

Corrosion characteristics of aluminum alloy in bio-ethanol blended gasoline fuel: Part 2. The effects of dissolved oxygen in the fuel

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ABSTRACT

This study examined the effects of dissolved oxygen on the corrosion properties of aluminum cast alloy, A384, in bio-ethanol blended gasoline fuel at 100 °C. Oxygen enhanced the corrosion resistance of the alloy by forming a protective hydrous oxide film. The acetic acid and water produced by ethanol oxidation in the fuel had a corrosive and protective effect, respectively. However, the protective hydrous oxide film enhanced by the water suppressed the corrosiveness of the acetic acid in the fuel when the two by-products coexisted. Therefore, the water formed by dissolved oxygen in the bio-ethanol blended gasoline fuel enhanced the corrosion resistance of the aluminum alloy by promoting the formation of a protective surface film.

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1. Introduction

Automotive fuel has met the challenges of reducing the consumption of fossil fuel and improving the efficiency and performance. Bio-ethanol is a promising candidate for alternative energy in automotive applications [1–4]. Winnington and Siddiqui [5] reported that ethanol-blended fuel improves the spark ignition performance of automotive engines. The fuel can reduce the content of harmful exhaust substances due to the high octane number (~113) and have superior anti-knock characteristics over hydrocarbon fuels with a similar octane number [3,4,6–8]. Moreover, the fuel has an advantage in fuel economy with a 1.5–5% cost reduction by blending of 10–30 vol.% ethanol compared to regular unleaded gasoline [4].

Nevertheless, there is controversy regarding the application of bio-ethanol to automotive fuel due to concerns regarding the corrosion problems that might occur in light metal components of fuel and engine system, particularly aluminum alloys. There are only a few literatures on the corrosion characteristics of automotive component materials under the modified fuel conditions, particularly at the high temperatures that might be reached in an engine [9–11]. Therefore, the reliability and safety of fuel system components operated under modified fuel conditions should be

confirmed by an investigation into the corrosion characteristics of aluminum alloys.

In the previous study [12], the corrosion characteristics of an aluminum alloy were investigated in bio-ethanol blended gasoline fuel with various bio-ethanol contents (10–20 vol.%) and temperatures (60–100 °C). Thermally activated chemical reaction between the metal and ethanol was found to be the mechanism of pitting corrosion, whereas the hydrous oxide film formed on the alloy surface inhibited the initiation of the pitting corrosion. However, the corrosion reaction and the property of surface oxide film can be affected by impurities or additives such as dissolved oxygen, water, sulfate, chloride ions and other organic contaminants incorporated into the fuel [13,14]. To confirm the reliability of fuel system components operated under bio-ethanol blended gasoline fuel, it is important to investigate the effect of these impurities or additives on the corrosion of aluminum alloys. In the present study, the effect of dissolved oxygen among the additives was focused on.

Oxygen which is included as an oxygenate in automotive fuel is known to improve engine performance and reduce the emission of harmful substances by increasing the octane rating [15–19]. Oxygenates provide the gasoline fuel with a better anti-knock performance due to the higher octane rating [15]. Eyidogan et al. [16] reported that the alcohol blended gasoline fuel with more oxygen rate caused to increase the performance of spark ignition engine by increasing the combustion and thermal efficiencies. Hamdan and Al-Subaih [17] reported that the addition of MTBE as an oxygenate up to 10% to gasoline fuel increased the octane number and power

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output of engine. They stated that the emission of pollutants such as carbon monoxide and hydrocarbons decreased because MTBE provides the combustion system with more oxygen content. Sendzikiene et al. [19] reported that the smoke index and the emissions of polycyclic aromatic hydrocarbons, NO_x and carbon monoxide decreased with increasing oxygen content in rapeseed oil methyl ester–diesel–ethanol fuel. They found that the optimal oxygen content in the fuel mixture was 15–19%.

On the other hand, the corrosion rate of a metal generally would increase with increasing dissolved oxygen content in the aqueous medium by activating a cathodic reduction reaction [20]. However, the effect of oxygen on the corrosion of metal components in bio-ethanol-blended fuel is not clearly understood because of the chemical complexity of the fuel.

