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# **Electrochemical Performance of Mg-air Battery Based on AZ61 Magnesium Alloy with Different Ambient Temperature**

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The purpose of this research is to determine the optimal ambient temperature for Mg-air batteries with AZ61 alloy as the anode to achieve, lower corrosion and higher discharge performance. The corrosion behaviors were evaluated by potentiodynamic polarization measurements in a 3.5 wt% NaCl solution and the discharge performance of the AZ61 alloy as an anode for Mg-air batteries was studied. The AZ61 anode has higher utilization and a lower self-corrosion rate at low ambient temperature. With increasing ambient temperature, Mg-air batteries have higher operating voltages and specific energies. Electrochemical impedance spectroscopy(EIS) and scanning electron microscopy(SEM) support the results of electrochemical and discharge performance tests. Combining the corrosion rate and discharge performance, the AZ61 alloy as anode for Mg-air batteries obtained the best electrochemical performance in a 3.5 wt% NaCl electrolyte at an ambient temperature of 40-60°C.

**Keywords:** Mg-air battery; AZ61 anode; Ambient temperature; Electrochemical performance; Discharge

# **1. INTRODUCTION**

Metal-air batteries are compact and potentially less expensive batteries. In addition, metal-air batteries have advantages of abundant raw materials, low cost, high security, and environmental friendliness. [1]. Among these types of Metal-air batteries, Mg-air batteries have the advantages of a theoretical specific capacity, of  $2205\text{mA}\cdot\text{h}\cdot\text{g}^{-1}$  and an energy density, of  $3910\text{kW}\cdot\text{kg}^{-1}$ . In addition, the theoretical voltage (3.1 V) of Mg-air batteries is higher than that of Zn-air (1.65 V) and Al-air (2.71 V) batteries [2-3]. The commonly used material of Mg anode is magnesium plate, but the disadvantage is the high degree of corrosion [4]. Therefore, improving the corrosion resistance of magnesium alloys has become the first research direction of magnesium alloy anode.

In existing studies, much research has been performed on the corrosion resistance and discharge performance of commercial AZ series magnesium alloy anodes. The increase in Al content will reduce

the corrosion sensitivity of Mg alloys and gradually increase the utilization rate of the anode of the Mgair battery [5]. Meanwhile, the number of corrosion products on the surface of AZ31 is the largest, and it decreases significantly with the increase of Al content [6]. Zinc can increase the corrosion resistance potential of Mg alloys [7]. The AZ61 alloy shows higher corrosion resistance and higher discharge potential, which is more suitable for Mg-air battery anodes than other AZ series alloys, including AZ31 and AZ91 [5,8]. Adding other elements to magnesium alloys to change the electrochemical performance of My alloys as anodes for Mg-air batteries is also the main research method, including the addition of Ca, Sm, Ga, La and Y [9-13]. Adding Sm to AZ80 can improve the corrosion resistance and discharge properties [9]. Ca and two types of REs (La and Sm) are used to modify AZ31 alloy and increase the output voltage and discharge capacity of the modified AZ31 alloy as anodes for Mg-air batteries [10].

In addition, ambient factors can also impact the electrochemical performance of magnesium alloys as anodes for Mg-air batteries. For example, the corrosion of magnesium alloy anode in NaCl solution depends with the chlorine ion concentration and pH value of NaCl solution [14-15]. The effects of organic media on magnesium anode properties have also been studied; for example, Genders and Pletcher used a microelectrode to study the basic chemical reaction of Mg<sup>2+</sup>/Mg in tetrahydrofuran and propylene carbonate [16]. Mg water-activated battery performance varies at different temperatures, and the battery voltage at 35°C is significantly higher than that at 0°C at the same current density [17].

In this work, the effects of ambient temperature on AZ61 as anode for Mg-air batteries were investigated. The electrochemical behaviors of the AZ61 magnesium anode were tested at different ambient temperatures. The discharge behavior of AZ61 alloy as an anode for Mg-air batteries was also tested under constant current at different ambient temperatures. To provide an optimal temperature environment for Mg-air batteries in practical applications, this work aims to study the electrochemical behavior of AZ61 anodes by changing the ambient temperature, thereby exploring the best discharge performance of Mg-air batteries.

