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Investigation on Corrosion of Magnesium and its Alloys

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Abstract

Magnesium alloys are well known for their high specific strength. There is growing interest in the alloys as structural materials in the automotive, aerospace and electronic industries. However, the corrosion performance of most magnesium alloys is not satisfactory for some practical applications. The Cooperative Research Centre for Cast Metals Manufacturing (CAST) has been investigating the corrosion behaviour of magnesium and its alloys since 1995. This paper presents some of the major achievements in this area recently made by CAST. The following issues are addressed: Negative Difference Effect (NDE), role of alloying elements, effect of microstructure and galvanic corrosion. It was found that: (a) the NDE is a common phenomenon for all alloys investigated; (b) alloying with aluminium and zirconium can stabilise the matrix phase of magnesium alloys in corrosion; (c) in the aluminium containing alloys the β phase appears to have a beneficial effect on the corrosion resistance especially where a fine grain size is produced, but in some cases, its presence in an alloy accelerates the corrosion of the alloy; (d) galvanic corrosion of magnesium alloys strongly depends on the cathode material and the manner by which the galvanic couples are exposed to the environment.

Keywords: corrosion, magnesium, electrochemistry

Introduction

The density of magnesium is only two thirds that of aluminium. Because of their high strength-to-weight ratio, magnesium alloys are being increasingly used, particularly in the automotive industry. Over the past 10 years, the weight of magnesium applications in North American automobiles has increased about 4 times [1].

However, the relatively poor corrosion performance of magnesium alloys [1, 2, 3, 4] is a major issue which limits their application. Currently, the use of magnesium alloys in the automotive industry is mainly limited to components in mild service environments [5]. For those parts exposed to outside environments, corrosion is still a serious issue. At a recent international conference "Magnesium Alloys 2003, Japan", corrosion was listed as one of the areas where technical progress is necessary for further expansion of magnesium use by automotive companies.

The Cooperative Research Centre for Cast Metals Manufacturing (CAST) was established as part of the Australian Commonwealth Government's Cooperative Research Centre's Program and commenced operations in July 1999. The Centre continues and extends research related activities on light metals previously carried out by the CRC for Alloy and Solidification Technology that operated between 1993 and 1999. The corrosion and prevention of magnesium and its alloys has been one of the most important areas under the CAST's research program since 1995. The research effort in this area is focused on solving corrosion problems relative to the application of magnesium alloys in the automotive industries. Nevertheless, encompassed by the requirements of the applied research, some essential fundamental studies have also been conducted in CAST. This paper presents a summary of some of the fundamental research achievements in this area recently made by CAST.

Negative Difference Effect (NDE)

Magnesium and its alloys have a special electrochemical behaviour, the Negative Different Effect (NDE). The NDE is usually defined by the difference Δ for a galvanostatic applied current density as

$$\Delta = I_{\text{corr}} - I_{\text{H,m}} \quad (1)$$

where I_{corr} is the corrosion rate or the spontaneous hydrogen evolution rate (HER) at the corrosion potential; $I_{\text{H,m}}$ is the measured HER at an applied anodic current density or potential. The polarization behaviour is called NDE when $\Delta < 0$. According to this definition, the NDE is a phenomenon under an anodic polarisation condition.

For most conventional metals, such as iron, copper, nickel, etc, the cathodic process normally slows down while the anodic dissolution of the metal is accelerated by anodic polarisation. However, for magnesium or its alloys, HER increases as the applied polarisation potential becomes more positive. Figure 1 displays a typical NDE case for pure magnesium in a NaCl solution [6, 7]. It can be seen that HER increases with increase in anodic polarisation current density.

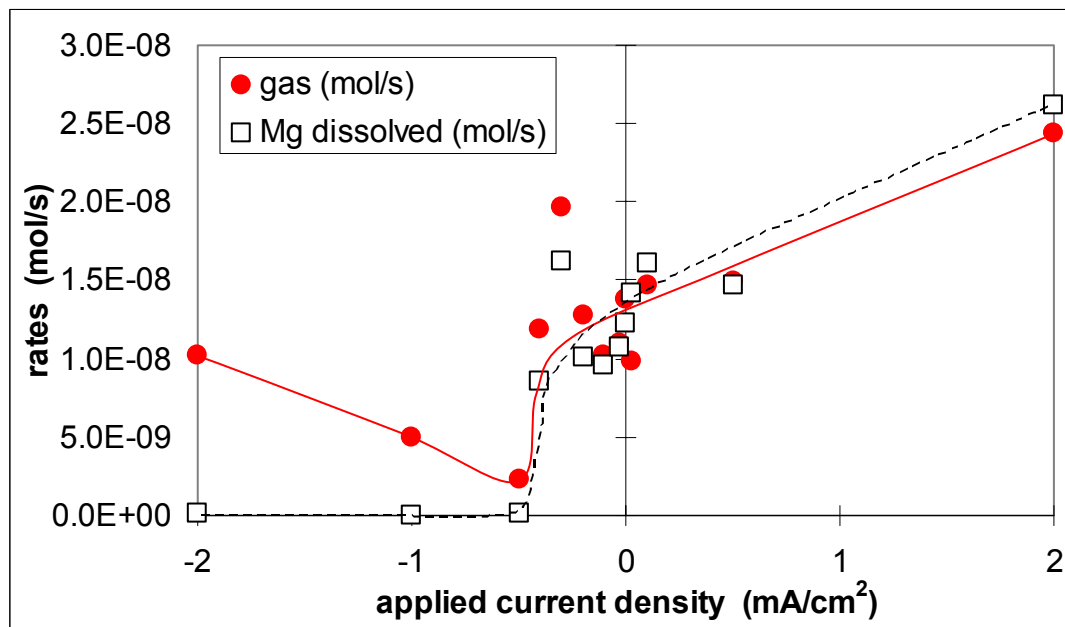


Figure 1. Hydrogen evolution rate and anodic dissolution rate for pure magnesium in 1 N NaCl of pH11 under different polarisation current densities

The NDE is a common phenomenon for magnesium alloys [8, 9, 10, 11, 12]. Figure 2 presents the NDE phenomena for some magnesium alloys.