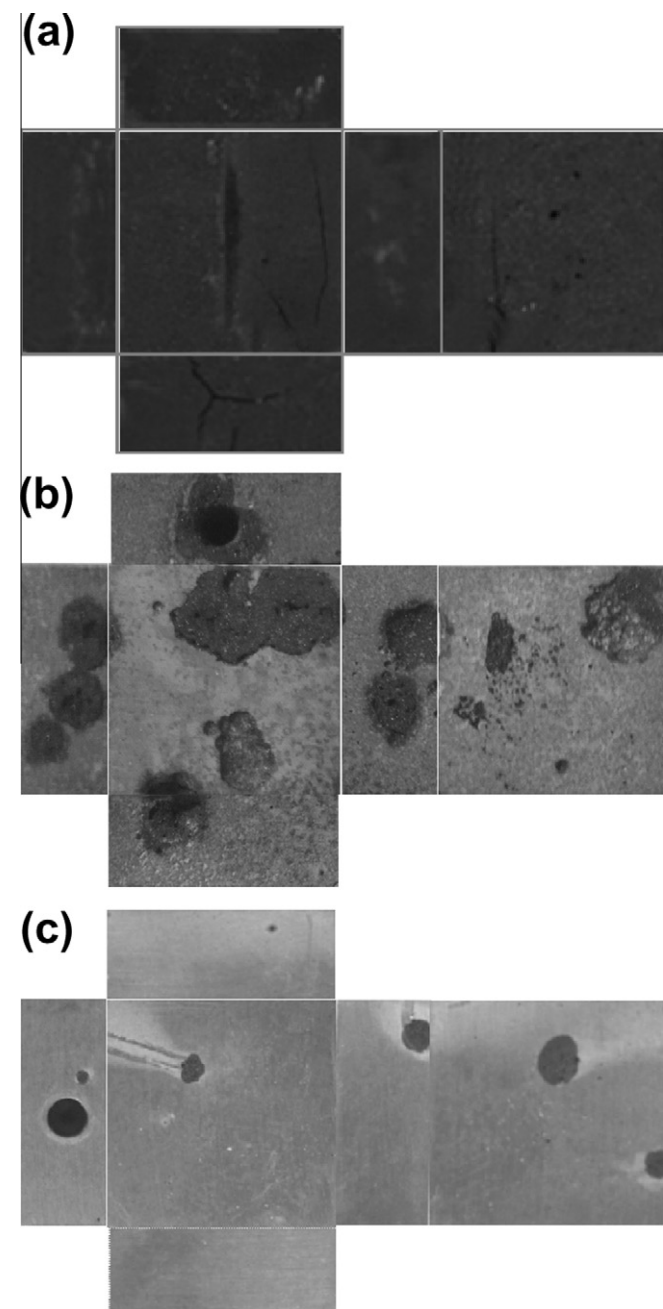


Fig. 1. Corroded surfaces of the aluminum alloy after the immersion tests at 100 °C for 24 h in (a) oxygen-free, (b) ambient, and (c) oxygen-enriched E20 fuel.

This study examined the effects of oxygen and agents (acetic acid and water) introduced by the oxygen on the corrosion of aluminum alloys in high temperature bio-ethanol blended gasoline fuel by surface observations and electrochemical tests.

2. Materials and methods

2.1. Materials

An aluminum cast alloy, T5 heat-treated A384 was prepared with a rectangular shape of $12 \times 12 \times 5$ mm. The detailed chemical composition is notated in elsewhere [21]. The surface was abraded with 600-grit SiC paper and rinsed ultrasonically with ethanol and dried with air. The preparation process for all specimens was completed within 10 min before the subsequent corrosion tests to obtain an identical state of surface oxide film, which might affect the corrosion properties of the aluminum alloy.

2.2. Immersion tests

The corrosion medium used this study was a fuel mixture of commercial unleaded gasoline and anhydrous bio-ethanol with a purity of 99.9% (less than 0.08 vol.% of water). The ethanol content was fixed at 20 vol.% (E20). To examine the effect of dissolved oxygen on the corrosion characteristics of the aluminum alloy, the oxygen concentration in the fuel was controlled by bubbling with nitrogen or oxygen gas (high purity grade of 99.999%) for 2 h before heating the fuel to the test temperature of 100 °C. Accordingly, three conditions were set up; oxygen-free, ambient and oxygen-enriched fuel. Corrosive agents such as acetic acid and/or water produced by the oxidation reaction of ethanol were injected into

