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ORIGINAL ARTICLE

Electrochemical Investigation of Ethylene Glycol as Corrosion Inhibitor of Aluminum 5052

I. Habibi[1], A. Nikitasari[2,3*], Saifudin[1], R. Kusumastuti[3], S. Musabikha[3], S. Prifiharni[3], J. Irawan[3], G. Priyotomo[3], and R. Riastuti[2*]

[1]Engineering Faculty, Universitas Muhammadiyah Magelang, Indonesia
[2]Metallurgical and Material Engineering Department, Engineering Faculty, Universitas Indonesia, Indonesia
[3]Research Center for Metallurgy, National Research and Innovation Agency (BRIN), Indonesia

ABSTRACT – Nowadays, electric cars use Aluminum (Al) 5052 in the cooling pad of the battery coolant system. Corrosion behavior study of Al 5052 by coolant is essential in the automotive industry. Ethylene glycol is the main compound of the battery coolant system that can inhibit the corrosion process of aluminum alloys. The corrosion inhibition performance of ethylene glycol (EG) for Al 5052 surface in 3.5% NaCl solution has been studied using electrochemical measurements such as potentiodynamic polarization and electrochemical impedance spectroscopy (EIS) in various concentrations of EG up to 50% and a temperature range from 30°C to 60°C. Based on the results, EG can inhibit the corrosion process of Al 5052. The corrosion rate (CR) of coolant containing EG decreases with increasing that concentration in various temperatures, and the CR of the coolant rises with the increased temperatures. The inhibition efficiency of coolant boosts with the increase in EG concentration in each operational temperature, and the highest is up to 75% at 60°C with 50% of EG concentration.

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INTRODUCTION

An electric car uses electrical energy stored in batteries or energy storage. Electric cars have several advantages compared to fossil fuel cars; among others, does not produce air pollution, save energy, and do not make noise [1]–[3]. One of the critical components of an electric car is the battery electricity. Currently, electric cars are driven by electric motors using stored electrical energy in lithium batteries. The performance of this battery is affected by its operating temperature, where its performance will decrease rapidly with increasing heat, so the temperature should be maintained between 0°C– 45°C (charging) and -20°C–60°C (discharging) using a battery coolant system [4].

Aluminum (Al) 5052 is a material commonly used as a component in the battery cooling system of electric cars [5]. The properties of Al 5052, such as good weldability and high corrosion resistance, make this alloy the most economical and attractive for a wide variety of uses in the automotive industry [6], [7]. Generally, Al 5052 resists corrosion due to the presence of a protective oxide layer. But, when chloride ions are present in the solution, the protective oxide layer on the Al 5052 surface is easily destroyed, leading to localized corrosion [8], [9].

The inhibition of Al 5052 corrosion was extensively studied using organic and inorganic compounds [8]. Currently, the main composition of battery coolant is 30–70%vol. ethylene glycol [9], [10]. Therefore, ethylene glycol, as a major content of the battery coolant, is expected to act as a corrosion inhibitor of Al 5052. Previous work results show that polyethylene glycol can inhibit the corrosion of aluminum at room temperature [11]. Ethylene glycol, produced by the reaction of ethylene oxide with water, was used as a coolant because of its high boiling point and flash point, which reduces the risk of coolant loss when the cooling system undergoes overheating. Moreover, the vapors from a hot glycol solution contain mostly water so that the antifreeze base is not lost by evaporation, and freezing protection is maintained. Unfortunately, some literature reported that ethylene glycol oxidizes at temperatures above 80°C to a mixture of corrosive acids, which are formic acid and oxalic acid [12]. However, the maximum temperature of an electric car's battery is only 60°C. Hence, the research about the effect of ethylene glycol on the corrosion behavior of Al 5052 at 60°C and below is essential to conduct. Most studies focused on the corrosion of aluminum alloys in chloride solution [13], [14]. However, very few investigations are available on ethylene glycol, which is significant in the automotive industry. Hence, in the present study, the effect of ethylene glycol in chloride solution was analyzed on the corrosion behavior of Al 5052 using inhibitor efficiency calculation. The efficiency of ethylene glycol as a corrosion inhibitor can be calculated using Equation (1) [15], [16].

$$Efficiency\ (\%) = \frac{CR\ 0\% - CR\ n\%}{CR\ 0\%} \times 100 \tag{1}$$

***CORRESPONDING AUTHOR** | A. Nikitasari & R. Riastuti | ✉ waltermellon@batan.go.id

where corrosion rate (CR) 0% is the corrosion rate without ethylene glycol, and CR n% is the corrosion rate with ethylene glycol [12].