#### 2. MATERIALS AND METHODS

## 2.1 Magnesium alloy anode and electrolyte

Commercial AZ61 magnesium alloy as the anode was used as the research object, the chemical composition is listed in Table 1. The microstructure of the AZ series Mg alloys is composed of an  $\alpha$ -Mg matrix, Al–Mn and  $\beta$ -Mg17Al12 intermetallics. The corrosion products of black aluminum-rich oxides can be formed in the thin  $\beta$ -Mg17Al12 phase; therefore, the as-cast anode has a lower corrosion rate. [18,19]. The 3.5 wt% NaCl solution used as the Mg-air battery electrolyte. It was made from 99% pure NaCl and deionized water.

**Table 1.** The chemical composition of the Mg anode in mass percent

Al	Zn	Mn	Si	Fe	Mg
5.8	0.82	0.335	0.05	0.005	Bal.

## 2.2 Self-corrosion rate

The weight loss method was applied to measure the self-corrosion rate of AZ61 alloy. AZ61 alloy samples were immersed in 3.5 wt% NaCl solution for 24 hours under different ambient temperatures. The corrosion products of the AZ61 alloy were removed in a 200 g/L CrO3 solution. The calculation formula for the weight loss rate is shown in Eq(1):

$$V = \frac{W_0 - W_t}{S \times t} \tag{1}$$

where V is the corrosion rate $(g \cdot cm^{-2} \cdot h^{-1})$ , W0 is the initial weight of the AZ61 alloy (g), Wt is the weight after corrosion of the AZ61 alloy (g), S is the area of the AZ61 alloy exposed to electrolyte (cm<sup>2</sup>), and t is the soaking time (h).

## 2.3 Electrochemical behavior

Through the use of a classical three-electrode cell, the electrochemical behaviors of the investigated alloys were tested in a ChenHua CHI660E electrochemistry workstation. The working electrode was an AZ61 alloy, the auxiliary electrode was a carbon rod, and the reference electrode was a saturated calomel electrode (SCE). Electrochemical impedance spectroscopy (EIS) was measured with a 5 mV sinusoidal disturbance at an open-circuit potential in different temperature environments. The sweep frequency was 100 kHz to 0.1 Hz. The obtained EIS curves were fitted using ZSimpWin software. With a scan rate of 1 mV·s<sup>-1</sup>, the dynamic potential polarization curve was tested from -2 to -1V. The corrosion potential and corrosion current density of the AZ61 alloy at different temperatures were obtained. To ensure repeatability, each test had was carried out in at least three replicates.

# 2.4 Discharge behavior

The magnesium air battery is composed of an AZ61 magnesium anode, manganese carbon air cathode and 3.5 wt% NaCl electrolyte. The constant current discharge test was conducted at a current density of 10-40 mA·cm<sup>-2</sup> to study the discharge behavior of Mg-air batteries at different ambient temperatures, and the test temperatures were 0, 20, 40, 60, and 80°C. First the battery was placed in the corresponding environment temperature for 30 minutes. Then, a discharge test was performed for 5 hours, the voltage change was recorded with time, and the cutoff voltage was set to 0.5 V. After the discharge of the Mg-air battery, the surface of the AZ61 alloy anode was cleaned with 200 g/L CrO3 to observe the corrosion of the magnesium alloy pattern, and then the mass loss method was used to calculate the discharge capacity and the anodic efficiency, as shown in Eq (2) and Eq(3):

Dischange capacity = 
$$\frac{i \cdot A \cdot t}{W_i - W_f}$$
 (2)  
Anode efficiency(%) =  $\frac{(i \cdot A \cdot t)M}{2F(W_i - W_f)}$  (3)

where i is the current density  $(mA \cdot cm^{-2})$ , t is the discharge time (h) of the discharge test, F is Faraday's constant (96485 C·mol<sup>-1</sup>), A is the surface area (cm<sup>2</sup>), M is the atomic mass (g·mol<sup>-1</sup>) of the

specimens, and  $W_i$  and  $W_f$  are the weights of the specimens before and after the discharge test, respectively[18].

## **3. RESULTS AND DISCUSSION**

#### 3.1 Self-corrosion

Table 2 lists the self-corrosion of the AZ61 alloy in a 3.5wt% NaCl solution at different ambient temperatures. With increasing the ambient temperature, the self-corrosion rate of the AZ61 alloy in 3.5 wt% NaCl solution increased significantly. The self-corrosion rates of the AZ61 alloy at ambient temperature of 40°C and 60°C are relatively close; however, when the ambient temperature reaches 80°C, the self-corrosion rate of the AZ61 alloy is approximately 1.9 times that at 60°C. Therefore, when the Mg-air battery is working, the ambient temperature should not be too high.