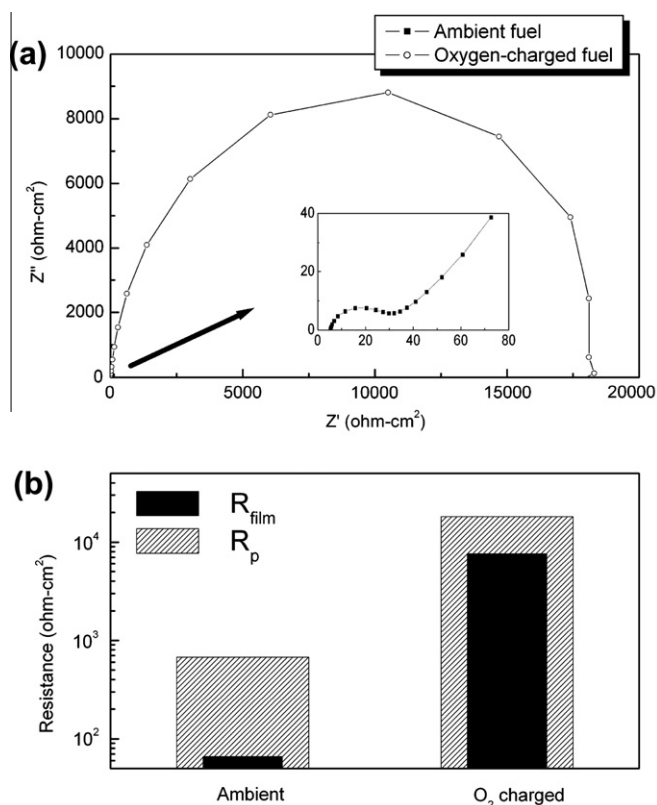


Fig. 2. Results of EIS measurements for the aluminum alloy immersed in E20 fuel with various oxygen concentrations: (a) Nyquist plots and (b) calculated oxide film and polarization resistance.

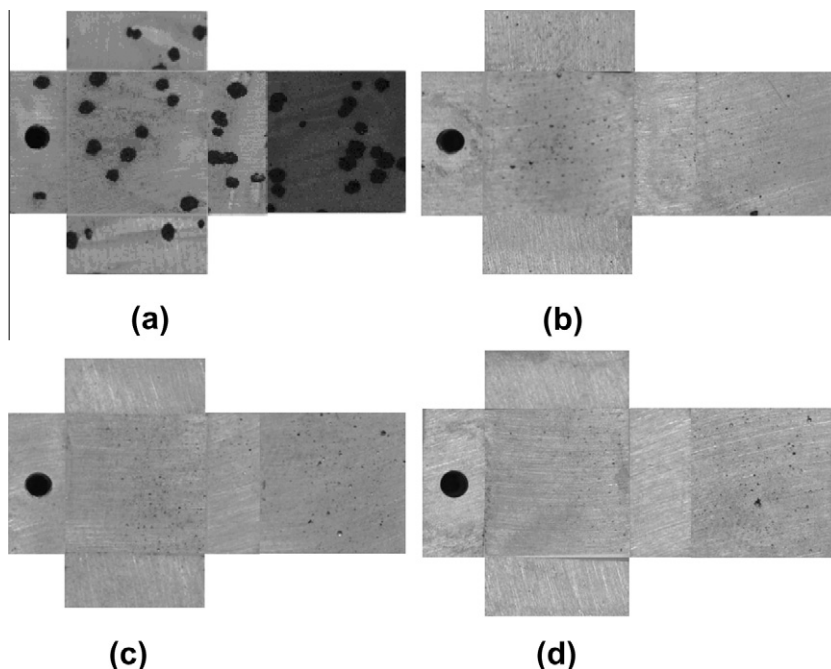


Fig. 3. Corroded surfaces of the aluminum alloy after the immersion tests at 100 °C for 6 h in oxygen-free E20 fuel with various water contents at (a) 0%, (b) 0.1%, (c) 0.5%, and (d) 1.0%.

Table 1
The weight change (mg/cm²) of the aluminum alloy after the immersion tests in oxygen-free E20 fuel at various water contents.

Water content	0%	0.1%	0.5%	1.0%
Changes of weight	-2.307	0.341	0.446	0.537

the fuel to confirm the effects of the chemical changes in the fuel by dissolved oxygen on the corrosion of the alloy. Each of acetic acid and water was injected into the fuel at the content of 0, 0.1, 0.5 and 1.0 vol.%. A mixture composed of 76 vol.% of acetic acid and 24 vol.% of water was injected into the fuel at the content of 0, 0.5, 1.0 vol.%. An autoclave was used in the immersion tests to isolate the corrosion system from the surroundings and withstand the high internal pressures induced by the elevated temperatures of the fuel.