EXPERIMENTAL METHOD

Materials and Instruments

A commercial product of Aluminum (Al) 5052 from the marketplace was used in this study. The chemical composition of Al 5052 was analyzed using optical emission spectroscopy (OES), and the results are listed in Table 1. Sodium chloride and ethylene glycol used in this experiment were pure analysis grades from Merck. Electrochemical measurements were carried out using Gamry Instrument G750.

Table 1. Chemical composition of the specimen

Element (%)						
Al	Mg	Si	Fe	Mn	Zn	Cr
97	2.4	0.08	0.3	0.02	0.03	0.2

Method and Procedure

The specimen under study, Al 5052, was annealed at 343°C for 1 hour and then cooled in a muffle furnace. The sample was divided into small coupons 3 mm × 10 mm × 10 mm. Each test coupon was welded to an electrical wire and mounted in epoxy resin to get a constant rectangular exposed area (1 cm^2). This exposed area was polished using emery papers of different grades (200–1200). After being polished with emery papers, the specimen was washed with distilled water and degreased with acetone.

The test solution was prepared using 3.5%vol. of sodium chloride and various ethylene glycol concentrations (0%, 10%, 20%, 30%, and 50%). Aquadest was also used as a blank in this experiment. All the experiments were conducted in a calibrated thermostat at temperatures of 30°C, 40°C, 50°C, and 60°C under unstirred conditions.

Electrochemical measurements were carried out using the Gamry G750 corrosion measurement system with a three-electrode set-up. Al 5052 specimen, a platinum mesh plate, and a saturated calomel electrode were used as the working electrode (WE), counter electrode (CE), and reference electrode (RE), respectively. For the open circuit potential (OCP), measurements were first done to ensure that a reasonably stable state was reached, where the OCP drift was less than 1 mV per minute, and the magnitude of the OCP fluctuation was less than 1 mV. The OCP measurements were immediately followed by potentiodynamic polarization. Potentiodynamic polarization measurements were performed via sweeping potential from -250 mV versus OCP to +250 mV versus OCP with a scan rate of 1 mV/s.

Electrochemical impedance spectroscopy (EIS) measurement was established using the same instrument with potentiodynamic polarization. EIS was carried out at the open circuit potential by applying a periodic amplitude of 10 mV and frequencies from 100 kHz to 0.01 Hz. The Bode plot and Nyquist plot were used to analyze the impedance data.

The visual of surface specimens after electrochemical measurement was observed in all concentrations of ethylene glycol and blank for comparison. Furthermore, the pH of 50% ethylene glycol at 30°C–110°C was measured to investigate the corrosive acid formed by ethylene glycol.

RESULT AND DISCUSSION

Potentiodynamic Polarization

The potentiodynamic polarization curves of Aluminum (Al) 5052 in 3.5% NaCl containing various concentrations of aqueous ethylene glycol at different solution temperatures are shown in Figure 1 to Figure 4. Figure 5 depicts the potentiodynamic polarization curves of Al 5052 in aquadest at different solution temperatures.

From Figure 1 to Figure 4, it is clear that curves exhibit nobler corrosion potential for a concentration of 50% of ethylene glycol at 30°C (Figure 1). At 40°C and 50°C solution temperatures (Figure 2) and (Figure 3), the curves have almost the same corrosion potential. The curves are shifted to the more positive value of corrosion potential with the increase of ethylene glycol concentration at 60°C (Figure 4).

Based on Figure 1 to Figure 4, the potentiodynamic polarization curves are shifted to higher current density with the increase in ethylene glycol concentration. It is evident that ethylene glycol inhibits the corrosion process of Al 5052 at 30°C–60°C. From Figure 5, corrosion potential (E_{corr}) decreases up to 60 mV, and current density (I_{corr}) increases with the increasing of temperatures. This result reveals that temperature can accelerate the corrosion process of Al 5052 in the absence of ethylene glycol.

