



Effects of Mg content on microstructure and electrochemical properties of Zn–Al–Mg alloys



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ABSTRACT

Zn–Al–Mg cast alloys with different Mg contents were prepared and characterized in this research. The solidification process of melt liquids and the formation mechanism of intermetallic were analyzed. The influence of microstructure on corrosion resistance property of alloy was also investigated. Results show that the microhardness of Zn–Al–Mg alloys increase with the increasing of Mg content, which is due to the grain refining effect. Ternary eutectic structures (Zn/Al/Zn–Mg intermetallic) were found in Zn–Al–Mg alloys. Polarization analyse reveals that Zn–5 wt.% Al–2 wt.% Mg (ZA5M2) alloy has the lowest corrosion current density of 1.03×10^{-3} A/cm² among all tested samples. Electrochemical impedance spectroscopy indicates that with the increasing of Mg content, the resistance of corrosion product increases from about 930 Ω of Zn–5 wt.% Al alloy to 8360 Ω of ZA5M2 alloy. The formation of nanostructured eutectic Zn/Al/Mg₂Zn₁₁ is probably the key factor of the improved corrosion property.

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1. Introduction

During the last 30 years, Zn–Al–Mg alloys have attracted worldwide attention due to their excellent mechanical and corrosion resistance properties as sacrificial coatings to substitute traditional Zn coating for the protection of steel structures [1–4]. Zn–Al–Mg alloys can also be used in light metal structures due to their advantages such as low-density, high-strength to weight ratio and high-specific toughness. [5,6]. Zn–Al–Mg coatings have been investigated extensively. However, the properties of Zn–Al–Mg alloys are seldom mentioned in literature. Due to the influence of steel substrate, it is expected that the microstructures of Zn–Al–Mg alloys may be different from Zn–Al–Mg coatings, which, in turn, influence their mechanical and corrosion resistance properties. This research aims to investigate the effect of Mg addition on the microstructure and corrosion properties of Zn–Al–Mg alloys, and explore how the microstructure influences the corrosion resistance property of Zn–Al–Mg alloys.

2. Experimental

2.1. Sample preparation

Zn–Al–Mg alloys with different Mg contents were prepared by casting method. Zn and Al metals (with purity >99.90%) were melt in a graphite crucible using

molten salt furnace at 680 °C, and then MgZn₂ intermetallic was added into the molten Zn–Al liquid with the shielding argon gas to inhibit the oxidation of Mg. The melt was homogenized by intense mechanical stirring, and then poured into a permanent mold and cooled down under Ar protection. The Zn–Al–Mg cast alloys were cut into 20 mm × 20 mm × 2 mm samples for tests. Zn, Al, Mg and Zn–5 wt.% Al were also tested for comparison purpose. MgZn₂ and Mg₂Zn₁₁ intermetallic were prepared using an induction vacuum furnace for further investigate their solidification processes and microstructure. The prepared Zn based alloys of the following composition were investigated, as shown in Table 1.

2.2. Characterization methods

Olympus BX60M optical microscope was used to conduct preliminary examination on the microstructure and morphology of the alloys. The alloy surface was polished and then etched using 2% Nital to reveal its microstructure. ESEM (Philips XL-30S) was employed for more detailed analyzes on the morphology and microstructure of prepared specimens. EDS (SUTW-Sapphire) attached to ESEM was used to analyze the elemental composition of the alloys. X-ray diffraction patterns of specimens were recorded using Bruker D2 PHASER desktop diffractometer. Cu Kα (0.15406 nm) radiation was used as X-ray source. A step-scan mode was used at the 2-theta range from 10° to 90° with a step length of 0.02°. The operating conditions were 40 kV and 100 mA. Diffraction patterns were analyzed using the DiffractPlus EVA software, which allows the identification of phases present in the material. The microhardness of coating was measured using a microhardness tester (Leco M400) with a Vickers diamond indenter. The applied loading time was 10 s with different forces ranging from 25 g to 100 g for different specimens. At least 5 measurements under the same condition were performed for each sample. The mean value was used as the nominal microhardness (HV) of the sample. The standard deviation was also calculated and presented with the mean value.

