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Effect of Stress and Galvanic Factors on the Corrosion Behave of Aluminum Alloy

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> **Abstract:** The interaction between stress and galvanic during the corrosion process of 5050 aluminum alloys was studied and the evolution of mechanics properties was indicated. Electrochemical impedance technique and scanning electron microscopy were used to analyze the surface electrochemical states and the corrosion morphology. At the same time, corrosion kinetics and thermodynamic theory were used to analyze the influencing mechanism of the stress factor and the galvanic factor. The results show that both of galvanic factor and tensile stress factor can increase the corrosion potential of aluminum alloys and result in the corrosion resistance decrease. With decreasing corrosion resistance, the mechanical properties of aluminum alloys decrease. These phenomena are attributed to two reasons: One is that aluminum alloys may under the condition of anodic polarization after coupled with 40CrNiMoA steel, so the corrosion can be promoted; The other is that with increasing stress, the electrochemical potential of 5050 aluminum alloys decreases and the potential difference between two materials increases, so the corrosion becomes serious. Compared to the stress factor, the galvanic factor is significant.

> Key words: aluminum alloy; stress; galvanic; mechanical-electrochemistry; electrochemical impedances spectroscopy

1 Introduction

For the present manufacturing industry, to lighten the weight becomes the mainstream philosophy of design. Especially in aerospace and automobile manufacturing industries, lightweight turns more and more important. Due to its high strength, low density and excellent machining performance, *etc*, aluminum alloys are widely used in industrial areas in the recent decades. Usually, aluminum alloys have high corrosion resistance in a wide environment due to the density passive film on the surface. However, in the corrosion ion containing environment, aluminum alloys may suffer various corrosions, such as stress corrosion, galvanic corrosion, $etc^{[1,2]}$. In some serious cases, it may arouse the components fracture^[3]. In recent years, many researchers pay attention to stress corrosion and galvanic corrosion of aluminum. Song R[4] *et al* studied the stress corrosion of aluminum and pointed out that the SCC susceptibility increases with increasing Mg segregation at the grain boundaries and hydrogen embrittlement (HE) increases with increasing hydrogen charging and decreases with increasing ageing time under the same hydrogen charging conditions. G Van Boven^[5] *et al* reported the influence of residual stress in the process of stress corrosion and indicated that micropitting occurs preferentially in areas where the tensile residual stresses are the highest, while SCC initiation occurs with a 71% normalized frequency in areas where the surface residual stress is in the range of 150-200 MPa . M Sakairi^[6] researched the galvanic corrosion behavior of AA5052 aluminum alloy in low chloride ion concentration solution by electrochemical noise impedance, and the results indicated that aluminum alloys tend to undergo galvanic corrosion because of low corrosion potential when contact with other alloys even in the low chloride ion concentration solution. However, there are few researches concentrated on the interaction between stress factor and galvanic factor to corrosion behavior of aluminum alloy and analyze the mechanical properties degradation rule^[7-10].

Steel or titanium alloy fasteners used to connect aluminum alloys in aircraft structure are widespread phenomenon and these structures are usually used to

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important structure bearing stress. When the warplanes serve in coastal areas or on the aircraft carrier, the fasteners corrosion under the connection of stress factor and galvanic factor has a significant impact on the service life and safety of warplane. So, to study the corrosion behavior and evolution of mechanical properties under the connection of stress factor and galvanic factor is necessary.

Due to those backgrounds, this article chooses 5050 aluminum alloy as the study object which has the good forming property and high corrosion resistance. Simulating the condition of 5050 aluminum alloy contacted with 40CrNiMoA steel, it suffers stress in the sea environments. The corrosion behaves and the degradation of mechanical properties are studied. EIS technology and surface morphology are used to analyze the internal mechanism.

2 Experimental

2.1 Materials and samples

 Materials investigated in this study included 5050 aluminum alloys (AA5050) as well as 40CrNiMoA steels. The chemical composition of 5050 aluminum alloys was 1.4%Mg, 0.2%Cu, 0.1%Mn, 0.1%Cr, 0.7%Si, 0.4%Ti, Al Bal (all in wt%) and the composition of 40CrNiMoA steel was 0.44%C, 0.8%Mn, 0.40 %S, 0.90% Cr, 1.75% Ni, 0.25 %Mo, Fe Bal (all in wt%). Mechanical properties of 5050 aluminum alloy are as follows: yield strength 149.6 MPa; tensile strength 199.9 MPa; elongation 21.5%; reduction of area 27.0%. Fig.1 shows the shape and size of galvanic corrosion sample.