2.3. EIS measurements

The effects of dissolved oxygen, water and acetic acid in the fuel mixture on aluminum corrosion were evaluated by electrochemical impedance spectroscopy (EIS). Owing to the extremely low conductivity of the fuel, the EIS measurements were carried out in a separated 3-electrode electrochemical cell after the immersion tests in an autoclave. The electrolyte selected for the EIS measurements was an aqueous solution containing 3 wt.% Na₂SO₄, which retards the further progress of aluminum corrosion and has a sufficiently high conductivity for the electrochemical tests [22]. The cell system was linked electrically to an impedance analyzer. A sinusoidal voltage of ±10 mV was applied over the frequency range, 100 kHz–0.001 Hz. An equivalent circuit that simulates the electrochemical reactions at the oxide film and the alloy surface was introduced to interpret the measured impedance data [23]. The circuit was composed of a resistor associated with the solution resistance (*R_s*) in series with two parallel RC circuits composed of the following elements: a capacitance of the oxide film (*C_{film}*) and an oxide film resistance (*R_{film}*) resulting from the formation

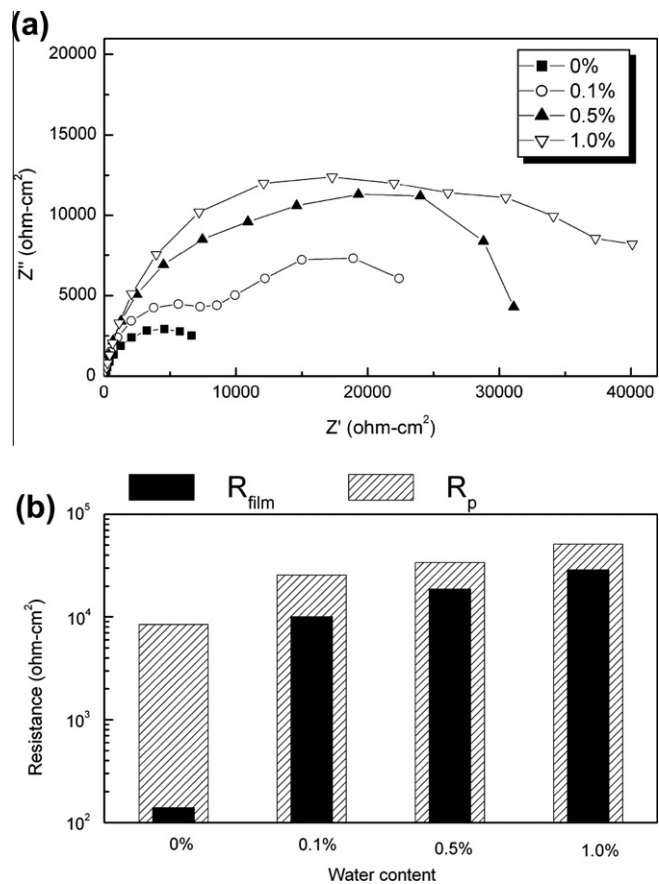


Fig. 4. Results of EIS measurements for the aluminum alloy immersed in oxygen-free E20 fuel with various water contents: (a) Nyquist plots and (b) calculated oxide film and polarization resistance.

of ionic conduction paths through defects in the film in the first RC circuit, and double layer capacitance (*C_{dl}*) and charge transfer

resistance (R_{ct}) between the electrolyte and metal surface in the latter. In the fitting process, the capacitances were replaced with constant phase elements (CPEs) for the best fit to allow for deviations from the ideal dielectric behavior related to surface heterogeneity [24,25]. The corroded surface of the specimens was observed after the corrosion tests.

3. Results and discussion

3.1. Corrosion properties as a function of the oxygen concentration

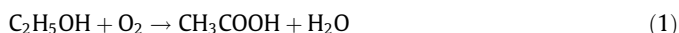
Fig. 1 shows the corroded surfaces of aluminum alloy after the immersion tests for 24 h at 100 °C in the modified oxygen-concentrated E20 fuels. A few localized pits were initiated after immersion in oxygen-enriched fuel. However, more pits were expanded with decreasing oxygen concentration in the fuel. In particular, severe damage developed in uniform corrosion aspect in the oxygen-free fuel. The corrosion, which was accompanied by the complete dissolution of the aluminum matrix, propagated to the inside of

the specimen, leaving a very weak porous structure composed mainly of platelet eutectic silicon phases.

Fig. 2 shows the results of the EIS measurements after the immersion tests in various oxygen-concentrated fuels. The impedance of the specimen immersed in oxygen-free fuel was not measurable due to complete dissolution of the aluminum matrix. The film resistance (R_{film}) and polarization resistance ($R_p = R_{film} + R_{ct}$) calculated from the Nyquist plots increased dramatically with increasing oxygen concentration in the fuel, as shown in Fig 2(b). The distinct increase in R_{film} indicates that the dissolved oxygen in the fuel promotes the formation of an oxide film on the surface that increases the corrosion resistance of the aluminum alloy.

