

# The Use of Mixed Electrolyte Solutions with Improved Electrochemical and Discharge Behavior of a Low-cost Commercial Aluminum Alloy for Aluminum-air Batteries

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## Abstract

The electrochemical and discharge behavior of low-cost commercial aluminum alloy 8011 as an anode was investigated in mixed electrolyte solutions between sodium chloride (NaCl) and sodium hydroxide (NaOH) for use in aluminum-air batteries. The results indicated that mixed electrolyte solutions improve passive film formation and reduce the self-corrosion of the aluminum alloy. In addition, the oxygen reduction reaction (ORR) of chain-like carbon nano-onions (CLCNOs) as a positive air electrode is also improved. The maximum capacity density in the mixed electrolyte solutions was returned as high as 1,232 mAhg<sup>-1</sup>, which was higher than the sodium chloride and sodium hydroxide solution only for 1.1 and 20.9 times, respectively.

**KEYWORDS:** Commercial aluminum alloy, Chain-like carbon nano-onions, Aqueous electrolyte, Oxygen reduction reaction, Metal-air battery

## 1 INTRODUCTION

At present, the world is searching for renewable sources of energy that are environmentally friendly in order to replace conventional energy sources such as coal, oil, and natural gas. Aluminum-air batteries are a clean energy source among metal-air batteries that are getting more attention because they have high energy density in addition to being environmentally friendly, inexpensive, and recyclable (Egan et al., 2013; Rahman et al., 2013; Liu et al., 2017; Faegh et al., 2021). Alkaline electrolytes have been widely used as electrolytes for aluminum-air batteries (Goel et al., 2020; Buckingham et al., 2021) because they provide good electrical conductivity and can dissolve the aluminum hydroxide formed on the surface of aluminum

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(Linden & Reddy, 2001). However, there are severe corrosion problems involving aluminum (Egan et al., 2013; Mokhtar et al., 2015; Ge et al., 2015) and the crystallization of carbonate in the porous positive air electrode (Li & Bjerrum, 2002). There are many ways to solve the corrosion problem of the aluminum anode such as alloying Al with other elements (Macdonald et al., 1988; El Abedin & Saleh, 2004; Li et al., 2019), low-temperature operating batteries (Zuo et al., 2019), or the addition of corrosion inhibitors in alkaline electrolytes (Moghadam et al., 2017; Sun & Lu, 2015; Salehi et al., 2017).

NaCl is an inorganic compound commonly used as an electrolyte in aluminum-air batteries due to its suitable electrical conductivity as well as being readily accessible, inexpensive, and less self-corrosive than the alkaline electrolyte. With such properties, it should reduce the self-corrosion of the aluminum anode if used in combination with an alkaline electrolyte. However, there are no definitive studies concerning the use of mixed electrolyte solutions between NaCl and NaOH for aluminum-air batteries.

Based on the aforementioned, we studied the performance of aluminum-air batteries in various mixed electrolyte solutions between NaCl and NaOH using low-cost commercial aluminum alloy 8011 as the anode. We also investigated the self-corrosion and electrochemical behavior of the aluminum anode, as well as the ORR of CLCNOs (Nit Namwong & Chaiyaput Kruehong, 2022) as a positive air electrode. This study aimed to develop high-performance aluminum-air batteries from materials that are easily available, economical, and environmentally friendly for future use.

## 2 RESEARCH OBJECTIVES

The purpose of this paper is to study the electrochemical and discharge behavior of aluminum-air battery using a mixed electrolyte solutions between NaCl and NaOH, with low-cost commercial aluminum alloy 8011 as the anode and the chain-like carbon nano-onions (CLCNOs) as a positive air electrode.