Fig.1 The test specimen's geometry (both aluminum alloys and steel were the same)

2.2 Test method

The AA5050 and 40CrNiMoA steel made up a galvanic couple. Before test, all materials were polished by diamond paper from 240# to 1000# step by step, and then ultrasonically cleaned in alcohol and washed with deionized water, finally dried in cool air. In the experiment, solution selected was 3.5 wt% NaCl and test temperature was 30±1℃. The distance between aluminum alloy and steel was 5 mm and the exposed area was 1 mm² and the other area was sealed with sealant. The schematic outline is shown in Fig 2.

In order to study the influence of stress factor during galvanic corrosion process of AA5050, 40CoNiMoA steel was used as the coupled material. Four levels tensile stresses were imposed on the aluminum alloys. The test was lasting for 20 h. The potential of the uncoupled materials was measured and the couple current during 20 h was recorded, then the average current density was calculated. Electrode potential before corrosion was measured after the electrode was immersed in the solution for 30 minutes when no coupled and electrode potential after corrosion was measured after the circuit disconnected 30 minutes when the test was finished.

In order to study the influence of galvanic factor during stress corrosion process of AA5050, 40CoNiMoA steel was used as the coupled material, four levels tensile stresses were imposed on the aluminum alloys during the galvanic corrosion. The test was lasting for 9 days then measuring the mechanical property of aluminum (five conditions were chosen: No coupled-not coupled 40CrNiMoA while no stress; Coupled-coupled 40CrNiMoA while no stress; $25\%R_{\text{p0.2Al}}$ -coupled 40CrNiMoA while suffer $25\%R_{\text{p0.2Al}}$ tensile stress; 50%R_{p0.2Al}-coupled 40CrNiMoA while suffer $25\%R_{p0.2A1}$ tensile stress; $75\%R_{p0.2A1}$ -coupled 40CrNiMoA while suffer $25\%R_{n0.2A1}$ tensile stress). To compare the change of mechanical property along with the corrosion time, three corrosion time $(3, 9, 9, 1)$ 18 days) was chosen and the tensile stress level was $75\%R_{n0.2\text{Al}}$. After the tests, the mechanical property of aluminum alloy was measured.

The electrochemical impedance spectroscopy (EIS) was performed on parstat2273 electrochemical workstation. Choosing three electrode systems, working electrodes was aluminum alloy, auxiliary electrodes

Tensile stress	Electrode potential before corrosion/mV		Electrode potential after corrosion/mV		Average current density
	Aluminum alloy	Steel	Aluminum alloy	Steel	/ μ A/cm ²
No stress	-775	-653	-786	-644	31.70
$25\%R_{n}0.2Al$	-890	-654	-895	-630	43.95
$50\%R_{n}$ 0.2Al	-912	-639	-874	-631	50.86
$75\%R_{\infty}0.2Al$	-923	-636	-847	-635	58.16

Table 1 Galvanic corrosion potentials and currents density of AA5050/40CrNiMoA

was platinum, and reference electrodes was saturated calomel electrode (SCE). The excitation signal was 10 mV and frequency range was 10^5 - 10^{-2} Hz. Zsimpwin was used to analyze the EIS.

 After corrosion, the surface morphologies were analyzed by SEM. The number of pitting and pitting area were analyzed by image-pro-plus and the depth of pitting was measured by a micrometer gauge.

3 Results and discussion

3.1 Influence of mechanical factor for galvanic corrosion

In this test, the 5050 aluminum alloys suffered different stresses while coupled with 40CrNiMoA steels in 3.5 wt% NaCl solution at 30 ℃ for 20 h. The result is shown in Table 1. Comparing the electrode potentials before corrosion, the potential difference between aluminum alloys and steels was large no matter aluminum alloys suffered stress or not. So the AA5050 had high galvanic corrosion activity when coupled with 40CrNiMoA steel. The electrode potential of AA5050 gradually decreased with increasing tensile stress. Stress led to the activation of AA5050 and increased the galvanic corrosion driving force. Compared the average current density, it was the same as the potential change that the larger of the stress was, the bigger of the current density was. Stress promoted the galvanic corrosion.