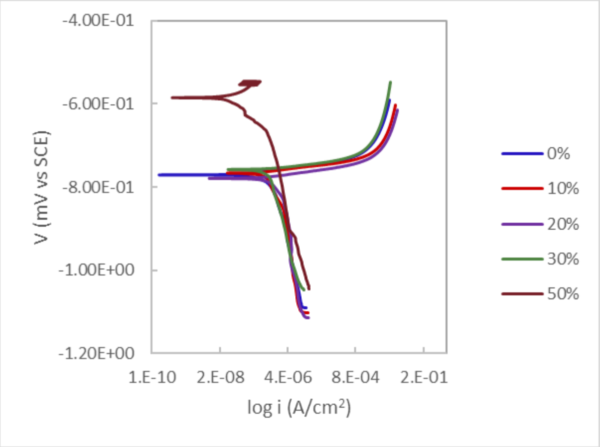

Figure 1. Potentiodynamic polarization curves of Al 5052 in 3.5% NaCl containing various concentrations of ethylene glycol at 30°C

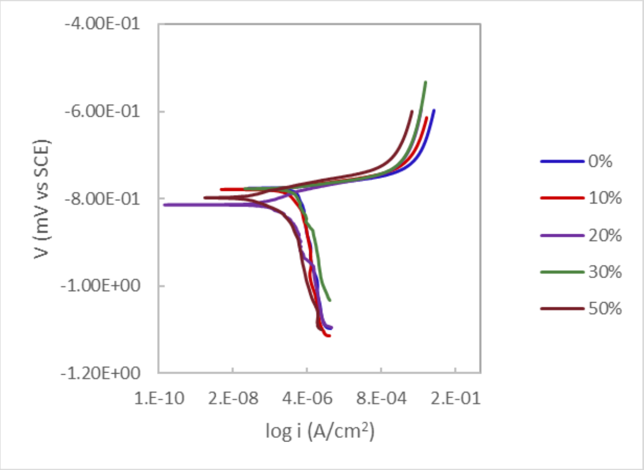

Figure 2. Potentiodynamic polarization curves of Al 5052 in 3.5% NaCl containing various concentrations of ethylene glycol at 40°C

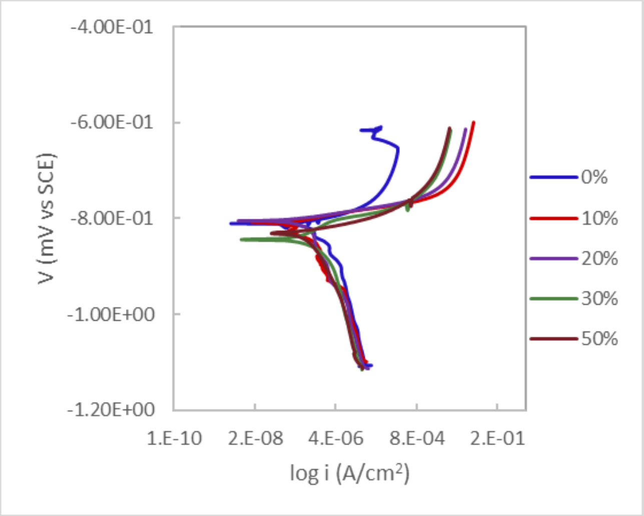

Figure 3. Potentiodynamic polarization curves of Al 5052 in 3.5% NaCl containing various concentrations of ethylene glycol at 50°C

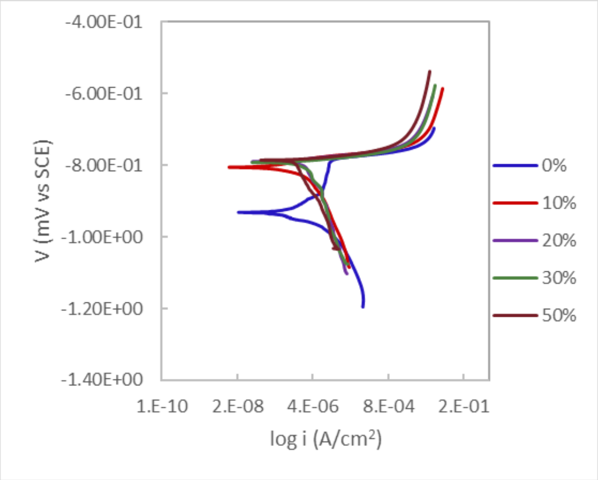

Figure 4. Potentiodynamic polarization curves of Al 5052 in 3.5% NaCl containing various concentrations of ethylene glycol at 60°C

