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Short Communication

# Effect of Dissolved Oxygen on Aluminum Corrosion in Simulated Cooling Water for HVDC Systems at 50 °C

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The corrosion behavior of aluminum in 50 °C deionized water with varying dissolved oxygen contents was studied. Compared with deionized water, an aqueous solution with dissolved oxygen promoted aluminum corrosion. With increasing dissolved oxygen content, the corrosion of the aluminum surface became more serious, and the corrosion resistance of the aluminum gradually decreased. When the oxygen content in the water was increased from 0 mg L<sup>-1</sup> to 4.0 mg L<sup>-1</sup>, the aluminum corrosion potential decreased from -1.282 V to -1.503 V, the corrosion current density increased from 1.987 × 10<sup>-7</sup> A cm<sup>-2</sup> to 4.904 × 10<sup>-7</sup> A cm<sup>-2</sup>, and the charge transfer impedance decreased from 8.057 × 10<sup>4</sup>  $\Omega$  cm<sup>-2</sup> to 2.427 × 10<sup>4</sup>  $\Omega$  cm<sup>-2</sup>. The corrosion products for aluminum in water containing oxygen are Al(OH)<sub>3</sub> and Al<sub>2</sub>O<sub>3</sub>, and the corrosion type is mainly pitting corrosion. The number of the pitting holes gradually increase with increasing oxygen content in the aqueous solution. Removing the dissolved oxygen from the cooling water of a valve cooling system with nitrogen is an effective method to limit corrosion of aluminum radiators and the scaling of grading electrodes in high-voltage direct current transmission systems.

Keywords: aluminum, erosion corrosion, dissolved oxygen, radiator, high voltage direct current

# **1. INTRODUCTION**

A high-voltage direct current (HVDC) transmission system has the characteristics of small line size, high transmission efficiency and low power consumption, and thus has become the first choice for excellent resource allocation and long-distance power transmission [1,2]. The converter valve in HVDC realizes the conversion between direct current and alternating current, which generates considerable heat

during operation. A heat exchange system composed of an aluminum radiator and cooling water can maintain a normal working temperature of ~50 °C [3,4]. Deionized water has the characteristics of high specific heat, high thermal conductivity and good voltage stability, so it is often used as the coolant in heat exchangers [5,6]. However, the oxygen in the air easily dissolves in the cooling water, and electrochemical corrosion occurs in the inner cavity of the aluminum radiator, which is in full contact with the cooling water [7,8]. The aluminum ions are deposited onto the surface of the platinum grading electrodes under the action of an electric field and electrochemistry. Scaling off can cause blockage, leakage, and emergency shutdown of HVDC. The scaling of grading electrodes can seriously affect the normal operation of HVDC. This fault caused by aluminum corrosion accounts for approximately 66% of the total faults of converter valves and has become a serious problem in the production of safe power transportation in recent years [9,10].

To completely put an end to the scaling of the grading electrode, it is necessary to completely restrain the corrosion of the aluminum radiator in cooling water [11,12]. At present, coolants with high heat capacity and good electrochemical stability have been reported [7,13,14], which are expected to adapt to HVDC valve cooling systems under harsh conditions such as high voltage, strong electric field, and high temperature (~50 °C).

Obviously, the dissolved oxygen in cooling water is an important factor affecting the aluminum corrosion reaction, so it is of great significance to study the action mechanism of oxygen in the aluminum corrosion process to restrain aluminum corrosion. To avoid the water splitting reaction in a high-voltage environment with a strong HVDC electric field, cooling water with a low ionic conductivity is required. At present, there is no report on the effect of oxygen on the corrosion behavior of aluminum in low conductivity water. In the simulated working environment of HVDC, the oxygen content in the solution was varied by injecting nitrogen into deionized water, and the corrosion behavior of the working electrode representing the aluminum radiator, was studied.

#### **2. EXPERIMENT**

#### 2.1 Experimental device

The inner cavity of the aluminum radiator in HVDC is exposed to deionized water with a high flow rate and is eroded. A magnetic agitator with a speed of 200 rpm is used to stir the solution to simulate the scouring process. The device is shown in Fig. 1.