3.2 Influence of galvanic factor for stress corrosion

In this test, AA5050 suffered different stresses while coupled with 40CrNiMoA steels in 3.5 wt% NaCl solution at 30 ℃ for 9 days. The mechanical properties of AA5050 after corrosion are shown in Table 2. Obviously, galvanic corrosion resistance

was significantly affected by the tensile stress and further impacted the mechanical property. Comparing the no stress in no coupled condition, all mechanical properties of AA5050 dropped in other conditions. When coupled with steel, the mechanical properties of AA5050, especially the elongation, gradually reduced with increasing stress. Plasticity losing was greater than intensity losing. It was indicated that galvanic factor influenced the stress corrosion process of aluminum alloy.

Table 2 Samples coupled while under different stress after corrosion 9 days

Sample	$R_{n0.2}$ /MPa	$R_{\rm m}$ /MPa	$A/\sqrt{2}$
No coupled	148	168	17
Coupled	136	163	14
$25\%R_{\rm p0.2Al}$	135	159	12
50% $R_{p0.2Al}$	128	156	11
$75\%R_{\rm p0.2Al}$	12.1	150	9

The surface morphologies of aluminum alloys after corrosion 9 days are shown in Fig.3 and Fig.4 providing the 3D morphologies of pits. Comparing Figs. $3(a)$, $3(b)$, $3(c)$, $3(d)$, $3(e)$, it could be found that there were a lot of shallow and small pits on the surface in the condition of no stress and no coupled $(Fig.3(a))$; after coupled with steel, the small pits grew deeper and larger, and then the small pits grew to the big pits. There were many cracks surrounding the pits and became more and more with increasing stress $[11]$. It was indicated that the pits would develop horizontally and lengthways and resulted in the surface damage, but the pits would not generate at other place. The change of mechanical properties was in the same rule of the change of pit area and depth. The larger pitting area and the deeper pitting depth were, the greater the mechanical properties lost.

Item	No coupled	Coupled	$25\%R_{\rm p0.2Al}$	$50\%R_{\text{p0.2Al}}$	75% $R_{\rm p0.2Al}$
Average pitting depth/µm	23.2	56.9	95.9	111.8	136.7
Pitting number	28				24
Pitting area/%	6.667	16.95	20.58	21.43	22.67

Table 3 Average depth and density of pitting under different stress

Fig.3 Surface morphology after corrosion 9 d: (a) No coupled; (b) Coupled; (c) $25\%R_{p0.2Al}$; (d) $50\%R_{p0.2Al}$; (e) $75\%R_{p0.2Al}$

Fig.4 3D morphology of pit after corrosion 9 d: (a) No coupled; (b) Coupled; (c) $25\%R_{p0.2\text{Al}}$; (d) $50\%R_{p0.2\text{Al}}$; (e) $75\%R_{p0.2\text{Al}}$

Table 3 provides the average depth, pitting number and pitting area of AA5050 under different stresses. As shown in Table 3, the pitting area became larger and larger, at the same time the pitting size was also changed. The number of pits increased at first and decreased later with increasing stress. Also, the small pits mixed together and grew up into the big pits. Combining the mechanical properties, the morphology and the depth and number of pits, it was obvious that the change of mechanical properties was affected by corrosion behavior, the bigger pitting area and the deeper pitting depth were, the lower the mechanical properties were.

3.3 Influence of corrosion time

Table 4 Samples coupled under $75\%R_{p0.2\text{Al}}$ **after corrosion for different time**

Time/day	$R_{n0.2}$ /MPa	R/MPa	$A/\%$
	140	166	19
9	121	150	9
18	118	146	

Table 4 provides the mechanical properties of AA5050 after corrosion for different time. Under the same stress conditions, the mechanical properties decreased with the extension of corrosion time. Mechanical properties lost quickly at first, especially the elongation and it decreased by 42.1% after corrosion for 9 days than corrosion for 3 days. Then

Fig.5 Surface morphology after corrosion for different time of 75%*R*_{p0.2Al} while coupled: (a) 3 days; (b) 9 days; (c) 18 days

Fig.6 3D morphology of pit after corrosion for different time of 75% R_{m2A} while coupled: (a) 3 days; (b) 9 days; (c) 18 days

the lost rate gradually slowed down, and the elongation after corrosion for 18 days only decreased by 9.1% than corrosion for 9 days.