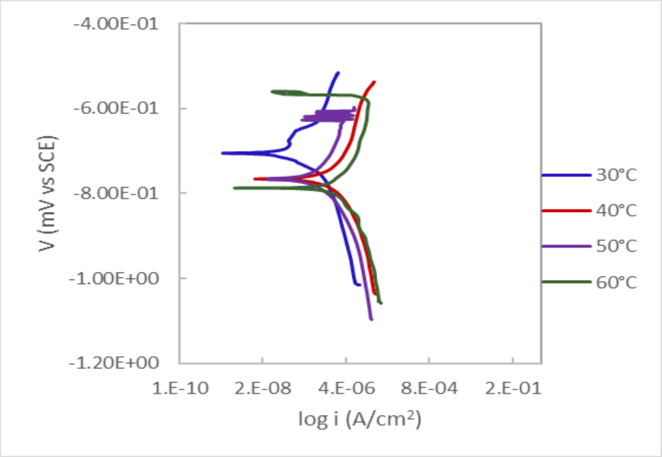

Figure 5. Potentiodynamic polarization curves of Al 5052 in aquadest at different solution temperatures.

The potentiodynamic polarization parameters such as corrosion potential (E_{corr}), current density (I_{corr}), cathodic slope (b_c), and corrosion rate (CR) are tabulated in Table 2. These parameters are obtained by extrapolating the linear Tafel region of the potentiodynamic polarization curves to the OCP. According to Table 2, the corrosion rate decreases with the increase in ethylene glycol concentration. This is attributed to the adsorption of ethylene glycol on the Al 5052 surface. Ethylene glycol reacts in anodic sites of electrochemical reaction and suppresses the anodic dissolution, Equation (2) and Equation (3) [17].

$$Anodic: \ Al \rightarrow Al^{3+} + 3e^{-} \tag{2}$$

$$Al^{3+} + 3(HO - CH2CH2 - O-) \rightarrow Al(HOCH2CH2O)_3 \tag{3}$$

The product $Al(HOCH_2CH_2O)_3$ has an inhibition effect on the Al 5052 corrosion process. Therefore, ethylene glycol is an anodic type of inhibitor indicated by increasing the corrosion potential with the increase in concentration. The maximum efficiency of ethylene glycol in inhibiting corrosion of Al 5052 is 76%. The efficiency of ethylene glycol is boosted with the concentration of ethylene glycol. At a solution temperature of 60°C, 10% ethylene glycol can directly increase the efficiency to 50%. This result indicated that temperature can accelerate the anodic reaction of ethylene glycol to $Al(HOCH_2CH_2O)_3$.