Figure 1. Experimental equipment used to study aluminum corrosión

#### 2.2 Electrochemical system

#### 2.2.1 Working electrode

The aluminum working electrode is taken from an aluminum radiator, and the exposed working area is 1 cm  $\times$  1 cm. The aluminum model is 3003, which consists of Si (0.57 *wt*%), Fe (0.63 *wt*%), Cu (0.14 *wt*%), Mn (1.27 *wt*%), Zn (0.09 *wt*%), Li (0.03 *wt*%) and Al (97.31 *wt*%) [15]. The auxiliary electrode is a platinum black electrode, and a saturated calomel electrode (SCE) is used as the reference electrode. The potential of the SCE at 50 °C is 0.228 V (relative to the standard hydrogen electrode).

Before the test, the working electrodes were polished with emery paper and nanoalumina powder, washed with distilled water and anhydrous ethanol many times, and then finally placed into a vacuum drying box.

# 2.2.2 Solution

High purity nitrogen was injected into the saturated oxygen solution. The oxygen content in the deionized water was adjusted to be 0 mg L<sup>-1</sup>, 0.5 mg L<sup>-1</sup>, 1.0 mg L<sup>-1</sup>, 2.0 mg L<sup>-1</sup> and 4.0 mg L<sup>-1</sup> by controlling the nitrogen supply time. The dissolved oxygen content in the deionized water was determined by using a JPSJ-605 dissolved oxygen meter. Table 1 shows the relationship between nitrogen supply time and dissolved oxygen content in the aqueous solution (50 °C).

Table 1. The nitrogen injection time and dissolved oxygen content in the solution (50 °C)

Nitrogen injection time (min)	60	33	25	15	3
Dissolved oxygen content (mg L <sup>-1</sup> )	0	0.5	1.0	2.0	4.0

#### 2.3 Electrochemical test

The working electrodes were fixed in position 2 cm away from the bottom of the beaker, and then the solution was poured into the beaker to rest for 10 min. After the corrosion potential of the

aluminum electrode in the solution became stable, the polarization curve and electrochemical impedance spectroscopy (EIS) for the aluminum in the deionized water with varying dissolved oxygen content were measured.

The potentidynamic polarization curve and EIS were measured by a CHI 660D electrochemical workstation. A potential scanning rate of 1 mV s<sup>-1</sup> was used for the potentidynamic polarization curve, and the scanning range was varied from  $E_{open}$  - 0.4 V to  $E_{open}$ + 0.4 V, where  $E_{open}$  is the open-circuit potential. The frequency range for the EIS test was 0.1 Hz - 10<sup>5</sup> Hz, and the amplitude was 5 mV.

For all the solutions, the temperature was 48-52 °C, and the test electrochemical systems were placed into shielded boxes during the test.

# 2.4 Physical characterization

The hanging piece method was used for the aluminum electrode in the aqueous solution with varying oxygen content with a test time of 7 days, and then morphology and energy spectrum analysis for the working face of the electrode were carried out.

Scanning electron microscopy (SEM, Hitachi-S4800) and energy dispersive X-ray spectrometry (EDX) were used to detect the morphology of the corrosion surface and study the distribution of the elements in the corrosion products. The composition of the corrosion products was determined by X-ray diffraction (XRD) at a scanning rate of  $10^{\circ}$  min<sup>-1</sup> between  $10^{\circ}$  and  $90^{\circ}$  with a RigakuUltima IV powder diffractometer with a Cu target.

## **3. RESULTS AND DISCUSSION**

#### 3.1 Effect of dissolved oxygen content on aluminum corrosion

#### 3.1.1 Potentidynamic polarization curve

With increasing dissolved oxygen content in the aqueous solution, the corrosion potential ( $E_{cor}$ ) of the aluminum continued to shift towards negative values, and the corrosion current density ( $J_{cor}$ ) gradually increased (Fig. 2 and Table 2). The dissolved oxygen content in the aqueous solution was higher, the cathodic reaction could be carried out more easily [16], and the dense oxide film formed on the surface of the aluminum electrode was more easily destroyed. This resulted in a slightly lower corrosion potential for the electrochemical corrosion system with a high dissolved oxygen content.