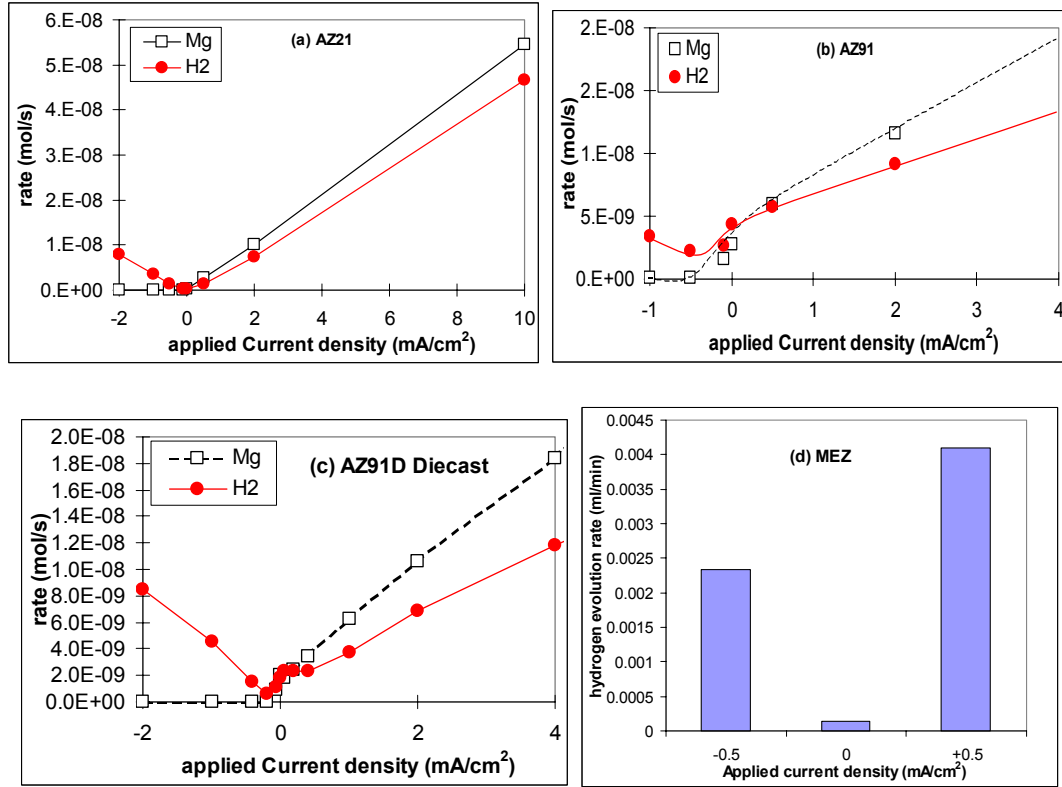


Figure 2. Hydrogen evolution rate and anodic dissolution rate under different polarisation current densities for (a) AZ21 (Al₂Zn₁) in 1N NaCl of pH11, (b) AZ91 (Al₉Zn₁) in 1N NaCl of pH11 (c) diecast AZ91 (Al₉Zn_{0.8}Mn_{0.3}) in 1N NaCl of pH11 and (d) sand cast MEZ (REZn) in 5% NaCl

The basic feature of the NDE phenomenon is that HER increases as the polarisation current density becomes more positive. As shown in Figure 1 and Figure 2 (b) and (c), this special hydrogen evolution behaviour actually starts at a cathodic current density. This means that the NDE can occur under a cathodic polarisation condition, not only under anodic polarisation as specified in the definition. The NDE is not always associated with anodic polarisation. This point has not been clearly illustrated before.

The above figures also show another interesting phenomenon; the HER first decreases and then increases as the polarisation current changes from a negative to a positive value. These two different hydrogen

processes are termed as “cathodic hydrogen evolution” and “anodic hydrogen evolution” respectively [11] in this paper. This is because the first one occurs when the electrode is under cathodic polarisation and the second one is closely associated with the anodic shift of the applied polarisation potential or current density and it mainly occurs in the anodic polarisation region. The different trends of the dependence of the HER on polarisation current suggest that different mechanisms are controlling the “cathodic” and “anodic” hydrogen evolution processes. It is a normal mechanism by which HER decreases as polarisation current or potential becomes more positive. However, the increase in HER with increasing anodic current density or anodic polarisation potential is an abnormal behaviour from an electrochemical point of view. It means that this hydrogen evolution process could follow a different mechanism.

The differences in these two HER behaviours can be further illustrated by in-situ observation [11] of a polarised MEZ magnesium alloy (0.18wt%Ce, 0.34wt%Nd, 0.9wt%Pr, 0.65wt%La, 0.54wt%Zn) in a NaCl solution (see Figure 3). When MEZ was cathodically polarised to -1.8V/SCE , “cathodic hydrogen evolution” was observed from many sites (e.g. particles) in grains or grain boundaries. When the polarisation potential became less negative, e.g. at -1.7V/SCE , less sites produced hydrogen bubbles, and the hydrogen evolution rate was lower. So far, “anodic hydrogen evolution” was not observed. If the polarisation potential was increased to a more noble value, e.g. -1.4V/SCE , then “cathodic hydrogen evolution” became even less and almost stopped. Meanwhile in another area where corrosion was occurring, “anodic hydrogen evolution” was observed to be very intensive. When the specimen was cathodically polarised back to a cathodic potential, “anodic hydrogen evolution” became weaker and weaker, while the “cathodic hydrogen evolution” became faster again. At -2V/SCE , “anodic hydrogen evolution” stopped in the corroded area (corrosion also stopped there at this potential), but “cathodic hydrogen evolution” was even more intensive in a number of sites in the uncorroded areas.

These phenomena strongly suggest that “cathodic hydrogen evolution” is mainly from the uncorroded area of a magnesium alloy, which is

responsible for the cathodic polarisation behaviour as represented by the cathodic branch of the polarisation curve of the alloy; and the