**Table 2.** Self-corrosion of the commercial AZ61 alloy for 24 h in 3.5 wt% NaCl solution at different ambient temperatures

Ambient temperature/°C	0	20	40	60	80
Self-corrosion rate / $g \cdot cm^{-2} \cdot h^{-1}$	0.09	0.23	0.36	0.43	0.82

#### 3.2 Electrochemical performance

Figure 1 shows the polarization curves of AZ61 alloys in 3.5 wt% NaCl electrolyte at different ambient temperatures. Table 3 lists the parameters calculated by the Tafel linear extrapolation. In Figure 1, the current density of the AZ61 magnesium alloy in the anode and cathode potential regions at an ambient temperature of 0°C is lower than those at other ambient temperatures. Under the corrosion potential (no discharge current), the current density of the AZ61 alloy increases with temperature, and the potential shifts negatively, indicating that the degree of activation of the AZ61 magnesium alloy increases with temperature. In Table 3, the AZ61 alloy at an ambient temperature of 0 °C has the lowest corrosion current density ( $J_{corr}$ ) and the highest polarization resistance (Rp) compared with the other ambient temperatures. As the ambient temperature increases, the AZ61 alloy has a higher corrosion rate in the 3.5 wt% NaCl electrolyte. Compared with the test results at room temperature in other studies, the AZ61 alloy has better corrosion resistance at low temperatures, but has a higher corrosion rate at high temperatures [4].



**Figure 1.** Anodic polarization curves of the commercial AZ61 alloy with different ambient temperatures in 3.5 wt% NaCl solution

**Table 3.** φcorr, Jcorr and Rp values of the commercial AZ61 alloys derived from polarization curves with different ambient temperatures in 3.5 wt% NaCl solution

Ambient	(Deerr (VS SCE)/ V	$I_{aorr}/\mu A \cdot cm^{-2}$	$Rn/Q \cdot cm^{-2}$
temperature			
0°C	-1.525	3.489	4386.5
20°C	-1.520	129.9	182.6
40°C	-1.553	319.3	76.2
60°C	-1.564	607.4	51.3
80°C	-1.609	874.9	35.3

In the high temperature environment, the AZ61 alloy has a higher self-corrosion current density. The higher the self-corrosion current density is, the worse the corrosion resistance of the AZ61 anode, but it is more conducive to the rapid dissolution of the magnesium anode under corrosion potential and shortens its activation time during use.

Figure 2a shows the Nyquist plots of the AZ61 alloy in 3.5 wt% NaCl solution with different ambient temperatures. Five tests were conducted, and the shape of the EIS plots was similar for all samples, indicating similar corrosion behavior of the AZ61 alloy in the 3.5 wt% NaCl solution with different ambient temperatures. The EIS fitting result is presented by the equivalent circuit in Fig 2b, including  $R_s$ ,  $R_t$ , CPE, L and  $R_l$ .  $R_s$  is the solution resistance;  $R_t$  is the charge transfer resistance and CPE is the double-layer capacitance. L is the corresponding inductance and  $R_l$  is mass transport resistance in the surface corrosion product layer. General speaking, a capacitive semicircle is attributed to the Mg-Mg<sup>2+</sup> reaction at the anode/electrolyte interface and charge transfer at high frequency [20,21]. The low-frequency inductive arc is caused by the deposition of corrosion products [22].

According to the equivalent circuit in Fig. 3b, table 4 shows the electrochemical parameters obtained by using the ZSimpWin software. The Rt of the AZ61 alloy at ambient temperature of 0°C, 20°C, 40°C, 60°C, and 80°C decreased sequentially. This indicates that the self-corrosion rate of the AZ61 alloy at different ambient temperatures increases in the following order:  $0^{\circ}C < 20^{\circ}C < 40^{\circ}C < 60^{\circ}C < 80^{\circ}C$ . Meanwhile, the smaller the value of R<sub>1</sub> is, the fewer corrosion products there are on the surface of the tested alloy, and the lower the anode utilization rate may be.



Figure 2. EIS of the commercial AZ61 alloy in 3.5 wt % NaCl electrolyte with different ambient temperatures: (a) Nyquist plots and (b) equivalent circuits used for fitting the EIS  $R_s(Q(R_t(L_1R_1)))$ .