## 3 LITERATURE REVIEW

Aluminum-air battery research related to solving the corrosion problem of the aluminum anode found that alloying Al with other elements such as In, Ga, Te, P, and Mg (Macdonald et al., 1988; El Abedin & Saleh, 2004; Li et al., 2019), can reduce hydrogen evolution and self-corrosion. However, there is additional cost for the aluminum anode. In addition, commercial aluminum alloys such as 5052 (Wang et al., 2015), 7475 (Pino et al., 2016), and 8011 (Fan et al., 2016) have been studied for use as anodes. These have been found to improve the performance of aluminum-air batteries, but their self-corrosion is higher than pure aluminum. Another method is to add corrosion inhibitors to the alkaline electrolytes, which include organics (Moghadam et al., 2017; Liu et al., 2019; Abiola et al., 2009), inorganics (Sun & Lu, 2015; Sarangapani et al., 1985; Chang et al., 2008), and hybrid inhibitors (Salehi et al., 2017; Nie et al., 2017; Zhu et al., 2019). The inhibitory performance of organic additives is inferior to inorganic additives such as ZnO due to the low conductivity in the electrolyte (Jiang et al., 2020). Besides, hybrid inhibitors are more effective against self-corrosion than inorganic and organic alone (Jiang et al., 2020; Li et al., 2022). Other studies have been able to reduce the

self-corrosion of the aluminum anode such as low-temperature operating batteries (Zuo et al., 2019), ionic liquid electrolytes (Gelman et al., 2014; Levy et al., 2020), dual-electrolyte systems (Teabnamang et al., 2020), and solid alkaline electrolytes (Di Palma et al., 2017). Furthermore, our previous research found that the use of mixed electrolyte solutions between NaCl and NaOH could reduce the corrosion of aluminum anode (Vichapol Manadee et al., 2017; Nit Namwong et al., 2019). However, the calculation of the battery's energy must be done carefully. Due to the mixed electrolyte solutions in which most of the corrosion products adhere to the aluminum surface after the test. If the corrosion products are not appropriately cleaned, weighing of used aluminum will not be accurate. As a result, the energy value calculated is error.

## 4 RESEARCH METHODOLOGY

### 4.1 Preparation of anode and cathode

The 8011 aluminum alloy anode (Al-0.73%Fe-0.55%Si-0.02%Mn) and carbon cathode were prepared following the method detailed in our previous work (Nit Namwong & Chaiyaput Kruehong, 2022; Saifon Kruehong et al., 2015). The size of the aluminum anode was controlled to be equal to 1 cm<sup>2</sup>. A sample of carbon cathode was synthesized onto a nickel foam sheet (2 cm<sup>2</sup>) by candle flame combustion at 50 s, of which the carbon nanomaterials had a mass of 0.49 mgcm<sup>-2</sup>.

### 4.2 Self-corrosion

Self-corrosion was performed in 4M NaCl, 4M NaOH, and mixed electrolyte solutions between NaCl and NaOH electrolytes at various volumetric ratios of 100:0, 99:1, and 0:100. Aluminum alloy sheets prepared according to 4.1 were weighed before testing and immersed in the mixed electrolyte solutions for 1 hour. The surface was cleaned with a cotton ball moistened with ethanol. It was left to dry and then taken back to identify the amount of weight loss due to self-corrosion. Subsequently, the lost weight, time spent, and size of the workpiece was used to calculate the corrosion rate of the workpiece at different electrolyte ratios.

### 4.3 Electrochemical testing

The electrochemical behavior was studied using anodic and cathodic polarization techniques by means of a potentiostat (Gamry, model Reference 600 (R600)). This test connected a three-electrode cell to be tested at room temperature. The counter and reference electrodes were platinum wire of 1 mm in diameter and 5 cm in length and silver/silver chloride (Ag/AgCl), respectively. The electrode potentials described in this part are referred to as the saturated KCl/AgCl/Ag electrode (SSE). The working electrodes were CLCNOs coated Ni foams and aluminum alloy sheets for cathodic polarization and anodic polarization, respectively. The open-circuit potential was tested for 10 minutes, after which it was tested by anodic and cathodic polarization by adjusting the ratio of the mixed electrolyte solutions to 100:0, 99:1, and 0:100.