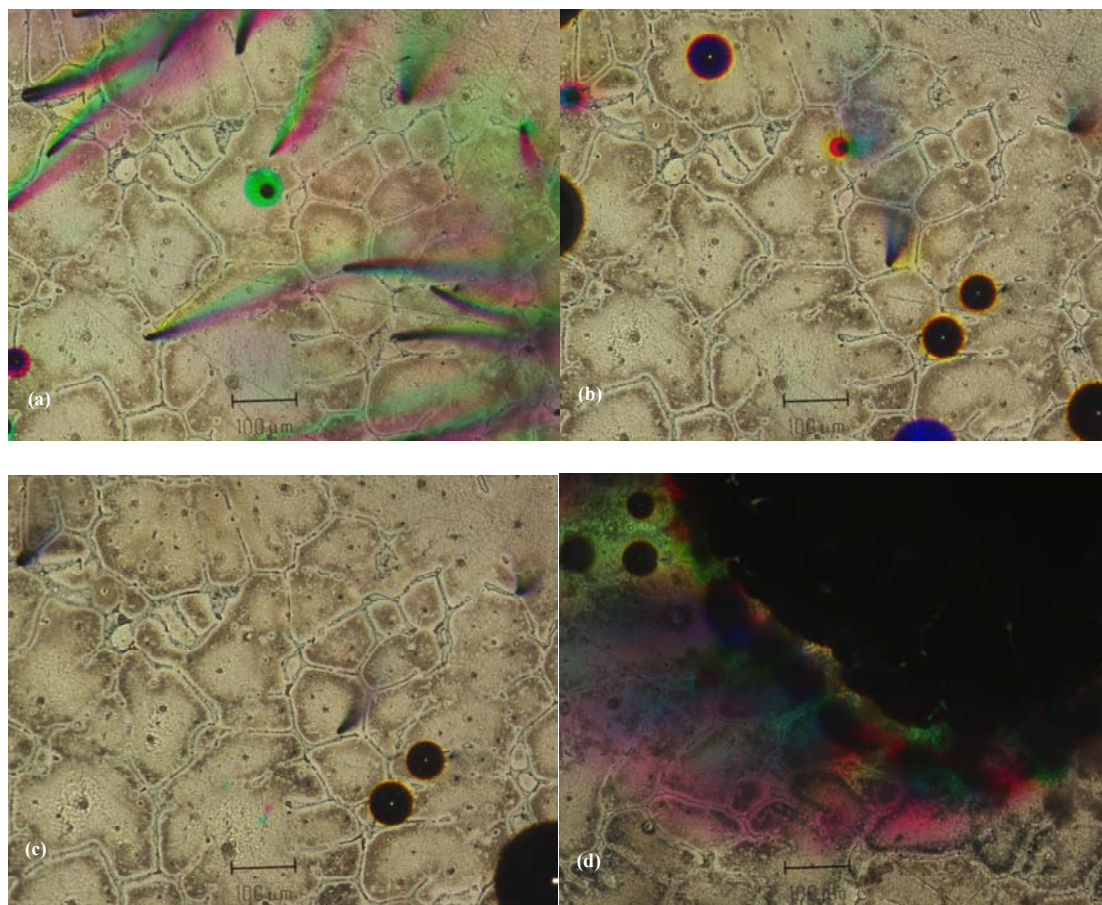


Figure 3 Hydrogen evolution and corrosion of polarised MEZ magnesium alloy: (a) at cathodic potential -1.8V/SCE in an area; (b) at cathodic potential -1.7V/SCE in the same area as (a); (c) at anodic potential -1.4V/SCE in the same area as (a); (d) at anodic potential -1.4V/SCE in another area; (e) at cathodic potential -2V/SCE in the same area as (d) after being anodically polarised at -1.4V/SCE . The cloudy “trails” of hydrogen bubbles and the trapped hydrogen bubbles associated with “cathodic hydrogen evolution” are clearly observed in (a) to (e). The cloudy zones along the edges of black (corroding) areas associated with “anodic hydrogen evolution” are observed in (d) and (e).

anodic dissolution of a magnesium alloy is closely associated with “anodic hydrogen evolution” from corroding areas of the alloy, and the anodic dissolution and “anodic hydrogen evolution” are responsible for the anodic polarisation behaviour of the magnesium alloy.

The “cathodic hydrogen evolution” can be easily understood as a normal cathodic electrochemical process. However, the “anodic hydrogen evolution” appears to be a cathodic reaction that is “abnormally” accelerated as polarisation potential becomes more positive. To explain such an electrochemical behaviour, the following model is proposed [4, 7] (see Figure 4):

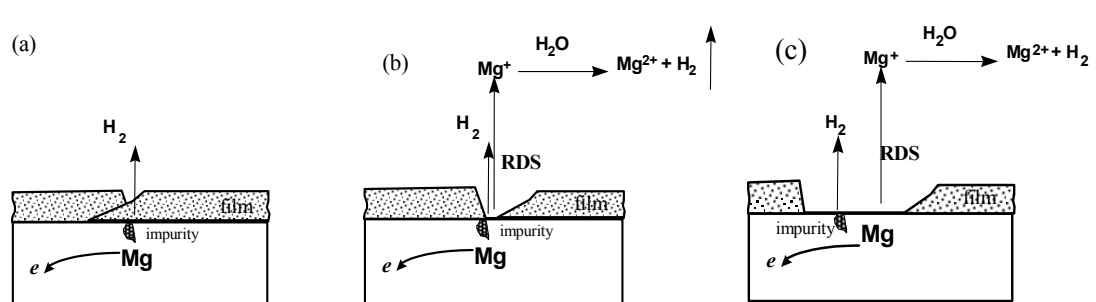
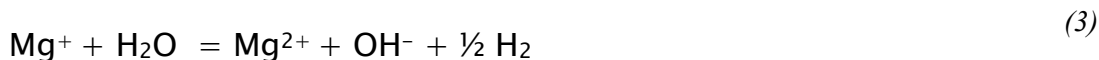


Figure 4. Electrochemical corrosion and Negative Difference Effect on the magnesium surface: (a) at a very negative cathodic potential; (b) at the “pitting potential”; (c) at a potential more positive than the “pitting potential”

At a very negative potential or current density, the surface film on magnesium is intact. There is no film-free area, so the anodic dissolution of magnesium is very low, almost zero. “Cathodic hydrogen evolution” can still proceed on the surface film at such a negative potential. The hydrogen evolution rate decreases as the potential becomes more positive until a “pitting potential” is reached. At the “pitting potential”, the surface film starts to breakdown, and both hydrogen evolution and magnesium dissolution become much easier in the defects or film-free areas. In these areas, magnesium corrosion occurs, which produces univalent magnesium ions and subsequently leads to generation of hydrogen:



The anodic dissolution rates of reactions (2) increase as polarisation potential or current density becomes more positive. Therefore, more hydrogen is produced at a higher potential by reaction (3). This explains the “anodic hydrogen evolution” phenomenon. The “pitting potential” is not necessarily the same as the corrosion potential and could be more negative than the corrosion potential in most cases. So NDE can occur even at a cathodic potential. When the electrode is polarised back to a very negative potential, reaction (2) will stop. As a result, reaction (3) slows down, and the “anodic hydrogen evolution” ceases too. Certainly, in the later stage after the surface has been severely corroded, undermining of magnesium particles would occur.

It is quite clear that the “special” electrochemical behaviour is caused by complicated reactions. At the corrosion potential, there are at least three reactions occurring on the surface: anodic dissolution of magnesium, “anodic hydrogen evolution” and “cathodic hydrogen evolution”. The “cathodic hydrogen evolution” from uncorroded areas should be different from that from corroding areas. Therefore, the polarisation curve may not necessarily follow a simple Tafel equation and the corrosion rate at the corrosion potential cannot be estimated through Tafel extrapolation.