The surface morphologies of AA5050 after coupled with steel and under the stress of $75\%R_{p0.2A}$ are shown in Fig.5 and Fig.6, providing the 3D morphologies of pits. Obviously corrosion time was a major influence factor. With the extension of corrosion time, the pitting area became larger and many cracks occurred around the pits. The surface of AA5050 desquamated under the influence of stress and corrosion. Also there were not new pits occurred but only the original pits developed.

Table 5 Average pitting depth and density after different time

Time/day			18
Average pitting depth/ μ m	35.73	136.7	183.2
Pitting number	21	24	18
Pitting area $\frac{9}{6}$	11.95	22.67	56.3

Table 5 lists the average pitting depth, pitting number and pitting area of AA5050 after corrosion. Average pitting depth and pitting area increased with the extending of corrosion. But the pitting number increased at first and decreased later. It is indicated that the pits grew up and were combined. The change of mechanical properties and corrosion had relationships and the more serious corrosion were, the lower mechanical properties were.

3.4 Electrochemical test

The Bode and Nyquist plots of AA5050 under different stresses after corrosion for 9 days are shown in Fig.7. From the bode plots in Fig.7(b), it could be seen that there were two time constants in the no stress conditions and three in the suffered stress conditions. No stress samples' Nyquist plot composed by two capacitive reactance arcs (Fig. $7(a)$): the first one appeared at high frequency represents the double-layer capacitance and charge transfer resistance between alloy and solution. And the second one appeared at low frequency corresponded to the corrosion reaction of AA5050 in the corrosion medium^[12]. For the suffered stress conditions, three capacitance arcs and one inductance arc could be seen in the Nyquist plot in Fig.7(a): the first one appeared at high frequency represented the double-layer capacitance and charge transfer resistance between alloy and solution. Meanwhile, the second one appeared at middle-low frequency corresponded to the corrosion reaction of passive films on the AA5050 in the corrosion medium^[13]. The inductance arcs appeared at low frequency represented the pitting process caused by the fresh substrate exposed in the solution which was broken by mechanical $[14]$. And the last capacitance arcs appeared at low frequency represented other areas of pits which were covered by corrosion products.

Comparing those curves, it could be found that impedance was very large in the no coupled and no stress conditions, so the corrosion resistance was high.

Fig.7 EIS plots under different conditions after corrosion for 9 day: (a) Nyquist;(b) Bode

Fig.8 Equivalent circuit: (a) no stress; (b) under stress

When coupled with steel, the impedance was only a small percentage compared with no coupled conditions, obviously the adding galvanic factors significantly affected the corrosion resistance of AA5050. The impedances were further decreased with increasing stress and the higher the stress was the lower the impedance was.

By using the equivalent circuit in Fig.8, the experi-mental data could be well fitted for all states. Fig.8(a) provides the impedance equivalent circuit under no stress conditions, in which R_s represents the solution resistance and Q_{d} represents the double-layer capacitance, while R_{d} indicates the charge transfer resistance between the alloy and the solution. Q_c and R_c correspond to the corrosion reaction of AA5050 in the solution. Fig.8(b) provides the impedance equivalent circuit of the sample suffered stress conditions. In Fig.8(b), R_s represents the solution resistance and Q_{dl} represents the double-layer capacitance, while $R_{\rm dl}$ indicates the charge transfer resistance between the alloy and the solution. Q_c and R_c correspond to the corrosion reaction of passive film in the solution. C_{p1} and R_{p1} correspond to the corrosion reaction of pitting covered by the product in solution. L_{p2} and R_{p2} correspond to the corrosion reaction of fresh substrate at the pitting bottom in the solution.