#### 4.4 Battery performance testing

A galvanostatic technique was used to test the performance of the aluminum-air battery. This test was connected to a two-electrode cell using the CLCNOs coated Ni foam dimensions of 2 cm x 2 cm as a cathode, controlling the exposed area of 2 cm<sup>2</sup>. The aluminum sheet with dimensions of 1 cm x 2.5 cm was an anode electrode by controlling the exposed area of 1 cm<sup>2</sup>. This was conducted by using a constant current density of 1 mAcm<sup>2</sup> compared to the size of the aluminum area used in the test under room temperature by adjusting the ratio of the mixed electrolyte solutions at various proportions to find the cell voltage, capacity density, and energy density. In addition, the mass of aluminum consumed was compared between a surface without and with corrosion products to be used to calculate the capacity density and energy density.

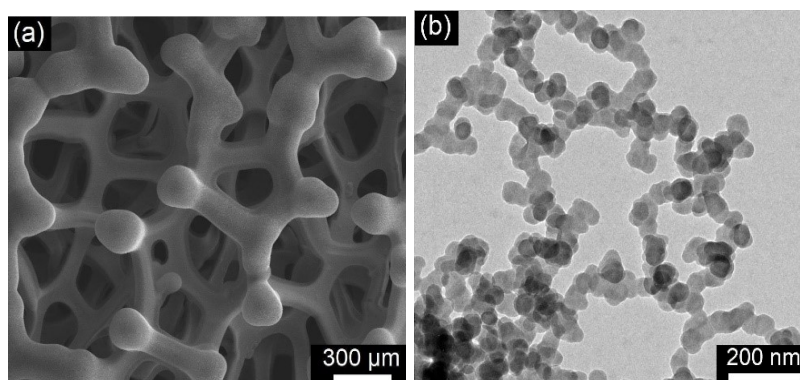
#### 5 Physical characteristics testing

The morphology of CLCNOs was analyzed using field emission scanning electron microscopy (FE-SEM, FEI Helios Nanolab) and transmission electron microscopy (TEM, FEI Tecnai G2 20) operated at 20 kV and 200 kV, respectively.

### 6 RESULTS

#### 6.1 Physical characteristics of the positive air electrode

Figure 1a shows an FE-SEM image of CLCNOs coated on a nickel foam substrate after candle flame combustion synthesis. It is observed that the three-dimensional structures of the nickel foam coated with carbon nanomaterials and nanometer-sized particles are densely scattered on the surface. TEM images of the synthetic carbon nanomaterials on the nickel foam sheet are shown in Figure 1b. The carbon nanomaterials formed on the nickel foam sheet are spherical and connected in a chain-like fashion with a diameter of 20-50 nm.



**Figure 1:** Physical characteristics of the positive air electrode: (a) FE-SEM image of CLCNOs coated on a nickel foam substrate and (b) TEM image of CLCNOs.

Self-corrosion of aluminum alloy sheet The corrosion rate is a test of the condition of aluminum alloy sheets when there is no current in the circuit. The test is used to determine the stability of aluminum alloy sheets under a parasitic reaction. This is the time when the

aluminum reacts with the electrolyte to produce hydrogen gas, as in the reaction in equation (1).

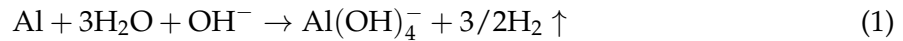


Figure 2 shows the results of the corrosion test of aluminum alloy sheets in NaCl, NaOH, and mixed electrolyte solutions. It is also seen that the aluminum alloy sheet in the NaCl electrolyte (100:0) was the least self-corrosion, whereas the NaOH electrolyte exhibited severe self-corrosion and produced large quantities of hydrogen gas. For the mixed electrolyte solutions at ratios of 99:1, and 98:2, the corrosion rates were reduced by 11.2, and 7.3 times, respectively, compared to the NaOH electrolyte. Therefore, the corrosion rate decreases when decreasing the amount of NaOH electrolyte in the mixed electrolyte solutions, which affects the discharge time of the batteries.