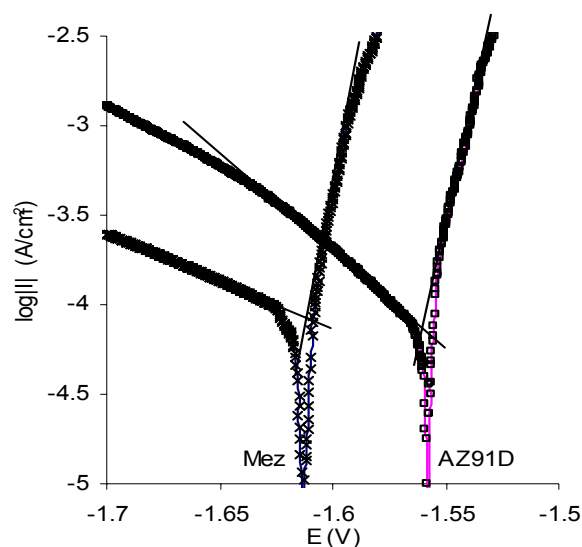


Figure 5. Polarisation curves for sand cast MEZ and diecast AZ91D in 5% NaCl (pH11)

For example, the polarisation curves of sand cast MEZ and diecast AZ91D (Al₉Zn_{0.8}Mn_{0.3}) shown in Figure 5 indicate that the corrosion rates of these two alloys are almost the same if estimated through “Tafel” extrapolation. However, the weight-loss measurements showed a very big difference between their corrosion rates. The weight-loss rate of MEZ is about 25 times higher than that of AZ91D [13]. The estimation based on the polarisation curves alone is incorrect.

Effect of alloying elements

Magnesium alloys in general can be divided into two groups: 1) those containing aluminium as the primary alloying element; and 2) those free of aluminium and containing a small amount of zirconium for the purpose of grain-refinement.

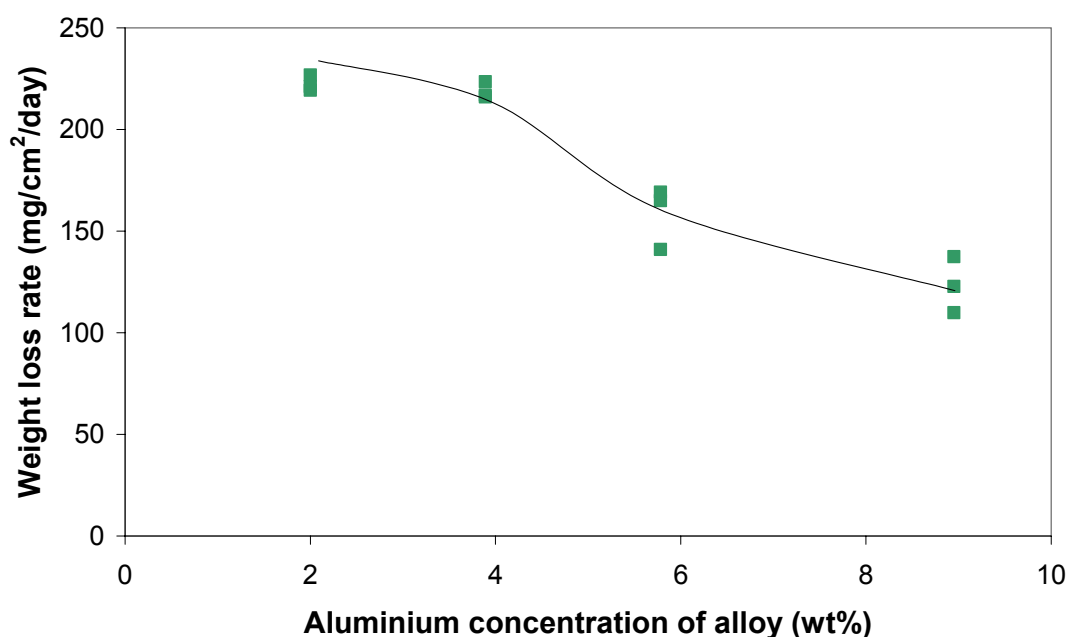


Figure 6. Average weight loss rates of Mg–Al single phase alloys with various aluminium contents after immersion in 5wt.% NaCl solution for 3 hours.

Aluminium is the most important alloying element in the first group of magnesium alloys. The addition of a certain amount of aluminium can introduce a secondary phase (β -Mg₁₇Al₁₂ intermetallic) into the

magnesium alloys. In most cases, the improved corrosion resistance is principally attributed to the secondary phase. This will be addressed later in this paper.

Solid solution aluminium in the matrix phase plays an important role in the corrosion of this group of magnesium alloys. The beneficial effect of aluminium in the α phase can be elucidated by the corrosion rates of several single α -phase alloys with various aluminium contents (see Figure 6) [14]. The corrosion resistance of the α phase matrix increases with increasing solid solution aluminium level. This is contrary to the explanation by Lunder et al [15] that the matrix phase becomes more anodically active as the aluminium content increases up to 8%.

For an AZ (Al–Zn) alloy, the solid solution aluminium content can vary from 1.5wt % in the grain centre to about 12wt % in the vicinity of the β phase [16]. Therefore, the corrosion behaviour could be very different in a grain of the α matrix phase. The evidence for this difference is the corrosion morphologies of AZ91E as shown in Figure 7 [17]. Corrosion mainly occurred in the interior of the α grain, and developed from grain to grain. In many cases, corrosion stopped at the grain boundary before it reached the β phase, where the aluminium content is much higher than the grain centre.

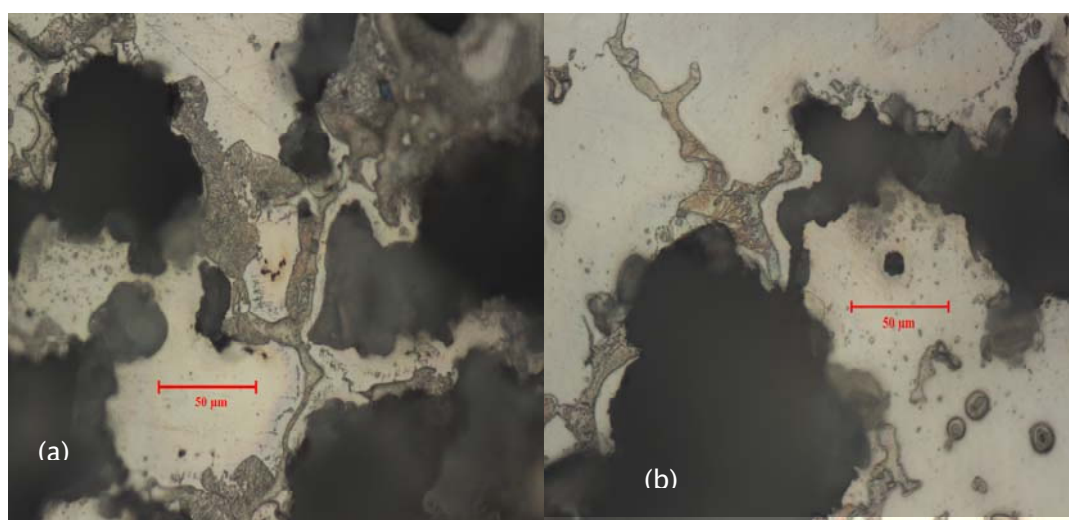


Figure 7. Corrosion morphologies after 4 hour immersion in 5% NaCl. (a) sand cast AZ91E and (b) sand cast AZ91E (Ca)