According to the electrochemical theory, the surface capacitance Q_c (Q_c was C_c in this text) could be

used to characterize the thickness of the product films on the surface:

$$
d_{\text{ox}} = \frac{\varepsilon_0 \varepsilon A}{C_{\text{ox}}} \tag{1}
$$

where
$$
C_{ox} = \frac{(Q_c \times R_c)^{\frac{1}{n}}}{R_c}
$$
, ε_0 is the permittivity of

vacuum, *ε* is the actual permittivity and *A* is the area of sample, d_{ox} is the thickness of product films. As shown in Eq.(1), it is inversely proportional between the thickness of product films and the surface capacitance^[15]. Measuring the EIS of AA5050 after corrosion for 1, 3 and 9 days gives the fitting values of Q_c . According to Eq.(1), the thickness of product films could be obtained. Fig.9 provides the fitting value of *Q*c. By the results, the surface impedance decreased and

Fig.9 *Q*ox changes along with the corrosion time and stress

the capacitance increased (with increasing stress level) or the extension of corrosion time would all deal to the product films thickness decreased.

3.5 Discussion

In the elastic range, it is assumed that the elastic deformation variable is *ε* and the relevant strain energy is *U*. Under stress, the corrosion rate of metals is $[16]$:

$$
i = i^{0} \left(\exp\left(\frac{\beta nF(\varphi_{e} - \varphi_{e,l})}{RT}\right) \cdot \exp\left(\frac{\beta M\sigma^{2}}{2 y \rho RT}\right) - \exp\left(\frac{\alpha nF(\varphi_{e,l} - \varphi_{e})}{RT}\right) \cdot \exp\left(-\frac{\alpha M\sigma^{2}}{2 y \rho RT}\right) \right)
$$

where *M* is the molar atomic weight and ρ is the density of the metal and σ is the tensile stress, whereas $\varphi_e - \varphi_{e,l}$ is the over potential of the metal and the $\varphi_e \cdot \varphi_e$ > 0 for aluminum alloys. Obviously, two factors can increase corrosion, one is increasing the over potential φ_e - φ_e , and the other is raising stress *σ*.

For galvanic factor, the main effect is increasing the over potential of aluminum alloys and enhancing the speed of electrode reaction. Under normal conditions, like the passive metal, there is a dense passive film on the surface. In the no Cl^- containing solution, the passive film has good protective function for metal. But, in the Cl^- containing solution, as the hydrolysis of passive film, the corrosion is severe. The surface energy spectrum of aluminum alloy after

galvanic corrosion is shown in Fig.10 and the process of corrosion is shown in Fig.11.

Eq.(1) and Eq.(3) are the main processes of the dissolution and production of products^[17]. Under the effect of Cl^- , the passive film is damaged and decomposed $(Eq. (4))$. In the corrosion process, $Cl^$ plays a role as the catalyst $(Eq.(5))$. So, the presence of Cl⁻ seriously affects the stability of the passive film and decreases the aluminum alloy corrosion resistance. When the passive film damage rate and generation rate are equal, the corrosion process is stabilized. As for the galvanic factor, it causes a strong polarization to aluminum alloy and the process of Eq.(1) is accelerated. So the dissolution of aluminum alloy increases and the corrosion becomes serious $^{[18]}$:

$$
Al-3e^{-}\rightarrow Al^{3+}
$$

\n
$$
O_{2}+2H_{2}O+4e^{-}\rightarrow 4OH^{-}
$$

\n
$$
Al^{3+}+3OH^{-}\rightarrow Al(OH)_{3}
$$

\n
$$
Al(OH)_{3}+3Cl^{-}\rightarrow AlCl_{3}+3OH^{-}
$$

\n
$$
AlCl_{3}+3H_{2}O\rightarrow Al(OH)_{3}+3H+43Cl^{-}
$$
 (3)

As for the stress factor, the main influence is two aspects. One is the destroy of the surface: Under normal conditions, the passive film is uniform on the surface, but due to the mechanical force, the passive film cracks in some weak positions and the fresh matrix may be exposed in solution (Fig.8). Meanwhile in most