Electrochemical testing has become increasingly straightforward with the increasing computerization of electrochemical instrumentation. Electrochemical testing is a quick and efficient method to assess the corrosion resistance property

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Table 1
Chemical composition of the investigated specimens.

Number	Designation of specimens	Bath compositions (in wt.%)
#1	Zn	>99.90% Zn
#2	Al	>99.90% Al
#3	Mg	>99.90% Mg
#4	ZA5	Zn–5Al
#5	ZA5M1	Zn–5Al–1Mg
#6	ZA5M2	Zn–5Al–2Mg

of a specimen. It uses an external auxiliary power source to apply a voltage or current to drive the cathodic or anodic reaction out of the equilibrium state, causing electrons to flow between the anodic and cathodic sites, thus the specimen is corroded faster than in normal environment [7,8]. In this research, CHI604D electrochemical analyzer was used to test the electrochemical behaviors of prepared alloys, including open circuit potential (OCP)-time plots, polarization curves and electrochemical impedance spectroscopy (EIS). All the electrochemical tests were performed in non-de-aerated 3.5 wt.% NaCl aqueous solution in a flat cell at ambient temperature (20 °C). Saturated calomel electrode (SCE) and platinum electrode (Pt) were used as reference electrode and counter electrode, respectively. The working electrode was the specimen that to be tested. Pretreatments in ethanol and water were performed before testing. The exposed active surface area was 1 cm² when the specimen was clamped on a commercial flat cell designed for 3-electrode electrochemical test. The polarization measurement was obtained at a scanning rate of 0.001 V/s between –0.9 V vs. SCE and –1.7 V vs. SCE. The EIS was tested in a frequency range of 10^{–2}–10⁵ Hz to investigate the corrosion mechanism.

3. Results and discussion

3.1. Microstructure

The microstructures of prepared Zn–Al–Mg alloys are shown in Fig. 1. Their elemental compositions at different locations are listed in Table 2. Phase diagrams of Zn–Mg [9] and Zn–Al [10] are given in Fig. 2. There are mainly two types of morphologies in ZA5 alloy: Zn crystal (with corresponding composition of “eds a1” in Table 2, same for following) and Zn–Al binary area (eds a2 and eds a3 in Table 2). The elemental compositions at a2 and a3 are similar. They might have the same structure with different orientations. According to Zn–Al binary phase diagram in Fig. 2b, the equilibrium structure of ZA5 alloy at room temperature is composed of simple Al and Zn phases. During the solidification of ZA5, Al is rejected into the inter-dendritic region of primarily solidified Zn. Intermetallic compounds will not form in the ZA5 binary system under equilibrium condition. For ZA5M1 alloy, there are mainly three types of morphologies: Zn crystal (eds b1 in Table 2), Zn–Al–Mg ternary eutectic area (eds b2 in Table 2) and Al rich area (eds b3 in Table 2). With the increasing content of Mg, the crystal size of Zn decreased and more eutectics formed around Zn crystals in ZA5M2 alloy. ZA5M2 alloy mainly has three types of morphologies: Zn crystal (eds c1 in Table 2) and Zn–Al–Mg ternary eutectic areas (eds c2 and eds c3 in Table 2). As indicated by arrows in the inset c2, the ternary eutectic area of ZA5M2 alloy has three phases. In combination with the XRD results in Fig. 3, the three phases are Al, Zn and Mg₂Zn₁₁ intermetallic phases. During the solidification of ZA5M2, A primary Zn phase nucleates first; Zn–Mg intermetallic forms around Zn crystals and Al is rejected into the inter-dendritic region. Similar results were reported by Arndt et al. [11] in their research about Zn–Al–Mg coatings.