In the second group of alloys, the role of zirconium is, to some extent, as important as aluminium in the first group of alloys. The beneficial effect of zirconium is remarkable.

Zirconium is not only a powerful grain refiner, but also a very effective purifier for magnesium. Iron is an unavoidable impurity that can be easily introduced into most magnesium alloys during melting and casting processes. A trace level of iron impurity can significantly deteriorate the corrosion performance of a magnesium alloy. Zirconium can react with iron in molten magnesium to form iron–zirconium intermetallics which quickly settle out due to their high density. Therefore, the addition of zirconium in the second group of magnesium alloys can lead to higher purity and hence a more corrosion resistant magnesium alloy.

An example for the relationship between the zirconium addition, purity and the corrosion resistance of a magnesium alloy is presented in Table 1[[#ref12](#)]. A higher concentration of zirconium results in a lower iron impurity and slower corrosion of the magnesium alloy.

Table 1. Zirconium additions, iron impurity levels and corrosion rates of MEZ and grain refined MEZ magnesium alloys

	Zr (wt%)	Fe (wt%)	5 day B117 salt spray corrosion rate (mg/cm ² /day)
MEZ	0.005	0.013	15.65
Grain refined MEZ	0.60	0.004	0.31

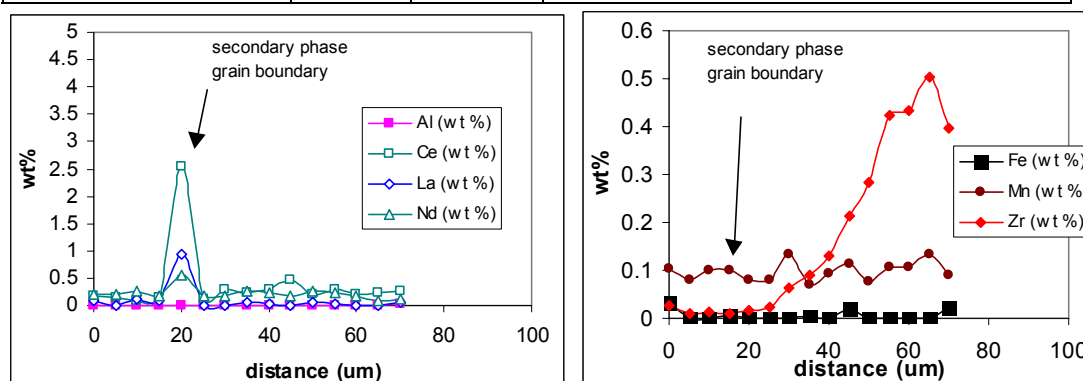


Figure 8 Elemental distribution across a grain boundary in a grain refined MEZ magnesium alloy

Similar to aluminium in the first group of alloys, the distribution of zirconium in the grain of the second group of alloys is not uniform. The grain centre is rich in zirconium (see Figure 8) [ref12].

The enrichment of zirconium in the grain centre leads to higher corrosion resistance of this zone. A simple optical observation of the corrosion morphology (see Figure 9) of the grain refined MEZ magnesium alloy has verified the relationship between the zirconium enrichment and the corrosion resistance of the grain interior[ref12]. From this photo, it can be clearly seen that many grain central areas remain uncorroded while the grain boundaries have been severely corroded. This indicates that zirconium can stabilise the magnesium matrix phase and reduce its corrosion rate.

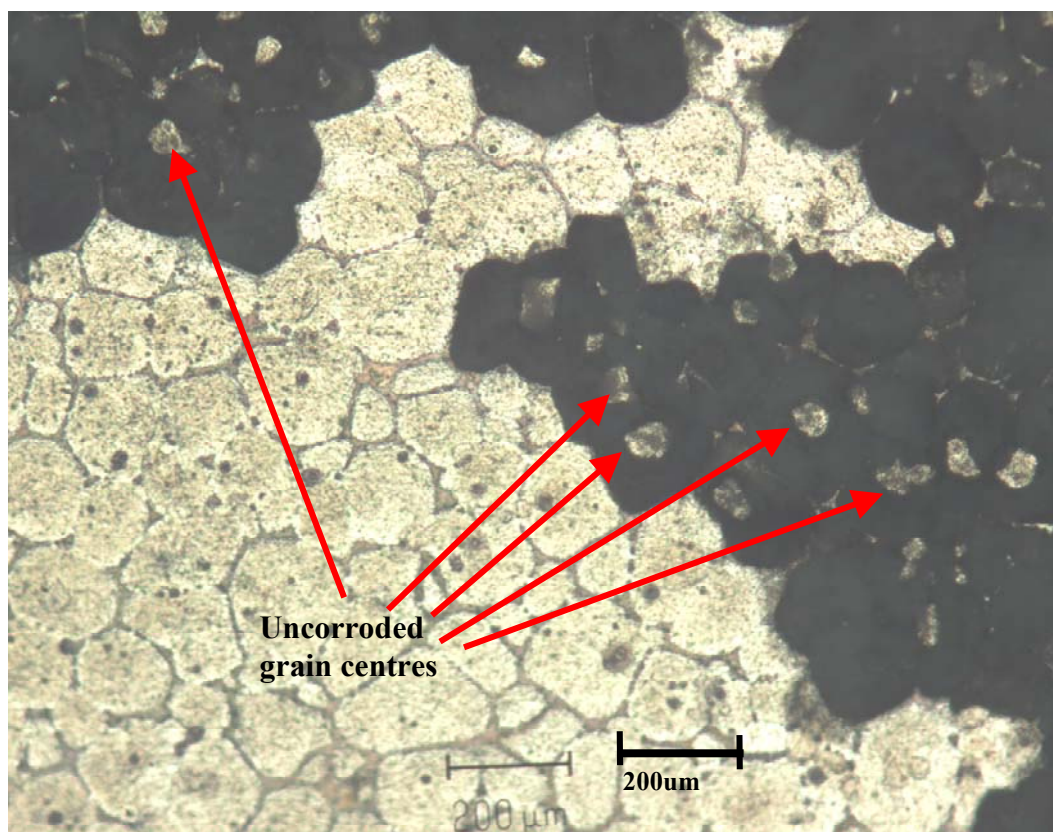


Figure 9 Optical micrograph of the surface of grain refined MEZ after immersion in 5% NaCl for 3 hours

The Effect of microstructure

The microstructure of an alloy usually refers to the phase constituents and their distribution as well as the grain size. All these have a

significant influence on the corrosion performance of a magnesium alloy.