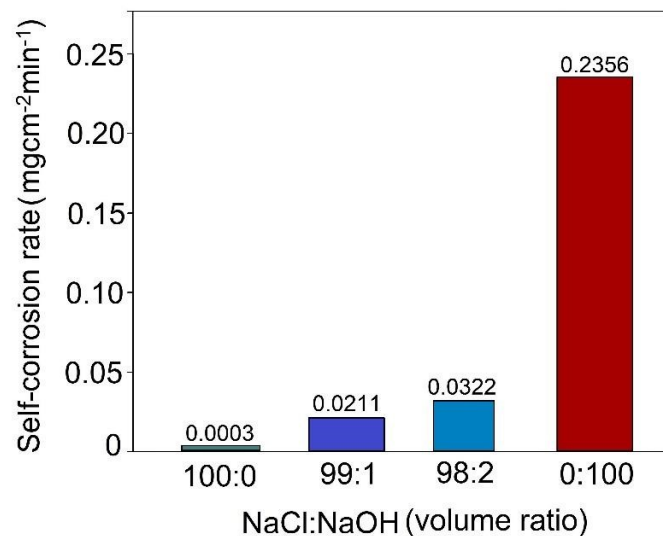
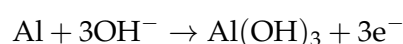


Figure 2: Self-corrosion rate of aluminum in various electrolytes.

## 6.2 Electrochemical testing

Figure 3 shows the anodic polarization curves of the aluminum anode in the mixed electrolyte solutions of 99:1 compared to the NaCl electrolyte and NaOH electrolyte. It was found that the corrosion potential in the mixed electrolyte solutions is less negative than that of the NaOH electrolyte since the NaCl electrolyte in the system causes a decrease in NaOH concentration and an aluminum hydroxide film is formed (Chen et al., 2015), as in equation (2).

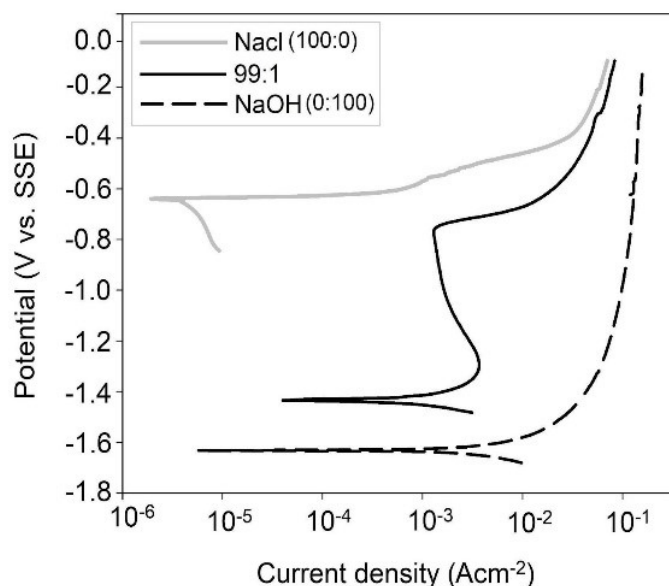


For the NaOH electrolyte, however, the hydroxide film at the aluminum anode decomposes, as in equation (3).



Meanwhile, the NaCl electrolyte has a corrosion potential of  $-0.65$  V vs. SSE, which is less negative than the mixed electrolyte solutions ( $-1.4$  V vs. SSE).