**Figure 2.** Potentidynamic polarization curves for aluminum immersed in aqueous solution for 7 days with scanning rate of 1 mV s<sup>-1</sup>

Table	2.	Fitting	parameters	for th	e po	tentidynam	nic j	polarization	curves	for	aluminum	immersed	in
	aq	ueous s	olution for 7	7 days									

Dissolved oxygen content (mg L <sup>-1</sup> )	0	0.5	1.0	2.0	4.0
Corrosion potential $E_{cor}(V)$	-1.282	-1.324	-1.344	-1.377	-1.503
Corrosion current density $J_{\rm cor}$ (nA cm <sup>-2</sup> )	198.7	291.8	366.0	372.3	490.4

The reduction reaction of the aluminum corrosion process was also affected by the amount of dissolved oxygen in the aqueous solution. The corrosion reaction for the aluminum can be divided into a hydrogen evolution reaction and oxygen absorption reaction, but the anodic reaction always involves the oxidation of aluminum. Because the dissolved oxygen content was a variable in this experiment, the oxygen absorption reaction played a dominant role in the cathodic reaction. At the initial stage of immersion, an oxide film was formed on the surface of the aluminum, which prevented further corrosion of the aluminum. However, the corrosion reaction for the aluminum did not stop, and its speed decreased significantly. The OH<sup>-</sup> produced by the cathodic reaction over an extended period increases the pH in the local area of the aluminum surface, which can continue to destroy the oxide film protection layer on the aluminum [17], resulting in a decrease in the resistance of the coating layer on the aluminum surface and an increase in the corrosion current density. The higher the dissolved oxygen content in the aqueous solution, the more serious the corrosion of the aluminum surface layer.

## 3.1.2 EIS

In the Bode diagram (Fig. 3a), the logarithmic  $[\log (Z)]$  value of the impedance modulus of the aluminum corrosion reaction increased with decreasing frequency logarithm  $[\log (Freq)]$  in aqueous solutions with a dissolved oxygen content of 0 mg L<sup>-1</sup>, 0.5 mg L<sup>-1</sup>, 1.0 mg L<sup>-1</sup>, 2.0 mg L<sup>-1</sup>, and 4.0 mg L<sup>-1</sup>

<sup>1</sup>. This indicates that the corrosion reaction of aluminum in aqueous solution is difficult, and that the corrosion reaction for aluminum needs a higher energy barrier. Under logarithmic values for different frequencies, the largest value for the impedance modulus for aluminum was obtained with a dissolved oxygen content of 0 mg L<sup>-1</sup>, and the impedance modulus decreased with increasing dissolved oxygen content at the same frequency. This result suggests that the increase in dissolved oxygen content can reduce the resistance of the aluminum corrosion reaction and promote the aluminum corrosion reaction.



**Figure 3.** Electrochemical impedance Bode pattern (a) and Bode phase diagram (b) with frequency range of 0.1 Hz -  $10^5$  Hz for aluminum immersed in aqueous solution with dissolved oxygen content of 0, 0.5, 1.0, 2.0 and 4.0 mg L<sup>-1</sup> for 7 days

In the Bode phase diagram (Fig. 3b), each curve shows two peaks, which shows that the corrosion reaction of aluminum in the aqueous solution with varying dissolved oxygen content has two time constants, with two state variables (E and X) in the corresponding system. The two state variables correspond to two capacitive arcs on the Nyquist diagram: the small capacitive arc in the high frequency band and the large capacitive arc in the middle frequency band [18]. The first capacitive arc is caused by the state variable E through charge-discharge relaxation in the circuit, and the second capacitive arc is caused by the formation and adsorption of corrosion products in the corrosion process [19].

Figure 4 shows the impedance spectrum for aluminum immersed in aqueous solution with varying dissolved oxygen content for 7 days and its corresponding fitting circuit diagram. The fitting results for the impedance spectrum are shown in Table 3. *R*s represents the solution resistance between the reference electrode and the aluminum electrode,  $C_{\rm f}$  and  $R_{\rm f}$  represent the capacitance and resistance of the oxide film on the aluminum surface,  $C_{\rm dl}$  represents the aluminum surface double layer capacitance,  $R_{\rm ct}$  represents the aluminum surface transfer resistance, and *W* represents the liquid phase transfer impedance [20].