The secondary phase in a magnesium alloy is normally inert and stable in corrosion. For example, in AZ alloys, β -phase is very corrosion resistant and normally not corroded if exposed to a sodium chloride solution [#ref9]. This can be clearly illustrated by the corrosion morphologies of AZ91E as shown in Figure 7. No corroded β -phase can be observed in the alloy. In most cases, even in the corroded areas where the matrix phase has been severely removed, the β -phase is still intact. Similarly, the secondary phase in the MEZ alloy can not be corroded [#ref11, #ref12]. In Figure 9, the secondary phase is also intact in the corroded areas.

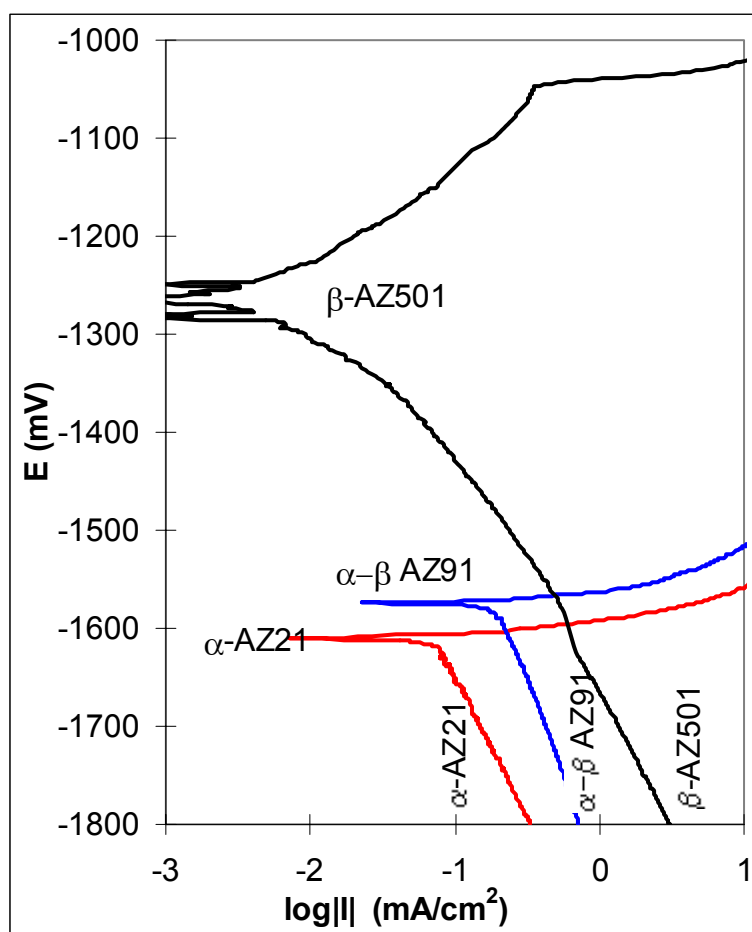


Figure 10. Polarisation curves for β phase (AZ501), α phase (AZ21) and α - β binary phase (AZ91) alloys in 1N NaCl (pH11)

The β -phase has an electrochemical polarisation behaviour different from the α matrix phase or the α - β binary phase alloys [#ref9]. Their

differences are compared in Figure 10. AZ501 is a β -phase alloy, AZ21 is an α -phase alloy and AZ91 is an α - β binary phase alloy. According to this figure, the corrosion potential of the β -phase is much more positive than the α -phase. This means that indicates that the β -phase is cathodic to the α phase and can accelerate the anodic dissolution of the α -phase through the galvanic effect in a binary phase alloy.

Even worse, The β -phase is a very effective cathode if it is coupled with the α -phase as its cathodic polarisation curve of the β -phase is much higher than the α -phase. Figure 11 shows that the corrosion rate increases after the introduction of β -phase into a Mg-Al matrix phase. The alloys presented in the figure were permanent mould cast. The microstructure of the Mg-1%Al alloy is a single α -phase matrix; the Mg-5%Al alloy has some β precipitates along the grain boundary; and more β precipitates are present along the grain boundary in the Mg-10%Al alloy.

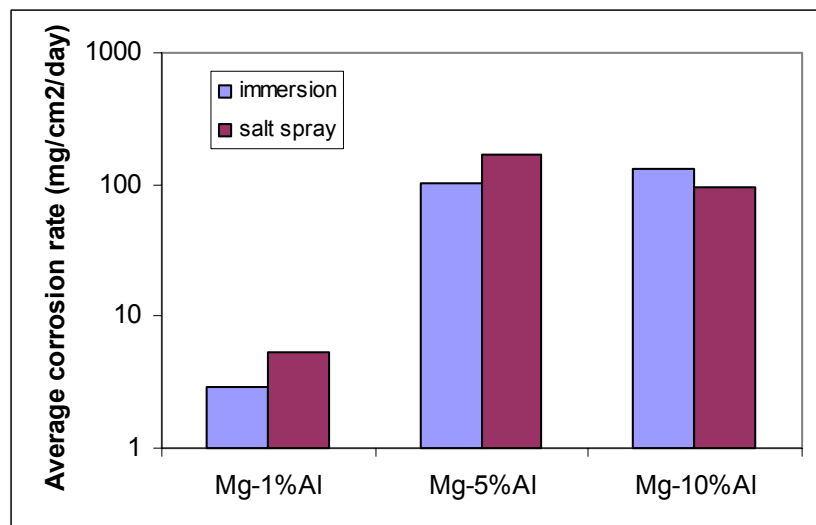


Figure 11. Influence on corrosion rate of the introduction of β -phase into the permanent mould cast matrix phase

This is contradictory to some other experimental results [ref10, ref17, 18]. For example, diecast AZ91D specimen has different microstructures in its interior and skin (see Figure 12). The skin has a greater amount of the β -phase than the interior, and the corrosion rate of the skin is about 10 times lower than the interior under the same salt solution (see Table 2).