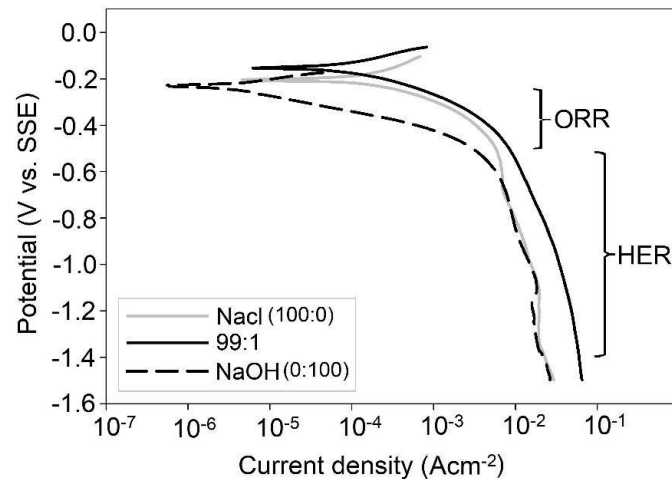
It is observed that the mixed electrolyte obtained by adding NaOH electrolyte to the NaCl electrolyte increased the corrosion of aluminum, which is perceived as the corrosion potential increased negatively from  $-0.65$  V to  $-1.40$  V vs. SSE, which behaves similarly to NaOH electrolyte. However, the NaCl electrolyte used as the main ingredient lowers the self-corrosion compared to the NaOH electrolyte (Fig. 2) because the surface of the aluminum has a complex film formed. This corresponds to the current density at the potential range of the passive region ( $-0.79$  V to  $-1.30$  V vs. SSE). Corrosion in the mixed electrolyte is higher than that of the NaCl electrolyte but lower than that of the NaOH electrolyte. Such behavior is beneficial to solve the problem of anode operation in traditional batteries run in NaCl electrolyte and NaOH electrolyte. Usually, NaCl electrolyte is limited in terms of low current (Mokhtar et al., 2015; Pino et al., 2016), while NaOH electrolyte is excessively self-corrosive (Egan et al., 2013; Mokhtar et al., 2015; Ge et al., 2015).



**Figure 3:** Anodic polarization curves of the aluminum anode in mixed electrolyte solutions at various ratios.

Figure 4 shows the cathodic polarization curves of the positive air electrode in the mixed electrolyte solutions at a ratio of 99:1 compared to NaCl electrolyte and NaOH electrolyte. It is observed that the mixed electrolyte has a significantly higher current density than the NaCl electrolyte and NaOH electrolyte. The electrochemical behavior of the mixed electrolyte can be considered in two potential ranges, where the potential between  $-0.25$  V to  $-0.50$  V vs. SSE is the range of ORR in which the oxygen consumption occurred on the surface of CLCNOs (Nit Namwong & Chaiyaput Kruehong, 2022). The mixed electrolyte has a higher current density than the NaOH electrolyte but is near the NaCl electrolyte. If considering the potential between  $-0.50$  and  $-1.40$  V vs. SSE (the range of hydrogen evolution reaction (HER)), however, the mixed electrolyte has a higher current density than the NaCl electrolyte and NaOH electrolyte. This indicates that the mixed electrolyte solutions enhance the oxygen

intake of the positive air electrode more than the NaCl electrolyte and the NaOH electrolyte alone.

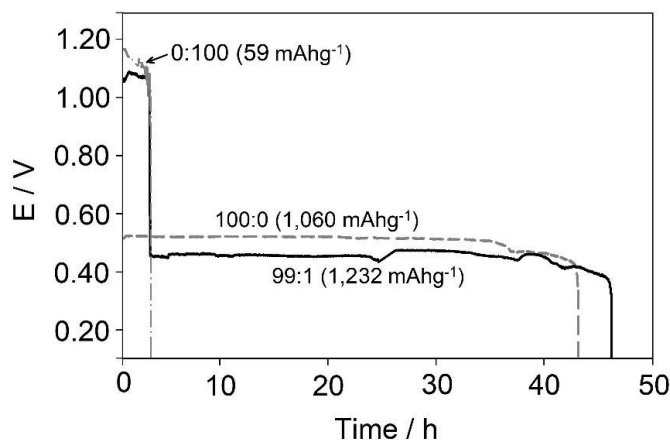


**Figure 4:** Cathodic polarization curves of the positive air electrode in mixed electrolyte solutions at various ratios.

### 6.3 Discharge performance

Discharge performance testing of the aluminum alloy as an anode was performed at different electrolyte ratios and current densities to study the behavior and specific capacities of aluminum-air batteries. Figure 5 shows an improvement in the discharge profile by a constant current density of  $1 \text{ mAcm}^{-2}$  at different electrolyte ratios (100:0, 99:1, and 0:100). It is apparent that the mixed electrolyte solutions at a ratio of 99:1 give two cell potential plateaus, which are 1.07 V for a plateau of 2 h and 0.45 V for another main plateau between 2 to 45 h due to the consumption of  $\text{OH}^-$  in the mixed electrolyte solutions, as in equation (2) (Chen et al., 2015), thus providing a specific capacity of about  $1,232 \text{ mAhg}^{-1}$ , higher than other mixed electrolyte solutions at ratios of 100:0, 0:100 (Fig. 5), and 98:2 ( $773 \text{ mAhg}^{-1}$ ). Its specific capacity is the highest due to the formation of complex film and the lower self-corrosion rate than those of other electrolytes in the period of 2-45 h. This makes the discharge time significantly longer.