3.2. Ethanol oxidation reaction in bio-ethanol-blended fuel

Ethanol can be oxidized by the dissolved oxygen in the ethanol-blended fuel, producing 1 mol of acetic acid and water according to Eq. (1) [26,27].



The evidence of ethanol oxidation in the fuel was confirmed by Karl Fisher analysis of water and a methyl orange indicator for acetic acid after heating the ethanol-blended fuel to 100 °C. Karl Fisher water analysis as a function of the oxidation time resulted in 0, 0.34 and 0.72 vol.% at 0, 2 and 12 h, respectively. The water content increased with increasing oxidation time. This indicates that a fraction of ethanol in the fuel was oxidized into acetic acid and water, and each of them might affect the corrosion of aluminum alloy. Acetic acid might cause the aluminum alloy to corrode according

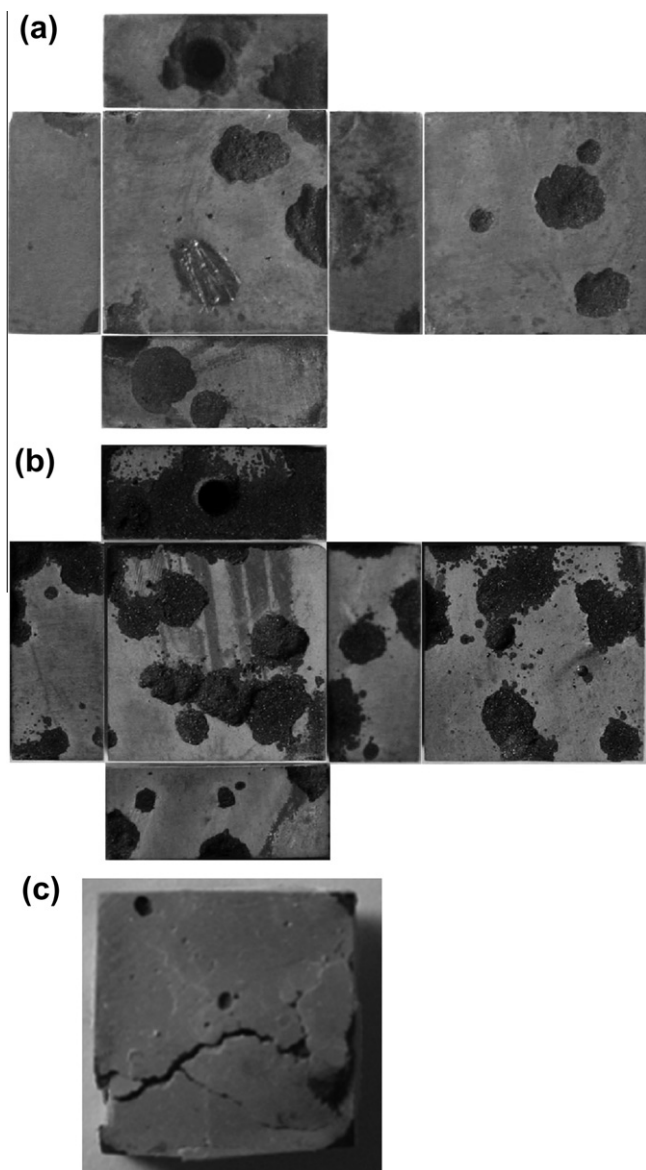


Fig. 5. Corroded surfaces of the aluminum alloy after the immersion tests at 100 °C for 6 h in oxygen-free E20 fuel with various acetic acid contents at (a) 0.1%, (b) 0.5%, and (c) 1.0%.