3.2. Phase structure

The XRD patterns of ternary Zn–Al–Mg alloys are given in Fig. 3, and are compared with those of binary Zn–Al alloy and pure Zn. The results are consistent with morphologies and phase diagrams shown in Figs. 1 and 2: Zn–Al intermetallic compounds were not found in ZA5 alloy. It consists of primary Zn and Al phases.

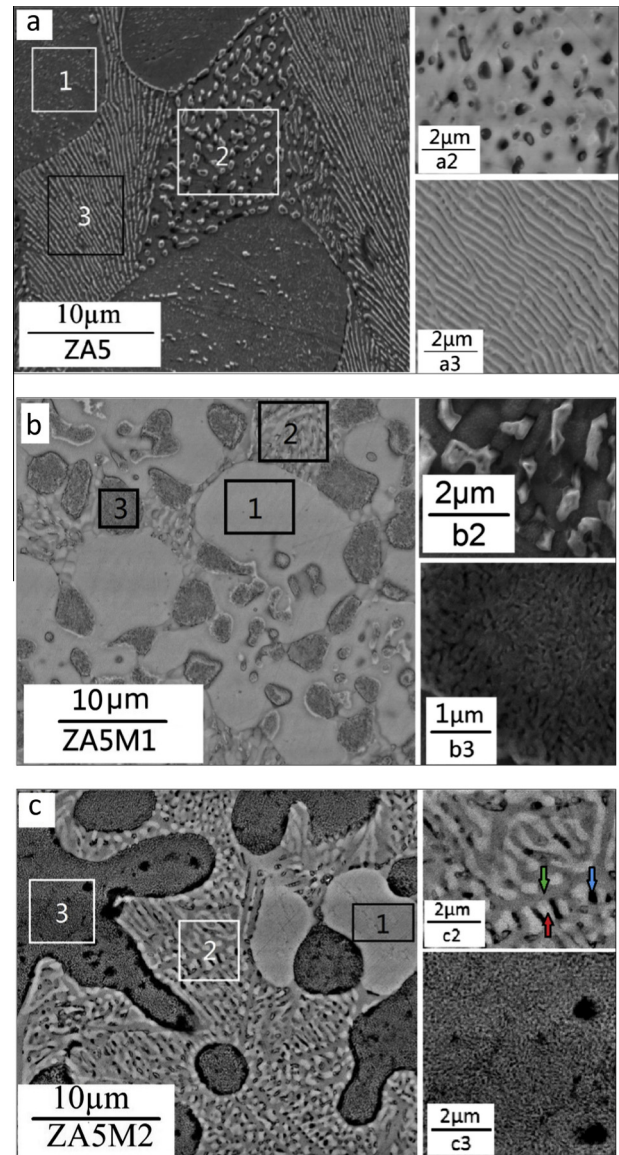


Fig. 1. ESEM images of etched Zn–Al–Mg alloys with different Mg contents, (a) ZA5 alloy, (b) ZA5M1 alloy and (c) ZA5M2 alloy.

Table 2
Elemental composition of different locations in Fig. 1.

Positions	Zn (wt.%)	Al (wt.%)	Mg (wt.%)	O (wt.%)
eds a1	97.21	2.16	0	0.62
eds a2	90.39	8.96	0	0.65
eds a3	90.85	8.36	0	0.79
eds b1	97.60	1.63	0	0.77
eds b2	88.75	7.44	3.2	0.6
eds b3	69.19	29.35	0.11	1.35
eds c1	98.79	1.21	0	0
eds c2	92.08	5.06	2.85	0
eds c3	89.11	9.61	1.28	0

Zn–Al–Mg ternary intermetallic compounds were not identified in ZA5M1 and ZA5M2. Zn–Mg intermetallic compounds laves phase MgZn₂ and cubic lattice Mg₂Zn₁₁ were observed in ZA5M1 alloy. While, large amount of cubic lattice Mg₂Zn₁₁ were found in ZA5M2 alloy. It may due to the equilibrium eutectic reaction L → Zn + Mg₂Zn₁₁ and the non-equilibrium eutectic reaction L → Zn + MgZn₂ [9,12,13].

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