In the immersion test, the self-corrosion of an aluminum anode in the mixed electrolyte solutions is usually higher than the NaCl electrolyte (Fig. 2), but it is lower than the NaOH electrolyte for the battery discharge test due to a complex film formation, which is observed as a black film on the aluminum surface. This is consistent with the research of Daufin et al. (1977) and Alaneme & Bodunrin (2011), which found that the formation of the black film on the aluminum alloy surface mitigates the self-corrosion under alkaline electrolytes. In the case of the mixed electrolyte solutions (99:1), the black film can be observed with the naked eye to be more stable and thicker as the discharge time increases. As a result, it has lower self-corrosion in the period of 2-45 h and thus has a higher specific capacity than the NaCl electrolyte.



**Figure 5:** Discharge profiles of aluminum-air batteries at a constant current density of  $1 \text{ mAcm}^{-2}$  in mixed electrolyte solutions at various ratios.

A comparison of the energy density and specific capacity of aluminum-air batteries in different electrolytes. In past research, aluminum-air batteries using KOH and NaCl as electrolyte solutions had an energy density of lower than  $500 \text{ Whkg}^{-1}$  and a specific capacity of lower than  $1,200 \text{ mAhg}^{-1}$  (Pino et al., 2016; Yu et al., 2018; Liu et al., 2016; Choi et al., 2017). For this research, however, the mixed electrolyte solutions with ratios of 99:1 has an energy density of  $596 \text{ Whkg}^{-1}$  and a specific capacity of  $1,232 \text{ mAhg}^{-1}$ ; the energy is similar to aluminum anode improvement with carbon layer (Pino et al., 2016). However, the energy value is lower than that of hybrid inhibitors of KOH, ethanol, and Benzoic acid (Li et al., 2022), NaOH in paper-based aluminum-air batteries (Wang et al., 2019) and low-temperature alkaline electrolytes ( $2,480 \text{ mAhg}^{-1}$ ) (Zuo et al., 2019). These suggest the possibility of applying the mixed electrolyte solutions of NaCl and NaOH as gel or paper-based electrolytes and that working at low temperatures might be able to achieve a higher energy value. When this research is applied to commercial products, both mixed electrolyte solutions and electrodes (commercial-grade aluminum alloy as an anode, and CLCNOs as a positive air electrode) can reduce the production cost compared to traditional work or other research.

There is also an observation about calculating the specific capacity, which must be done carefully as the weight of the aluminum lost will greatly affect the specific capacity, especially in NaCl electrolyte, where most of the corrosion products are attached to the aluminum surface after tests. If the corrosion product is not completely cleaned from the aluminum surface, the weighing of the consumed aluminum will not be precise. As a result, the energy value calculated is greater than the accurate value. For example, this study performed the test in 4 M NaCl electrolyte. The specific capacity calculation without completely removing corrosion products yielded  $1,727 \text{ mAhg}^{-1}$  which is 1.6 times higher than the completely removed case ( $1,056 \text{ mAhg}^{-1}$ ). This suggests that researchers should be mindful and focus on weighing the anode consumed.



## 7 CONCLUSION

Aluminum-air batteries use mixed electrolyte solutions of NaCl and NaOH. The results show that mixed electrolyte solutions (99:1) can reduce the self-corrosion of an 8011 aluminum alloy anode and enhance the oxygen reduction reaction (ORR) of a positive air electrode better than using NaCl and NaOH electrolytes alone. Its specific capacity is the highest due to the formation of complex film and the lower self-corrosion rate than those of other electrolytes. This makes the discharge time significantly longer. These results will enable further improvements in the efficiency of aluminum-air batteries as a renewable energy source in the future.

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