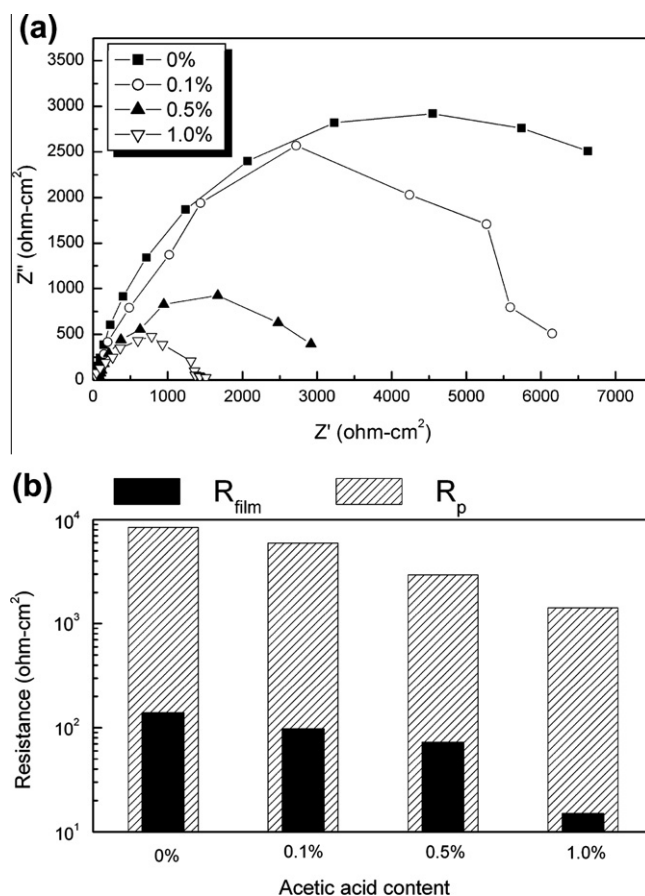
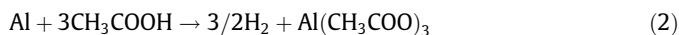
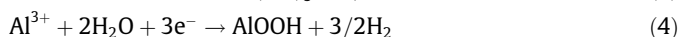
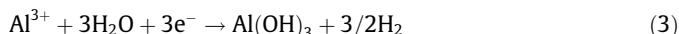


Fig. 6. Results of EIS measurements for the aluminum alloy immersed in oxygen-free E20 fuel with various acetic acid contents: (a) Nyquist plots and (b) calculated oxide film and polarization resistance.

to the chemical reaction shown in Eq. (2). In an acetic acid environment, the aluminum alloy corrodes by substitution between an aluminum atom in the metal or oxide film and a hydrogen atom in the OH group, producing hydrogen gas and aluminum acetate [28].



The water produced in the fuel might affect the corrosion of the aluminum alloy in two ways: facilitate corrosion by creating aqueous corrosion conditions, while mitigating it by contributing to the formation of a protective hydrous oxide surface film such as bayerite (Al(OH)₃) or boehmite (AlOOH) by the reaction in Eqs. (3) and (4) [28].



The effects of these corrosive agents produced from ethanol oxidation on the corrosion properties of the aluminum alloy in the fuel were investigated further.

3.3. The effects of acetic acid and water in bio-ethanol-blended fuel

Acetic acid and/or water were injected artificially into the oxygen-free fuel in which the self-formation of them by ethanol oxidation reaction was suppressed. Fig. 3 shows the corroded surfaces of the aluminum alloy after the immersion tests for 6 h at 100 °C in oxygen-free E20 fuels containing various water levels. The water content in the fuel was 0, 0.1, 0.5 and 1.0 vol.%. In the absence of water in the fuel, significant pitting corrosion developed on the surface (see Fig. 3(a)), whereas the corrosion damage was de-

creased remarkably by introducing a small amount of water. No corrosion damage was observed above a water content of 0.5% (see Fig. 3(c) and (d)). The weight change of the aluminum alloy specimen was measured after the immersion tests, and the results are listed in Table 1. The weight gain due to the formation of a hydrous oxide film on the surface was observed for the specimens immersed in water-containing fuels, which suggests that water promoted the formation of a hydrous oxide film on the surface

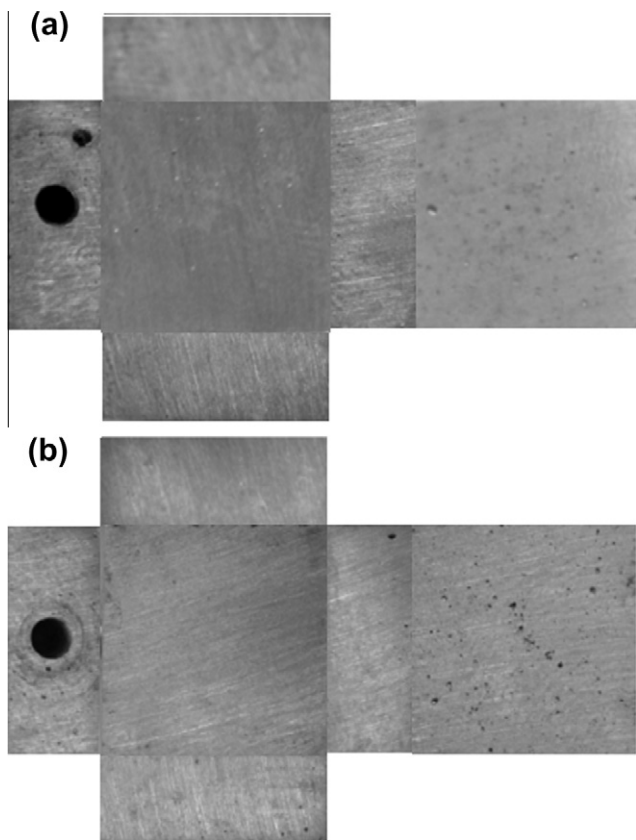


Fig. 7. Corroded surfaces of the aluminum alloy after the immersion tests at 100 °C for 6 h in oxygen-free E20 fuel with various acetic acid–water mixture contents at (a) 0.5% and (b) 1.0%.