Table 2. Potentiodynamic polarization parameters

Temperature (°C)	Solution	b_c (V/dec)	E_{corr} (mV)	I_{corr} (A/cm^2)	CR (mpy)
30	Aquadest	3.04E-01	-705	8.49E-07	1.101
	0% Ethylene Glycol	4.50E-01	-771	1.89E-06	2.445
	10% Ethylene Glycol	4.21E+02	-766	1.83E-06	2.367
	20% Ethylene Glycol	2.61E-01	-878	1.22E-06	1.585
	30% Ethylene Glycol	3.24E-01	-757	1.12E-06	1.453
	50% Ethylene Glycol	2.67E-01	-585	4.47E-07	0.580
40	Aquadest	1.86E-01	-767	2.18E-06	2.822
	0% Ethylene Glycol	4.55E-01	-779	2.57E-06	3.327
	10% Ethylene Glycol	4.66E-01	-779	2.47E-06	3.205
	20% Ethylene Glycol	4.25E-01	-815	1.53E-06	1.978
	30% Ethylene Glycol	1.60E-01	-777	1.46E-06	1.892
	50% Ethylene Glycol	3.62E-01	-797	1.17E-06	1.522
50	Aquadest	3.10E-01	-767	2.48E-06	3.218
	0% Ethylene Glycol	2.92E-01	-811	2.71E-06	3.520
	10% Ethylene Glycol	3.03E-01	-808	2.37E-06	3.067
	20% Ethylene Glycol	2.61E-01	-805	1.63E-06	2.119
	30% Ethylene Glycol	2.82E-01	-844	2.14E-06	1.968
	50% Ethylene Glycol	2.12E-01	-831	1.24E-06	1.606
60	Aquadest	3.17E-01	-788	5.67E-06	7.35
	0% Ethylene Glycol	1.57E-01	-931	8.09E-06	10.500
	10% Ethylene Glycol	2.29E-01	-806	3.71E-06	4.804
	20% Ethylene Glycol	2.84E-01	-791	3.57E-06	4.633
	30% Ethylene Glycol	2.57E-01	-793	3.31E-06	4.297
	50% Ethylene Glycol	2.26E-01	-785	1.92E-06	2.488

The corrosion rate of Al 5052 in aquadest is lower than that in 3.5% NaCl without ethylene glycol because of the absence of ion chloride in the aquadest solution. However, ethylene glycol can inhibit ion chloride attack. Therefore, the corrosion rate in the presence of ethylene glycol is lower than that in aquadest.

Electrochemical Impedance Spectroscopy

The corrosion behavior of Al 5052 in inhibited and uninhibited solutions of various concentrations of ethylene glycol was also investigated by electrochemical impedance spectroscopy. The impedance responses of Al 5052 in 3.5% NaCl with various concentrations of ethylene glycol are represented in the form of the Nyquist plot (Figure 6 to Figure 9). The Nyquist plot of Al 5052 in the aquadest solution is shown in Figure 10. It has been observed that the impedance response, as in the Nyquist plot, shows a semicircle capacitive loop. By increasing the concentration of ethylene glycol, an increment of the diameter of the semicircle appears in the Nyquist plot. This may be attributed to the increase in ethylene glycol inhibition power [18].

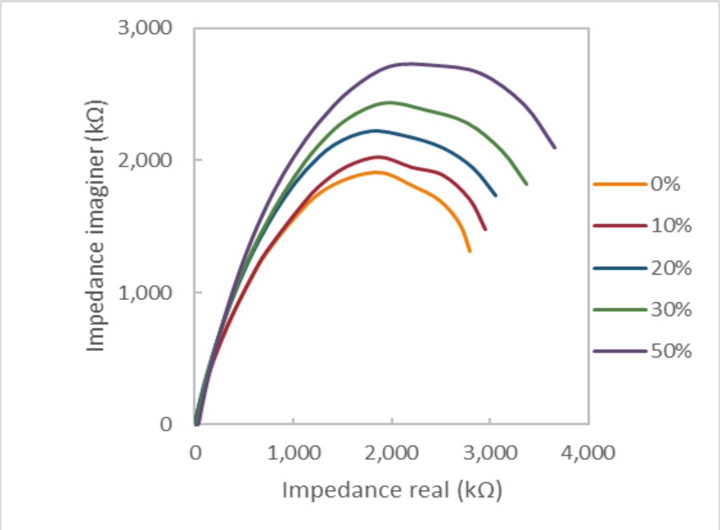

Figure 6. Nyquist plot of Al 5052 in 3.5% NaCl containing various concentrations of ethylene glycol at 30°C

