3	MgCO_3
PBR	0.57	0.8	1.26	1.80	1.35	2.04

Since Mg and Li are all active metal elements, the oxide film layer can be easily formed on the surface of Mg–Li alloys when exposed to the corrosive environments. However, its surface oxide film layer is not the same dense as that on the surface of Al alloys, resulting in lower corrosion resistance of Mg–Li alloys, which severely limits their widespread applications [81, 82]. So far, lots of work has been carried out on the study of surface films of traditional Mg alloys [83–85]. In aqueous solution, the outer surface layer of Mg alloys tended to form a loose and porous $\text{Mg}(\text{OH})_2$ product layer with a thickness of up to 500 nm, while the inner layer usually had a thin and dense MgO and its thickness was about 50 nm [86, 87]. The typical surface and cross-sectional morphologies of the corrosion films on the surfaces of pure Mg and Mg alloys are shown in Fig. 4.

In addition, MgO could react with water molecules to form $\text{Mg}(\text{OH})_2$ and a schematic diagram about the formation mechanism of corrosion products on the surface of Mg alloys was proposed, as shown in Fig. 5 [88]. Although the addition of alloying elements in Mg alloys can optimize the composition of surface films, the main component is still $\text{Mg}(\text{OH})_2$, and thus, it can hardly improve the corrosion resistance of Mg alloys [89–92]. Therefore, a passive corrosion-resistant Mg alloy would require a protective surface film and a surface film which can have self-repairing capability after being damaged [93].

However, so far, there are relatively few reports on the corrosion films of Mg–Li alloys. From the limited reference literature, it can be known that due to the addition of Li, the composition in surface corrosion products of Mg–Li alloys becomes much more complicated and has a typical multilayer structure. For example, the film structure on the surface of Mg–8Li alloy could be divided into four layers when exposed to the atmospheric environment. From the outermost layer to the innermost layer, their compositions

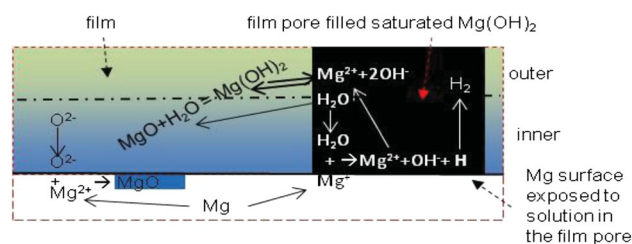


Fig. 5 Schematic illustration of the formation of Mg surface film [88]

are as follows: (1) the outermost layer contains $\text{Mg}(\text{OH})_2$ and Li_2O ; (2) the second layer contains $\text{Mg}(\text{OH})_2$, Li_2O and MgO; (3) the third layer contains $\text{Mg}(\text{OH})_2$, MgO, LiOH, Li_2O and Mg; and (4) the innermost layer contains MgO, Li_2O , Li and Mg [44]. Zeng et al. also reported that a four-layer structured film could be formed on the surface of Mg–9.29Li–0.88Ca (wt%) under the atmospheric environment, but the compositions in the corrosion product film was different from those of Mg–8Li alloy, i.e., the outermost layers were Li_2O , LiOH and Li_2CO_3 ; the second layer was LiOH, Li_2O_2 , Li_2CO_3 , MgCO_3 and LiH; the third layer was Li_2O_2 , Li_2O , MgO and CaO; and the innermost layer was composed of the oxides of Mg and Li [80]. Based on the PBR values of different compounds, it can be seen that the components in product film for the effective protection of Mg–Li alloys may be $\text{Mg}(\text{OH})_2$, LiOH and Li_2CO_3 . Since $\text{Mg}(\text{OH})_2$ is a porous loose structure and LiOH is soluble in water, Li_2CO_3 plays the key role in improving the corrosion resistance of Mg–Li alloys. Under natural conditions, Li_2CO_3 can be generated by the reaction (1) [80]:



In addition, LiOH is usually used as CO_2 absorbers and widely used in aerospace and seabed systems [94, 95]. Therefore, the Li_2CO_3 layer can be formed on the surface of Mg–Li alloy and plays a positive role in improving the