**Figure 4.** EIS diagram with frequency range of  $0.1 \text{ Hz} - 10^5 \text{ Hz}$  and its corresponding equivalent circuit (insert diagram) for aluminum immersed in aqueous solution with dissolved oxygen content of 0, 0.5, 1.0, 2.0 and 4.0 mg L<sup>-1</sup> for 7 days

Table 3.	The fitting	results of	obtained	from	EIS
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Dissolved oxygen content (mg L <sup>-1</sup> )	0	0.5	1.0	2.0	4.0
$R_{\rm s}(10^3\Omega{\rm cm}^{-2})$	1.877	1.602	1.508	1.429	1.373
$C_{\rm f}(10^{-10}{ m Fcm^{-2}})$	1.138	1.607	1.674	3.505	7.535
$R_{\rm f}(10^4\Omega~{\rm cm}^{-2})$	4.767	4.513	3.172	1.951	1.501
$C_{ m dl} (10^{-6}{ m F}{ m cm}^{-2})$	0.5321	1.920	2.948	5.321	6.084
$R_{\rm ct}(10^4\Omega{\rm cm}^{-2})$	8.057	4.705	4.069	3.506	2.427
$W(10^{-5}\Omega{ m cm}^{-2})$	1.175	1.028	0.9762	0.8811	0.7963

When dissolved oxygen was present in the aqueous solution, the solution resistance, film resistance, charge transfer resistance and Warburg impedance of the aluminum decreased, while the membrane capacitance and electric double layer capacitance increased. The dissolved oxygen accelerated the corrosion on the aluminum surface and formed corrosion pits that further reduced the corrosion resistance of aluminum [21]. The higher the oxygen concentration in the aqueous solution, the more serious the aluminum corrosion; in addition, the partially dissolved corrosion products reduced the solution impedance. In an aqueous solution with high oxygen content, the corrosion product coverage of the aluminum surface was higher, the surface double layer capacitance increased, the surface charge transfer resistance decreased, and the corrosion reaction for aluminum was accelerated.

3.2 Composition and morphology of corrosion products

# 3.2.1 XRD

The XRD patterns for aluminum surface immersed in aqueous solution with dissolved oxygen content of 0, 0.5, 1.0, 2.0 and 4.0 mg L<sup>-1</sup> for 7 days is shown in Fig. 5. The peaks observed at 39°, 45°, 65° and 78° for all samples correspond to the standard card Al (PDF#01-1176). The peaks at 18° and 25° correspond to Al(OH)<sub>3</sub> (PDF#37-1377) and Al<sub>2</sub>O<sub>3</sub> (PDF#04-0875) peaks, respectively. The corrosion products for the aluminum in the aqueous solution with dissolved oxygen were Al<sub>2</sub>O<sub>3</sub> and Al(OH)<sub>3</sub>, which is consistent with the corrosion products found for aluminum in wet environments [22].



**Figure 5.** XRD spectra for an aluminum surface immersed in solution with dissolved oxygen content of 0, 0.5, 1.0, 2.0 and 4.0 mg L<sup>-1</sup> for 7 days

In the aqueous solution with a dissolved oxygen content of 4.0 mg  $L^{-1}$ , the peaks due to Al(OH)<sub>3</sub> and Al<sub>2</sub>O<sub>3</sub> were stronger. With a decrease in dissolved oxygen content, the Al(OH)<sub>3</sub> and Al<sub>2</sub>O<sub>3</sub> peaks became weaker. Aluminum in low dissolved oxygen solution only formed hydroxide with low crystallinity, and the corrosion tendency for aluminum was weakened. Deoxidization in water can inhibit the corrosion of aluminum.

# 3.2.2 SEM and EDX

In the aqueous solution with a high dissolved oxygen content, the corrosion of the aluminum surface was more serious, and many pitting holes appeared (Fig. 6). However, with a decrease in dissolved oxygen content, the aluminum corrosion was gradually improved. For aluminum immersed in

 $4.0 \text{ mg L}^{-1}$  dissolved oxygen solution for 7 days, most of the elements on the surface were aluminum with a small amount of carbon and oxygen (Fig. 7, Table 4). With an increase in the dissolved oxygen content in the aqueous solution, the oxygen on the aluminum surface gradually increased. This indicated that the corrosion products on the aluminum surface gradually increased with increasing dissolved oxygen content. The high dissolved oxygen content accelerated the oxygen absorption reaction for the aluminum and promoted corrosion.