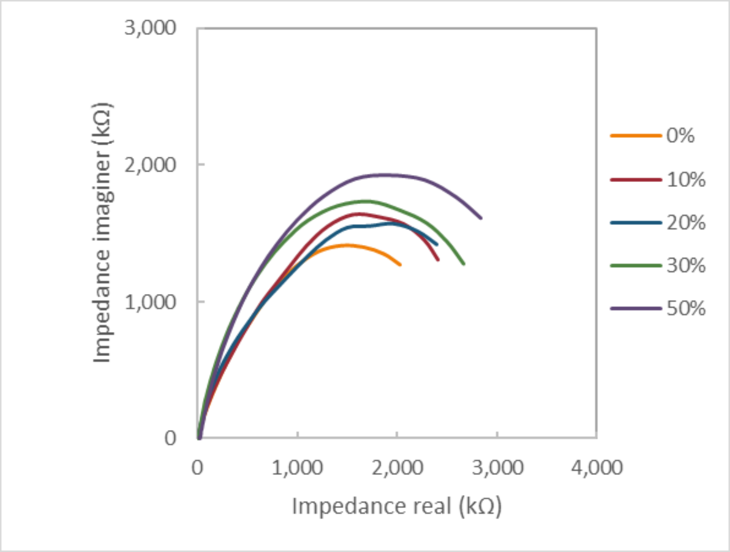

Figure 7. Nyquist plot of Al 5052 in 3.5% NaCl containing various concentrations of ethylene glycol at 40°C

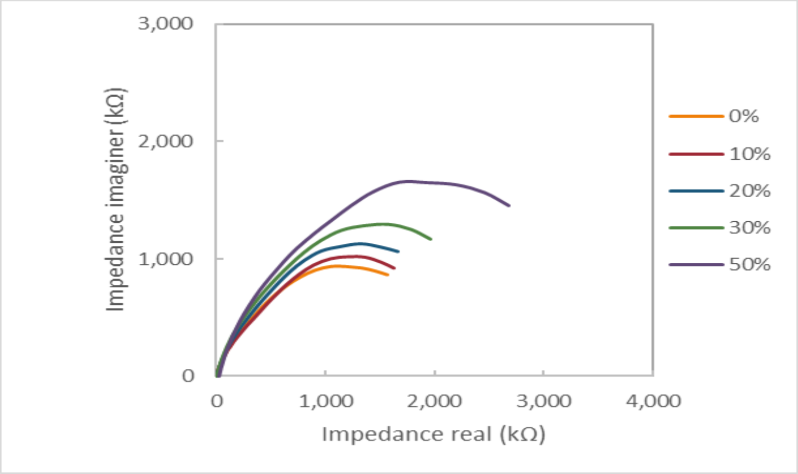

Figure 8. Nyquist plot of Al 5052 in 3.5% NaCl containing various concentrations of ethylene glycol at 50°C

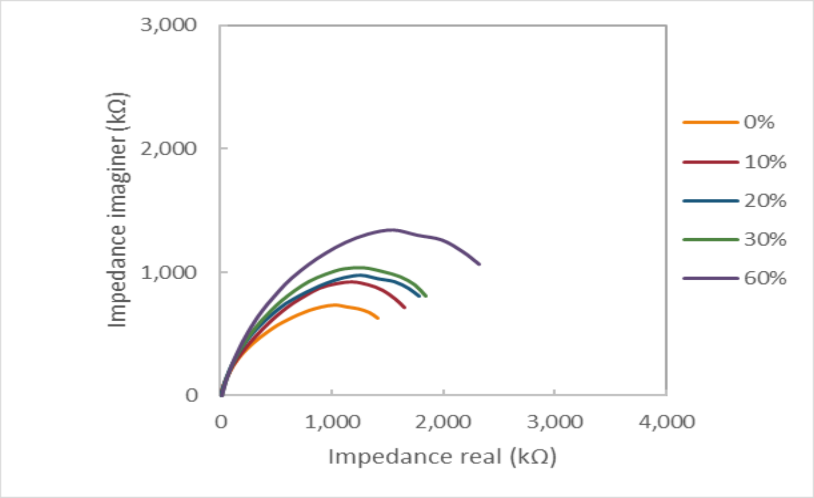

Figure 9. Nyquist plot of Al 5052 in 3.5% NaCl containing various concentrations of ethylene glycol at 60°C