**Table 4.** EIS simulated values of the commercial AZ61 alloy in the 3.5 wt% NaCl solution at different ambient temperatures

Ambient	റംറ	20°C	40°C	60°C	80°C
temperature	0 C	20 C	40 C	00 C	80 C
$R_s$ , $\Omega cm^2$	8.243	7.069	5.405	4.125	3.807
CPE, Fcm <sup>-2</sup>	9.788×10 <sup>-6</sup>	12.96×10 <sup>-6</sup>	15.69×10 <sup>-6</sup>	37.71×10 <sup>-6</sup>	21.26×10 <sup>-6</sup>
n (0 <n<1)< td=""><td>0.8752</td><td>0.8728</td><td>0.9717</td><td>0.9555</td><td>0.9598</td></n<1)<>	0.8752	0.8728	0.9717	0.9555	0.9598
$R_t, \Omega cm^2$	1943	366.4	8.18	20.44	13.33
L, Hcm <sup>2</sup>	2.373	331.9	4.549	1.677	1.497
$R_1$ , $\Omega cm^2$	$6.677 \times 10^9$	1062	37.36	16.89	13.71

## 3.3 Discharge performance

Figure 3 shows that the discharge behavior of the Mg-air batteries based on the commercial AZ61 alloys was measured at 10 mA·cm<sup>-2</sup>, 20 mA·cm<sup>-2</sup>, 30 mA·cm<sup>-2</sup>, and 40 mA·cm<sup>-2</sup> current densities for 5 h at different ambient temperatures. Table 5 summarizes the properties of these batteries.

For batteries at the same discharge current density, the operating voltage increases first and then decreases when the ambient temperature increases from 0 to 80°C. When the ambient temperature is 60°C, the operating voltage of the batteries reaches the maximum. The operating voltages are 1.2180, 1.0825, 0.9641 and 0.9033 V with discharge current densities of 10, 20, 30 and 40 mA·cm<sup>-2</sup> at this temperature, and the operating voltage of the Mg-air battery with the AZ61 alloy as anode is higher than the operating voltages of Mg-air batteries for other AZ series alloys as anodes at room temperature described in other papers [17,23].Meanwhile, the operating voltage decreases as the current density increases from 10 to 40 mA·cm<sup>-2</sup> for the batteries, at the same ambient temperature.



**Figure 3.** Discharge curves of Mg–air batteries for the commercial AZ61 with different ambient temperatures in 3.5 wt% NaCl solution: (a) 10 mA cm<sup>-2</sup>, (b) 20 mA cm<sup>-2</sup>, (c) 30 mA cm<sup>-2</sup>, and (d) 40 mA cm<sup>-2</sup>

Self-corrosion rate of AZ61 alloys is lower and the production of the self-corrosion of AZ61 alloy is higher at low temperature environment, and thus, it has a high utilization rate as a Mg-air battery anode. With increasing temperatur, the self-corrosion rate increases and the anode utilization rate decreases. However, as the ambient temperature increases, it brings more favorable effects. First, compared to the discharge performance in a low temperature environment, it has a more stable working voltage platform, for example, the working voltage fluctuates greatly at 0°C. Second, it has a longer discharge time. Mg-air batteries can maintain good discharge performance for a long time under different discharge current densities in high temperature environments, but in low temperature environments, especially 0°C, they usually cannot complete the set discharge time before reaching the cutoff voltage.

The electrode process of the Mg-air battery is mainly controlled by activation. With increasing temperature, the dissolution rate and charge transfer rate of the magnesium anode increase, and thus, it shows strong discharge activity and the discharge voltage increases. Meanwhile, the corrosion rate of the AZ61 alloy increases in a high temperature environment, and the anode efficiency of the AZ61 alloy as a Mg-air battery anode in a high temperature environment is significantly lower than that in a low temperature environment.

Current density/	Ambient	Operating	Anode utilization
mA·cm <sup>−2</sup>	temperature/°C	voltage/V	/%
	0	1.0630	45
	20	1.1182	43
10	40	1.2056	38
	60	1.2180	39
	80	1.2093	38
	0	0.9213	50
	20	1.0150	48
20	40	1.0577	45
	60	1.0825	45
	80	1.0196	40
	0	0.8711	50
	20	0.9136	48
30	40	1.0233	47
	60	0.9641	44
	80	0.9632	40
	0	0.7533	53
	20	0.8184	49
40	40	0.8435	48
	60	0.9033	44
	80	0.8860	40