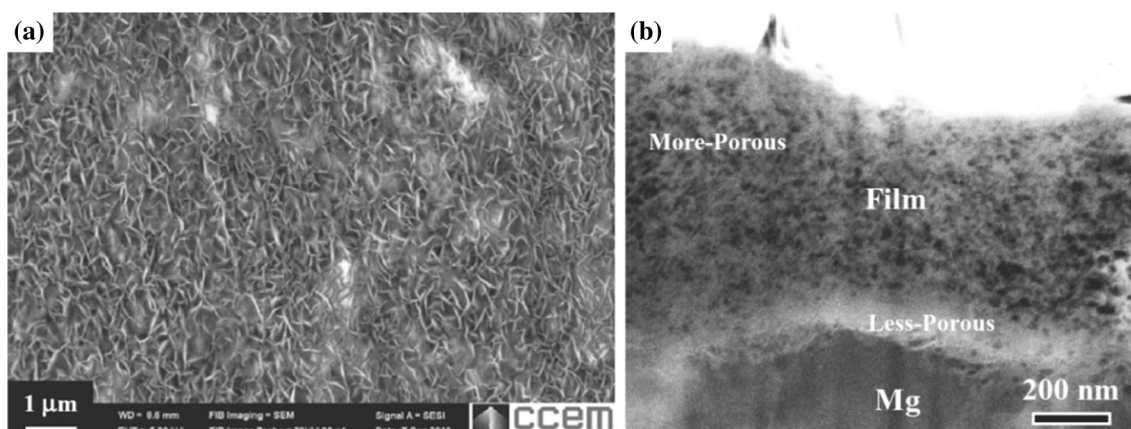


Fig. 4 Morphologies of surface film formed on pure Mg in 0.01 M NaCl after 24-h immersion: a surface morphology, b cross-sectional morphology [87]

corrosion resistance of the alloy. Similarly, Xu et al. and Hu et al. reported that the high corrosion resistance of Mg–10.95Li–3.29Al–0.19Zr–0.59Y alloy was attributed to the Li_2CO_3 protective film formed on the surface [28, 79]. However, Mg–Li alloys with single-phase HCP and duplex (HCP + BCC) structures did not exhibit excellent corrosion resistance. Thus, it can be seen that the formation of Li_2CO_3 protective film has a close relationship with the Li content in Mg–Li alloys, but the critical Li content in Mg–Li alloys for obtaining a high corrosion resistance is still unknown. On the other hand, adding a small amount of Mg into the Li matrix will form a Li–Mg alloy, which has the same crystallographic structure as the BCC-structured Mg–Li alloys. It has been reported that in the environment of removing CO_2 , Li– x Mg ($x = 0, 0.14, 1.16, 5$ wt%) alloys could also have good corrosion resistance. Through analyzing the surface composition, it was disclosed that the components in surface corrosion film were mainly MgH_2 , $\text{Mg}(\text{OH})_2$, LiOH and $\text{LiOH}\cdot\text{H}_2\text{O}$, but no Li_2CO_3 was detected. Among them, the presence of MgH_2 and $\text{Mg}(\text{OH})_2$ made the layer of (LiOH + $\text{LiOH}\cdot\text{H}_2\text{O}$) more dense [96, 97]. In addition, considering the CO_2 content in NaCl solution, the solubility of CO_2 in deionized aqueous solutions is very limited and NaCl can inhibit the solubility of CO_2 [98, 99]. Based on the description mentioned above, it is ambiguous whether the superior corrosion resistance of BCC-structured Mg–Li alloys in aqueous solutions is due to the formation of Li_2CO_3 in surface films or not. Moreover, previous characterization to the structure, morphology and constituents of surface protective films of Mg–Li alloys mainly relied on the X-ray photoelectron spectroscopy (XPS), X-ray diffraction (XRD) and scanning electron microscope (SEM). However, these techniques can hardly provide direct evidences about the existence of these constituents especially Li_2CO_3 in surface films. Thus, a deep characterization by employing the focused ion beam (FIB) technique to cut the surface oxide films formed on BCC-structured Mg–Li alloys for transmission electron microscopy (TEM) observation is needed.

6 Summary and Outlook

Based on the description mentioned above, it can be seen that the corrosion behavior of Mg–Li alloys with different crystallographic structures in saline medium solution is closely related to their corrosion product film layers. However, so far, the composition of the surface film layer and the distribution of the elements of Mg–Li alloys with high corrosion resistance are still not well understood. Moreover, the microstructure, cross-sectional morphology and detailed composition analysis of the protective films also need to be characterized. Since the corrosion time

reported in the literature is short, the evolution of the microstructural characteristics and protective capability of product films under longer immersion conditions are still unclear. Thus, future investigations on the corrosion behavior of Mg–Li alloys should focus on the structure, effective components and evolution of surface corrosion product film layers. At the same time, it is necessary to consider the effect of the addition of alloying elements on the protective effect of film layer. In addition, it is important to study the influence of the crystallographic texture of Mg–Li alloys induced by severe plastic deformation on corrosion behavior.

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