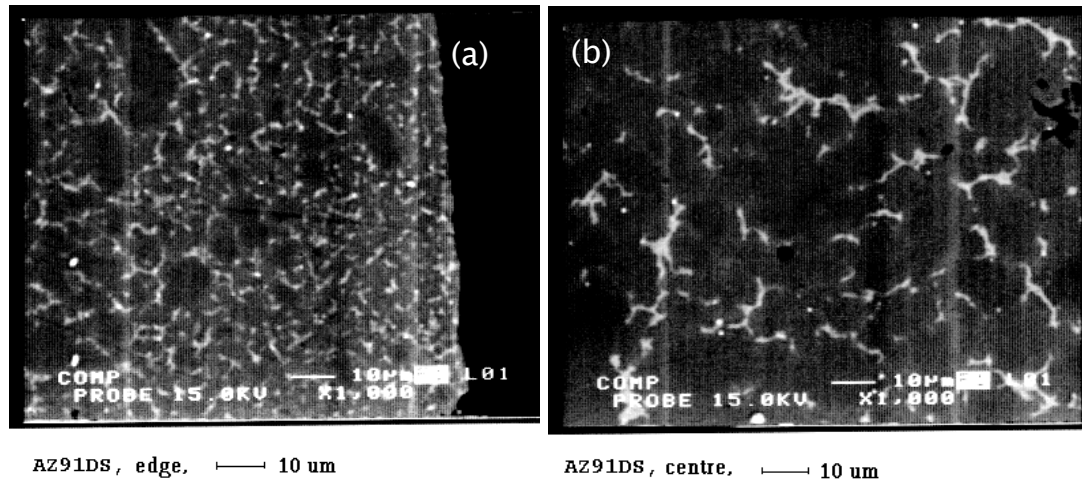


Figure 12. Back scattered SEM images of die cast AZ91D. (A) skin, (B) interior area

Table 2. Dissolution rates under open circuit conditions.

	penetration rate in 1N NaCl (pH11) (mm/y)
Interior of diecast AZ91D	5.72
Skin of diecast AZ91D	0.66

A proposed explanation for these contradictory results is that the β -phase has a dual role in corrosion. The β -phase in an AZ alloy can act as either a corrosion barrier or a galvanic cathode accelerating corrosion, depending on the amount and distribution of the β -phase. Finely and continuously distributed β -phase can more effectively stop the development of corrosion in an AZ alloy. Otherwise, the presence of β -phase will accelerate corrosion.

It should be stressed that the distribution of a secondary phase is closely associated with the grain size. Normally, a grain-refined microstructure has a more uniform and continuous secondary phase along the grain boundary. This is clearly demonstrated by the microstructures of diecast AZ91D and MEZ alloys (Figure 9 and Figure 12). Particularly, Figure 12 clearly shows that the continuously distributed secondary phase effectively confines corrosion in the grains. This signifies that grain refinement is an effective approach of improving corrosion resistance of a magnesium alloy.

Galvanic Corrosion

Galvanic corrosion is one of the major obstacles to the use of magnesium parts in the automobile industry [19]. This is because magnesium is the most active metal in the galvanic series [20], and a magnesium alloy component is always an active anode if it is in contact with other metals. Theoretically, galvanic corrosion can be eliminated by insulating or blocking the direct electrical contact between magnesium alloys and other metals. Unfortunately, direct electrical contacts are sometimes required or unavoidable in the practical designs because of mechanical and electrical demands.

In practice, aluminium, steel and galvanised steel are popular materials, and magnesium alloys will unavoidably be in contact with them. In this case, the metal that forms a galvanic couple with a magnesium alloy has a significant influence on galvanic corrosion.

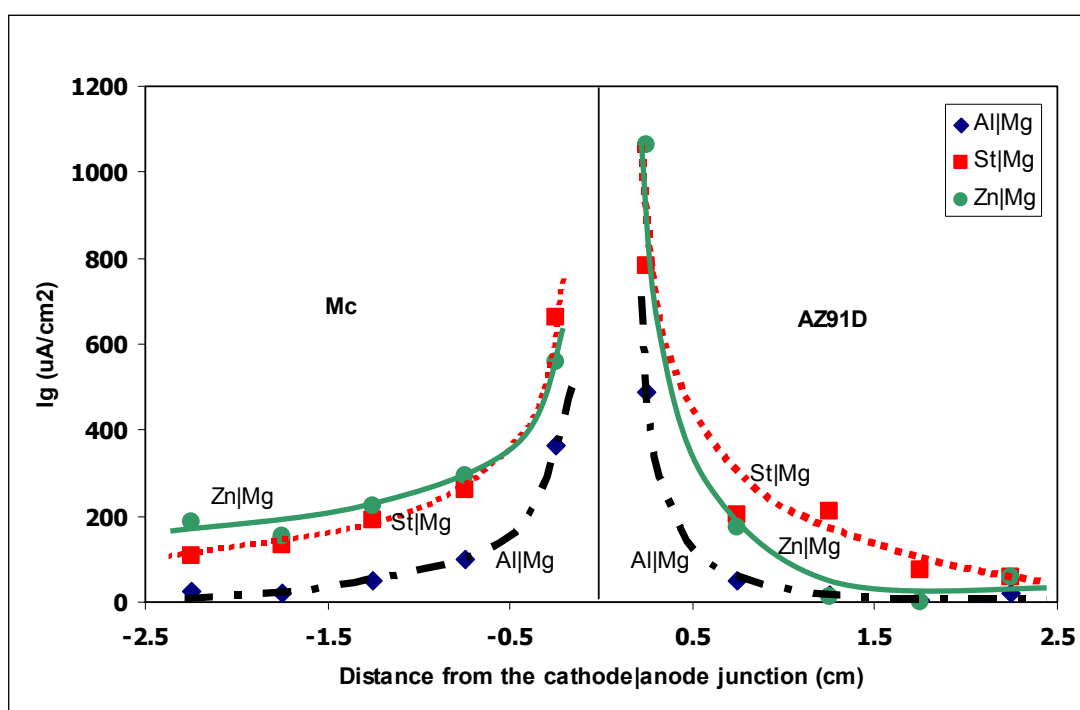


Figure 13. The distribution of galvanic current density on “Al|Mg” , “Steel|Mg” and “Zn|Mg” galvanic couples.