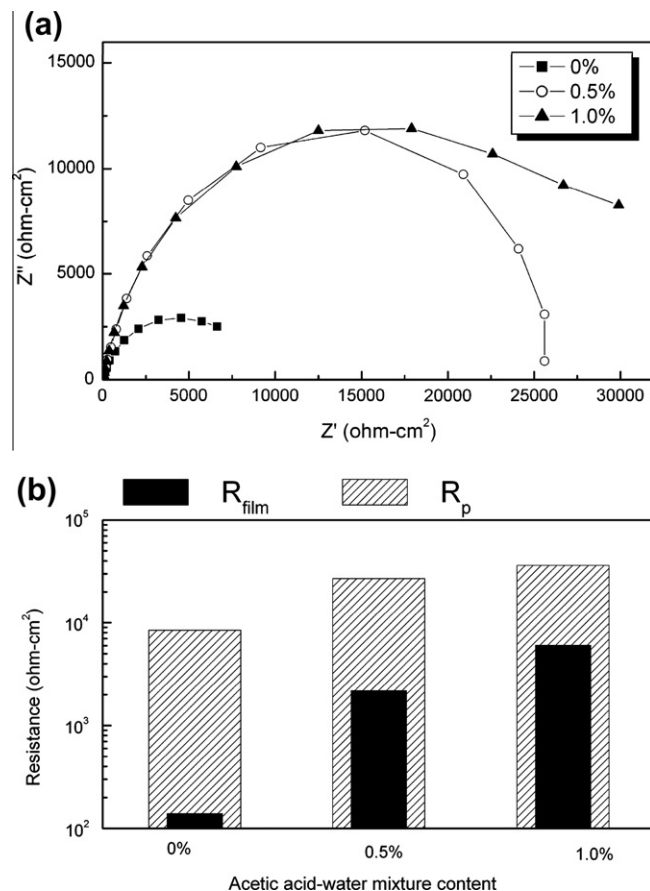


Fig. 8. Results of EIS measurements for the aluminum alloy immersed in oxygen-free E20 fuel with various acetic acid–water mixture contents: (a) Nyquist plots and (b) calculated oxide film and polarization resistance.

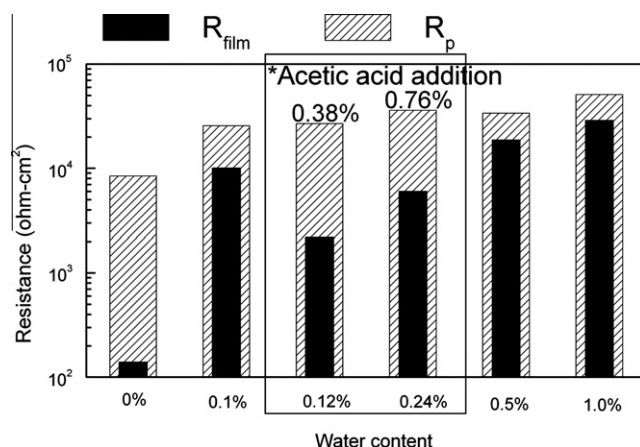


Fig. 9. Comprehensive diagram for the effects of water and acetic acid on the oxide film and polarization resistance of aluminum alloy in the E20 fuel.

[29]. The weight gain increased with increasing water content in the fuel. Fig. 4 shows the results of the EIS measurements for the specimens immersed in the water-added E20 fuel. The Nyquist plots were interpreted as the qualitative values of R_{film} and R_p , as shown in Fig. 4(b). R_{film} increased remarkably with the addition of water to the fuel and increased further with increasing water content. The distinct increase in R_{film} in the presence of water suggests that water promotes the formation of a hydrous oxide film on the surface, which increases the corrosion resistance of the aluminum alloy in the fuel. Furthermore, the increasing water content thickened the film, contributing to enhanced corrosion resistance. These results confirmed that the water acts as a corrosion inhibitor by promoting the formation and growth of a hydrous oxide film in the bio-ethanol blended gasoline fuel.