**Table 5.** Discharge performance of Mg-air batteries (discharge current of 10mAcm<sup>-2</sup>, 20mAcm<sup>-2</sup>, 30mAcm<sup>-2</sup>and 40mAcm<sup>-2</sup>) for the commercial AZ61 alloys as anodes in the 3.5wt% NaCl solutions with different ambient temperatures

The specific energy of a Mg-air battery with AZ61 alloy as the anode at different ambient temperatures is shown in Figure 4. In the case of constant ambient temperature, the specific energy

decreases with the increasing of current density. At a certain current density, the specific energy increases with the temperature increases, and it reaches the a maximum at 40°C. When the temperature continues to rise to 80°C, the specific energy decreases significantly. At an ambient temperature of 0°C, when the batteries are discharged with a high current density, the low temperature environment reduces the activation efficiency of the AZ61 and the ion transmission rate of the battery system; thus, the specific energy with increasing temperature may be because the increase in ambient temperature promotes the efficiency of the conversion of chemical energy to electrical energy. The specific energy reached a maximum at 40°C and then decreased, which may be due to the increased self-corrosion rate of the AZ61 alloy anode at high temperature and the significant decrease in anode utilization.



**Figure 4.** Specific energy of Mg–air batteries (discharge currents of 10mAcm<sup>-2</sup>, 20mAcm<sup>-2</sup>, 30mAcm<sup>-2</sup>, and 40mAcm<sup>-2</sup>) for the commercial AZ61 alloy as an anode with different ambient temperatures in 3.5 wt% NaCl solution

## 3.4 Surface morphologies after discharge

Figure 4 shows the surface morphologies of the AZ61 anode after 5 h of discharge at 30 mA·cm<sup>-2</sup> in 3.5 wt % NaCl electrolyte at different ambient temperatures with removal of the discharge products. After discharge the surface of the AZ61 alloy at ambient temperatures of 0°C and 20°C is relatively flat, and there are few large and obvious corrosion pits, as shown in fig.5a and 5b. Corrosion pits can be observed on the surface of the AZ61 alloy after discharge in an ambient temperature of 40°C, and the surface is no longer smooth, as shown in Fig.5c. At ambient temperatures of 60°C and 80°C, a large number of obvious corrosion pits appeared on the surface of the AZ61 alloy after discharge, as shown

in Fig.5d and 5e. In particular the corrosion pits appearing on the surface of the AZ61 alloy after discharge at an ambient temperature of 80°C are large and deep.

The Mg-air battery is accompanied by self-corrosion during the discharge process, and thus, the consumption of the AZ61 anode does not only come from battery discharge. The AZ61 anode's self-corrosion rate is lower, and the product of self-corrosion is easier to deposit on the surface of AZ61 anode in a low-temperature environment. There are no obvious corrosion pits and the anode surface is relatively smooth after discharge at an ambient temperatures of 0°C and 20°C, which explains why the anode efficiency of the Mg-air battery in a low-temperature environment is higher. In a high-temperature environment, the solution evaporates faster, and the chloride ion concentration increases, which also promotes the corrosion of AZ61 alloys [24]. The AZ61 alloy has obvious corrosion pits after discharge at an ambient temperature environment, the Mg-air battery obtains a smaller anode utilization rate due to self-corrosion during the discharge process.



**Figure 5.** Surface morphologies of the AZ61 anode after 5 h of discharge at 30 mA·cm<sup>-2</sup> in 3.5 wt % NaCl electrolyte at different ambient temperatures with removal of the discharge products: ambient temperatures of (a) 0°C, (b) 20°C, (c) 40°C, (d) 60°C and (e) 80°C

# 4. CONCLUSIONS

In this work, the effect of the ambient temperature on the electrochemical performance of AZ61 alloys as anodes for Mg-air batteries was investigated. The conclusions are as follows.

The self-corrosion rate of the anode in the low temperature environment becomes lower, which makes the Mg-air battery have a higher anode utilization rate in the low-temperature environment. Mg-air batteries are accompanied by stronger discharge activity with increasing ambient temperature and have a higher discharge voltage in a high-temperature environment. The discharge voltage reaches the maximum at 60°C, and the operating voltages are 1.2180, 1.0577, 0.9641 and 0.9033 V at the a current density discharge of 10 to 40 mA  $\cdot$  cm<sup>-2</sup>. After comprehensive analysis, the Mg-air battery can obtain the best electrochemical performance in the ambient temperature range from 40°C to 60°C. This has guiding significance for the Mg-air batteries' practical application.

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