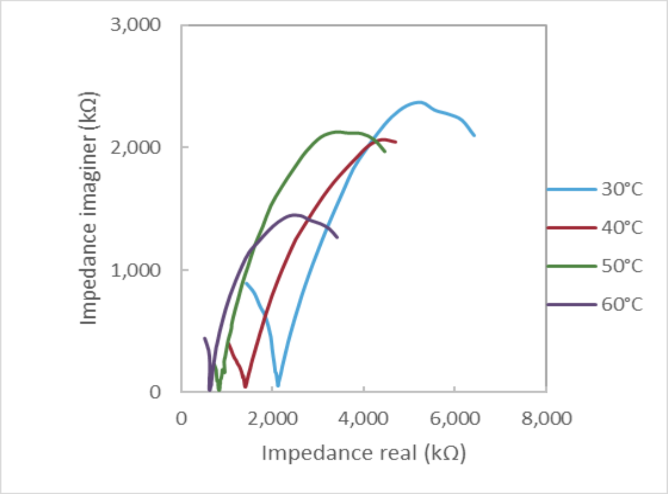

Figure 10. Nyquist plot of Al 5052 in aquadest at different solution temperatures

The semicircle capacitive loop of the Nyquist plot in Figure 6 to Figure 10 suggests that the corrosion process is governed by charge transfer without the contribution of the diffusion process to the total impedance [19], [20]. There is a difference between the Nyquist plot between Figure 6–Figure 9 and Figure 10. Nyquist plot in 3.5% NaCl (Figure 6–Figure 9) establishes a one-time constant due to chloride inducement. At the same time, the Nyquist plot in aquadest (Figure 10) has two times constants. Based on Figure 10, a decrease in temperature outcomes in the larger diameter of the semicircle capacitive loop in the Nyquist plot. This finding is appropriate for the potentiodynamic polarization result mentioned before.

The Bode plot illustrates the impedance and phase angle values of specimens at each frequency. Figure 11 to Figure 14 depict the Bode plot of Al 5052 in 3.5% NaCl with various concentrations of ethylene glycol at 30°C, 40°C, 50°C, and 60°C, respectively. The inspection of the figure shows that there is only one phase maximum impedance, which suggests that the charge transfer controlled the corrosion process at the metal-solution interface [21]. Furthermore, the maximum impedance increases with increasing the concentration of ethylene glycol due to the formation of a protective layer on the metal surface [22], [23].

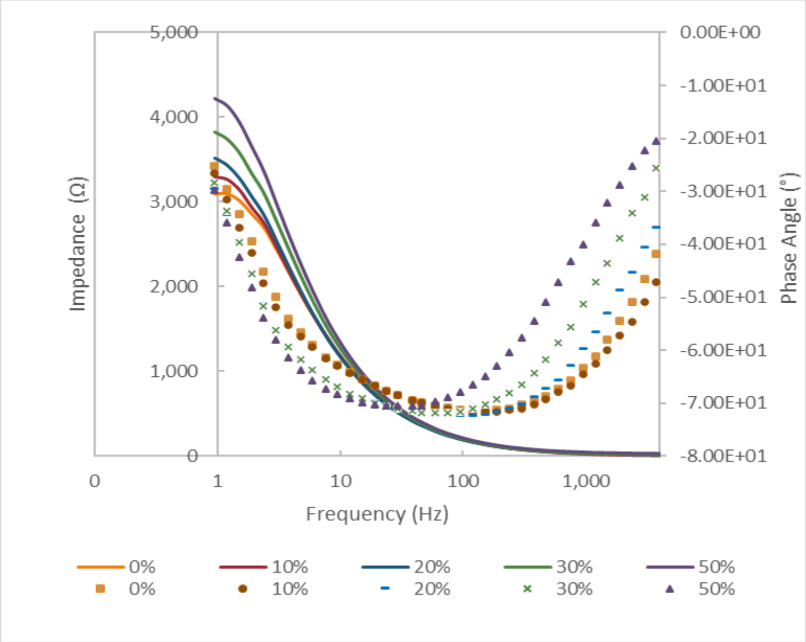

Figure 11. Bode plot of Al 5052 in 3.5% NaCl containing various concentrations of ethylene glycol at 30°C