**Figure 6.** SEM diagram of an aluminum surface immersed for 7 days in an aqueous solution with a dissolved oxygen content of 0 mg L<sup>-1</sup> (a), 0.5 mg L<sup>-1</sup> (b), 1.0 mg L<sup>-1</sup> (c), 2.0 mg L<sup>-1</sup> (d), and 4.0 mg L<sup>-1</sup> (e). Scale: 200 nm



**Figure 7.** EDX spectra for an aluminum surface immersed in an aqueous solution with a dissolved oxygen content of 4.0 mg L<sup>-1</sup> for 7 days

97.71

96.67

96.22

95.58

93.81

93.16

1.13

1.96

2.54

**Table 4.** EDX analysis of the aluminum surface after immersion in aqueous solution with dissolved oxygen content of 0, 0.5, 1.0, 2.0 and 4.0 mg L<sup>-1</sup> for 7 days

## 3.3 Corrosion mechanism analysis

1.0

2.0

4.0

The oxide film on the aluminum surface led to good corrosion resistance for the aluminum [23]. Generally, in neutral media, the corrosion reactions of aluminum in aqueous solution proceed as follows: Anodic reaction:

$$A1 - 3e \rightarrow Al^{3+}$$
 1)

$$Al^{3+}+3OH^{-}\rightarrow Al(OH)_{3}$$
 2)

1.87

3.21

4.15

1.16

1.37

1.24

2.55

2.98

2.69

Cathodic reaction:

$$0_2 + 2H_2O + 4e^- \rightarrow 4OH^-$$
 3)

$$2H^+ + 2e^- \rightarrow H_2 \tag{4}$$

Corrosion reaction:

$$4A1 + 3H_2O + 3O_2 = 2Al(OH)_3 + Al_2O_3$$
5)

The type of corrosion of aluminum in the aqueous solution containing oxygen was mainly pitting corrosion (Fig. 8). The oxide film on the aluminum surface was incomplete and defective, and the aqueous solution containing oxygen contacted the aluminum metal matrix through the defects. The oxide film with fewer defects had a lower local state and was more resistant to pitting corrosion [24]. The aluminum coated with an oxide film had better corrosion resistance and could be used as a cathode. The corrosion resistance of aluminum at the defect was poor and was used as the anode. This kind of corrosion cell with a large cathode and small anode can accelerate the corrosion of the defect. The defective local area is a relatively closed environment, which can easily form occluded cells.

In the beginning, due to the existence of dissolved oxygen, a large amount of  $OH^-$  can be formed in the occluded area, resulting in the enhancement of local alkalinity. The corrosion reaction is mainly oxygen inhalation corrosion, forming pitting holes, which aggravate the corrosion of aluminum. However, with the further development of the corrosion reaction, the hydrogen evolution reaction can begin to occur after oxygen consumption in the occluded area, which further aggravates the corrosion of the pitting holes. The  $A1^{3+}$  produced in this process spreads out of the pores and combines with OH<sup>-</sup> generated by the cathodic reaction to form precipitates, which accumulate around the pitting holes, making the environment more restrictive. The corrosion reaction shows the characteristics of autocatalysis [25]. This leads to pitting corrosion of the aluminum.



Figure 8. Schematic diagram for the pitting corrosion of aluminum in aqueous solution containing oxygen.

# 4. CONCLUSION

In this paper, the erosion behavior of aluminum in aqueous solution with varying dissolved oxygen content was studied. Compared with dissolved oxygen solution, the corrosion of aluminum in deoxidized water was found to be obviously weak. With increasing dissolved oxygen content from 0 mg  $L^{-1}$  to 4.0 mg  $L^{-1}$ , the corrosion current density for aluminum gradually increased, the coating resistance and corrosion charge transfer impedance gradually decreased, and the corrosion of the aluminum surface gradually worsened. The aluminum corrosion deteriorated with increasing oxygen content in the aqueous solution.

Deoxidation can enable one to delay corrosion. The type of aluminum corrosion in dissolved oxygen solution was mainly pitting corrosion, and the corrosion pits gradually increased with increasing dissolved oxygen. Deoxidation can reduce the corrosion rate for aluminum in aqueous solution. The inhibition of aluminum corrosion with the cathodic oxygen absorption reaction by deoxidation was achieved by reducing the dissolved oxygen in the solution. The introduction of high purity nitrogen to remove dissolved oxygen is a very effective method to restrain aluminum corrosion, which has a certain guiding significance for the corrosion protection of HVDC valve cooling systems.

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