Figure 13 [21] shows typical distributions of the galvanic current density on “Al|Mg”, “Zn|Mg” and “Steel|Mg” galvanic couples. The galvanic current density is the highest when AZ91D, which is the most widely used magnesium alloy, is used in practice, is in contact with steel. The least severe is the galvanic corrosion of AZ91D in contact

with aluminium. The galvanic current densities on the magnesium side and the coupling cathode metal side both decrease with increasing distance from the “anode|cathode” junction. This indicates that galvanic corrosion is more severe on magnesium in the area adjacent to the cathode and the cathode metal is better protected from corrosion attack in the area adjacent to the magnesium anode.

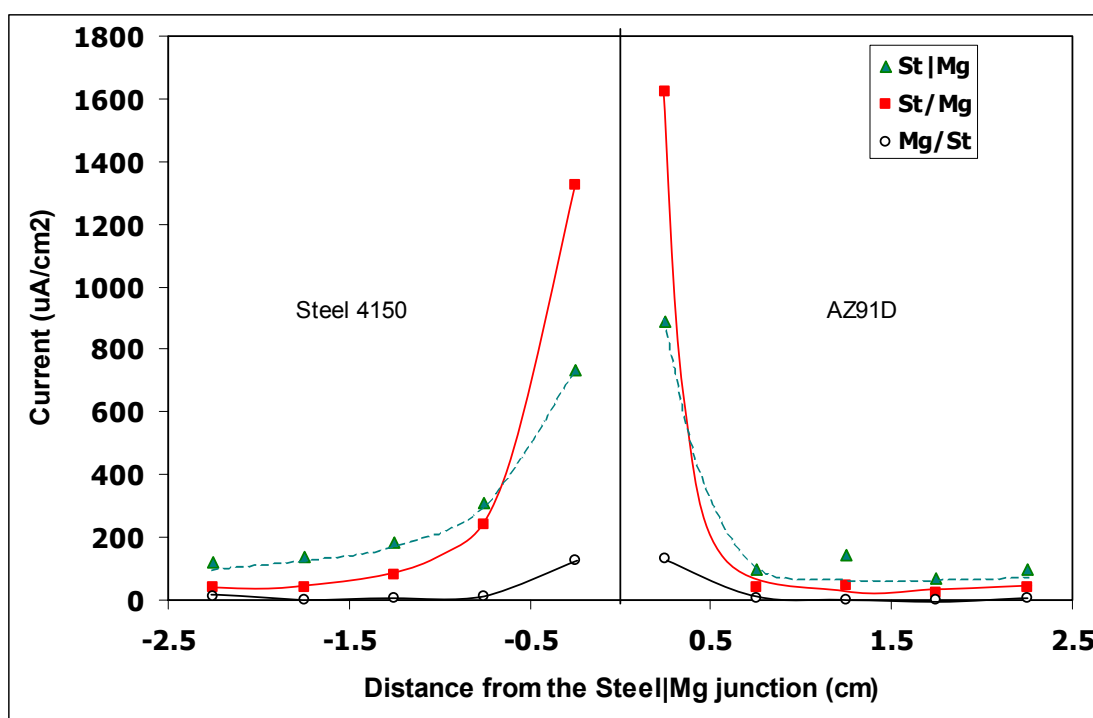


Figure 14. The distributions of galvanic current density of “Mg|Steel” galvanic couple in different exposure configurations: “Steel|Mg” –Steel and Mg are beside each other; “Steel/Mg” –Steel is above the Mg; “Mg/Steel” –Mg is above the Steel.

The relative positions of the anode and cathode of a galvanic couple can also affect galvanic corrosion [21]. Figure 14 shows the distributions of the galvanic current density of a “Steel|Mg” couple in different exposure configurations. The galvanic current densities of this couple in the “Mg/Steel” exposure configuration (Mg above the Steel) are all relatively lower than in the “Steel|Mg” exposure configuration (Steel and Mg beside each other), particularly in the regions adjacent to the Steel–Mg junction. The differences in corrosion caused by the different exposure configurations could be ascribed to the fact that the alkalised solution produced on the magnesium anode surface can easily flow down to the cathode surface

in this exposure configuration. Therefore, the smaller galvanic current densities in this exposure configuration can be attributed to the alkalisied solution on the cathode surface caused by the corrosion products coming from the magnesium surface.

In contrast, galvanic current density of the couple in the “Steel/Mg” (Steel is above the Mg) exposure configuration is higher than in the “Steel|Mg” exposure configuration. This is because the corrosion products from the steel cathode are flushed down to the anode surface, affecting the electrochemical behaviour of the magnesium anode. A possible mechanism involved in this process is that the iron ions from the steel surface deposit on to the magnesium surface, which dramatically increases the impurity level of the magnesium. Hence, the galvanic corrosion rate increases.

Summary

Although corrosion performance is currently a problem for magnesium alloys, the prospect for magnesium alloys is promising because of their attractive advantages and potential applications. Particularly, with the price of magnesium dropping dramatically and approaching that of aluminium recently, magnesium alloys are becoming a commercially attractive light material in many industries. How to solve the corrosion problem has now become an important issue in the application of magnesium alloys.

The investigation by CAST on the corrosion behaviour of magnesium and its alloys has clearly elucidated the following points:

1. Hydrogen evolution always accompanies the anodic dissolution of magnesium alloys and the Negative Difference Effect (NDE) is a common phenomenon for all investigated alloys.
2. In the aluminium containing alloys the β phase appears to have a beneficial effect on the corrosion resistance but it can also accelerate corrosion in some cases. The role of the β -phase depends on its amount and distribution. The beneficial effect of the β -phase on corrosion performance can be enhanced by grain refinement.

3. Alloying aluminium and zirconium can stabilise the matrix phase of a magnesium alloy against corrosion. In addition, zirconium is also an effective purifier for magnesium, and the purification can lead to significantly improved corrosion resistance of magnesium alloys.
4. The galvanic corrosion of a magnesium alloy depends on the cathode material. Steel is the worst coupling metal for magnesium alloys and aluminium is relatively less detrimental. The manner by which the galvanic couples are exposed is also an important factor influencing the severity of galvanic corrosion.

More importantly, these fundamental findings by CAST signify that:

1. For magnesium and its alloys, the corrosion rate of can not be estimated from polarisation curve alone. Electrochemical measurements should be used with caution in study of the corrosion of magnesium and its alloys.
2. Since zirconium can purify a magnesium alloy and the corrosion resistance of the magnesium alloy can be further improved through grain refinement, an approach for improving corrosion resistance could be to first purify the raw magnesium ingots with zirconium and then refine the grain size of the alloy through high pressure die casting, which would produce a skin with fine grains and a large amount of continuously distributed secondary phase on the surface of the casting.
3. In practice, if the electrical contact between a magnesium component and steel is unavoidable in design, then try to avoid the rust from the steel depositing onto the magnesium component.

Acknowledgments

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