Fig.10 Surface energy spectrum of 5050 aluminum alloy after corrosion

Fig.11 Corrosion process of aluminum alloys

other positions, the surface is not broken. Because of a high potential of passive film and the low potential of matrix, the corrosion of matrix is accelerated and the process of the stress corrosion is promoted. The other is the mechanical-electrochemical interactions: Stress can break the chemical bonds of aluminum alloys and activate chemical reaction. It can also improve the activity of the electrodes (Table 1). The relationship between the decrease of the active energy of chemical reaction and the tensile stress is linear. The rate of mechanics promoting chemical reaction depends on the change of chemical. For the solid materials, the relationship between chemical potential changes and tensile stress is:

$$
\Delta \mu = \int_{\sigma_1}^{\sigma_2} V(\sigma) d\sigma = V_0 \sigma \tag{3}
$$

When $\Delta \mu = 0$, there is the condition of no stress on metal, so:

$$
\mu = \mu \Delta \sigma_0 + \Delta \sigma V = \mu_0 + RT \ln \alpha + \Delta \sigma V
$$

= $\mu_0 + RT \ln \overline{\alpha}$ (4)

where μ_0 is the electrochemical potential of matal, whereas α is thermodynamic activity and $\bar{\alpha}$ is the mechanics promoting chemical reaction activity. When there is tensile stress on metal, the $\Delta\sigma$ is negative. With deceasing $\bar{\alpha}$ the μ decreases.

For a system of metal electrodes, the corresponding metal ion electrochemical potential is:

$$
\overline{\mu} = \mu_0 + RT \ln \alpha + \Delta \sigma V + zF\varphi = \mu_0 + RT \ln \overline{\alpha} \quad (5)
$$

where Z is the ion valence and F is the Faraday's constant, whereas φ is the potential of system and μ_0 is the constant of mechanics promoting electrochemical reaction. Obviously, the higher the outside stress is, the smaller the electrochemical reaction active energy is, and the easier the corrosion reaction is. From the EIS testing it can also be seen that with the increase of stress, the R_p and R_c (the surfaces reaction resistance) decrease gradually during the same time. The thinner the passive film on the surface is, the more serious the electrochemical reaction is at the same time. Mechanical factor causes reduction of electrochemical reaction and maks the corrosion reaction easier.

5050 aluminum alloy has good corrosion resistance in general environment. But as in this paper, after coupled with the 40CrNiMoA steel which is not a high corrosion resistant metal, the corrosion also becomes very serious. This is caused by three reasons. One is that coupling with the 40CrNiMoA whose electrode potential is higher has anode polarization function for the aluminum alloys essentially. With the increase of over potential, the corrosion becomes serious. Another is that stress can decrease the electrochemical degree of reaction and cause the corrosion reaction easier. The higher the stress is, the lower the electrochemical degree is and the heavier the corrosion is, which is promoted by mechanics. The last is that with the appearing of crack caused by mechanical action, corrosion products cannot fully cover the surface of aluminum alloys making a fresh substrate exposed in solution. With the formation of a big small cathode-node structure, the pitting develops inward and influences the mechanical properties of aluminum alloy seriously.

4 Conclusions

a) There is large potential difference between 5050 aluminum alloy and 40CrNiMoA steel. In 3.5 wt% NaCl solution, galvanic connection significantly promotes the corrosion process and degradation of the mechanical properties especially the plasticity of 5050 aluminum alloy. When aluminum alloy suffers tensile stress after coupled with 40CriNiMoA steel, corrosion would be accelerated. With increasing stress and the extension of time, the corrosion pits grow up and mix together which leads to the surface desquamate. With the surface damaged, the mechanical properties gradually degenerate.

b) Galvanic coupled factor mainly causes aluminum alloy under the condition of anode polarization and influence the electrode surface electrochemical reaction process. The potential of aluminum ally increases but does not meet the passive potential, so the process of anodic reaction is accumulated which influences aluminum alloy mechanical properties.

c) Tensile stress mainly influences the electrochemical potential of aluminum alloy, and promotes the corrosion process. The interaction between mechanics-electrochemical is obvious and the reaction activation energy decreases when the tensile stress increases which leads to the electrochemical potential reduction. So electrode potential difference increases and the corrosion tendency increases. Tensile stress will cause the aluminum alloy surface corrosion product to break and lead to fresh substrate exposed in solution. The productive film loses the protective

function for substrate. Compared to the tensile stress factor, the function of coupling 40CrNiMoA with steel factor which causes aluminum alloy electrochemical activation is more significant.

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