Subsequently, the effects of acetic acid on the corrosion properties of the aluminum alloy were examined by immersion tests for 6 h at 100 °C in an acetic acid-added oxygen-free E20 fuel. The acetic acid content in the fuel was 0, 0.1, 0.5 and 1.0 vol.%. Fig. 5 shows the corroded surfaces after the immersion tests as a function of the acetic acid content, and that for 0% is the same as that shown Fig 5(a). The surfaces reveal that the acetic acid considerably accelerated the corrosion of the aluminum alloy to propagate to the entire exposed surface in the fuel with 1.0 vol.% acetic acid. Fig. 6 shows the results of the EIS measurements for the specimens immersed in the acetic acid-added E20 fuel. R_{film} and R_p decreased gradually with the increasing acetic acid content. The decrease in R_{film} is considered to result from the contribution of the acetic acid to the weakening of the hydrous oxide film formed on the aluminum surface by dehydration and altering the structure, as well as the chemical reaction in Eq. (2) [28]. These results confirm that acetic acid accelerates the pitting corrosion of the aluminum alloy in the bio-ethanol blended gasoline fuel by weakening the protective film and the further propagation according to Eq. (2).

From the above results, the by-products from ethanol oxidation, water and acetic acid, have the opposite effects as an inhibitor and activator on the corrosion of aluminum alloy, respectively. In practice, these agents should coexist with the same mole fraction in the fuel according to Eq. (1), assuming the isolated system from surrounding. Therefore, the combined effect of these products was investigated by immersion tests in oxygen-free E20 fuel containing an acetic acid–water mixture. The solution consisted of the same mole fraction of water and acetic acid, thereby 24% and 76% in volume fraction, respectively. Fig. 7 shows the corroded surfaces of the aluminum alloy after immersion tests for 6 h at 100 °C in the fuels containing the acetic acid–water mixture contents of 0.5 and 1.0 vol.%. The image for 0% is the same as that shown in Fig 3(a). The corrosion damage by pitting decreased remarkably with the addition of the acetic acid–water mixture because only small pits were observed, as shown in Fig. 7(a), even though the corrosive acetic acid was incorporated in the fuel. A complete intact surface was observed with further increases in the concentration of the solution (see Fig. 7(b)). This suggests that the protective effect of water inhibits the corrosive effect of acetic acid, where the two agents coexisted. This was confirmed by the EIS measurements shown in Fig. 8. R_{film} and R_p increased significantly with the addition of the acetic acid–water mixture as shown in Fig. 9(b), which was attributed to the dominant water effect of promoting the growth of hydrous oxide film.

Fig. 9 summarizes the integrated effects of water and acetic acid on the corrosion of the aluminum alloy in bio-ethanol blended gasoline fuel by combining the results of EIS measurements in Figs. 4(b) and 8(b) as a function of the added water content. It is indisputable that acetic acid has a detrimental effect on the protective oxide film, i.e., decrease in R_{film} , thereby decreasing the corrosion resistance, compared to the resistances at a water content of 0.1% with 0.12%. However, the resistances were enhanced reason-

ably by the small amount the water, even though the corrosive acetic acid had been incorporated in the fuel. The resistances increased with further increasing water content.

Consequently, the enhanced corrosion resistance by the dissolved oxygen in the bio-ethanol-blended fuel was attributed to the protective hydrous oxide film promoted by water as a by-product of ethanol oxidation. Although acetic acid had a detrimental effect on the oxide film and the alloy matrix, the protective effect of water was superior to the corrosive effect of acetic acid. The water content in the fuel increased with increasing dissolved oxygen concentration, leading to high corrosion resistance of aluminum alloy in oxygen-enriched fuel.

4. Conclusions

The effects of dissolved oxygen on the corrosion of aluminum alloy in the high temperature E20 fuel were examined by electrochemical tests and surface analyses. The corrosion resistance increased with increasing dissolved oxygen concentration in the fuel. The enhanced corrosion resistance by dissolved oxygen was caused by competition between the by-products of ethanol oxidation. Among the by-products, water had a beneficial effect on the corrosion resistance of aluminum alloy by promoting the growth of a protective hydrous oxide film, whereas acetic acid had a detrimental effect by deteriorating the oxide film and alloy matrix. The corrosion resistance of aluminum alloy was determined by the predominant effect of water as a corrosion inhibitor, even though it coexisted with the corrosive acetic acid. The water content increased with increasing dissolved oxygen concentration in the fuel. Therefore, the corrosion resistance of the aluminum alloy was increased in oxygen-enriched fuel.

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