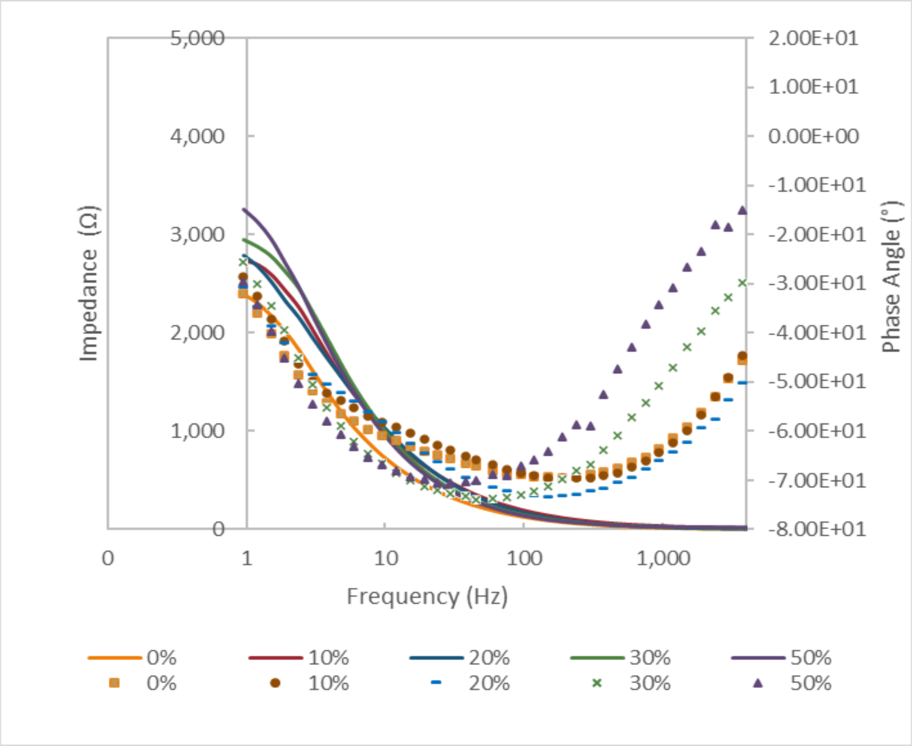

Figure 12. Bode plot of Al 5052 in 3.5% NaCl containing various concentrations of ethylene glycol at 40°C

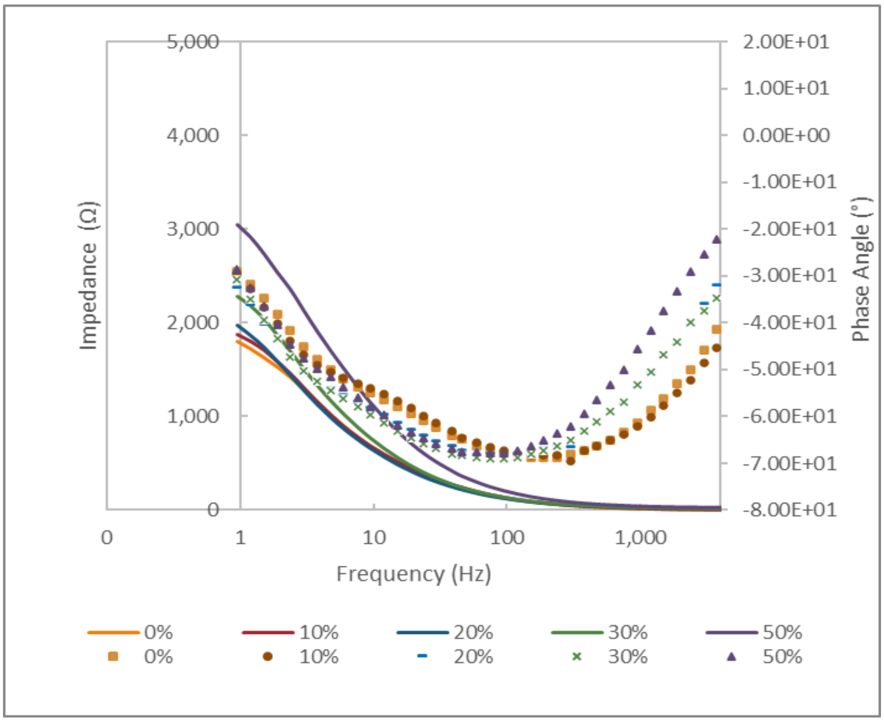

Figure 13. Bode plot of Al 5052 in 3.5% NaCl containing various concentrations of ethylene glycol at 50°C

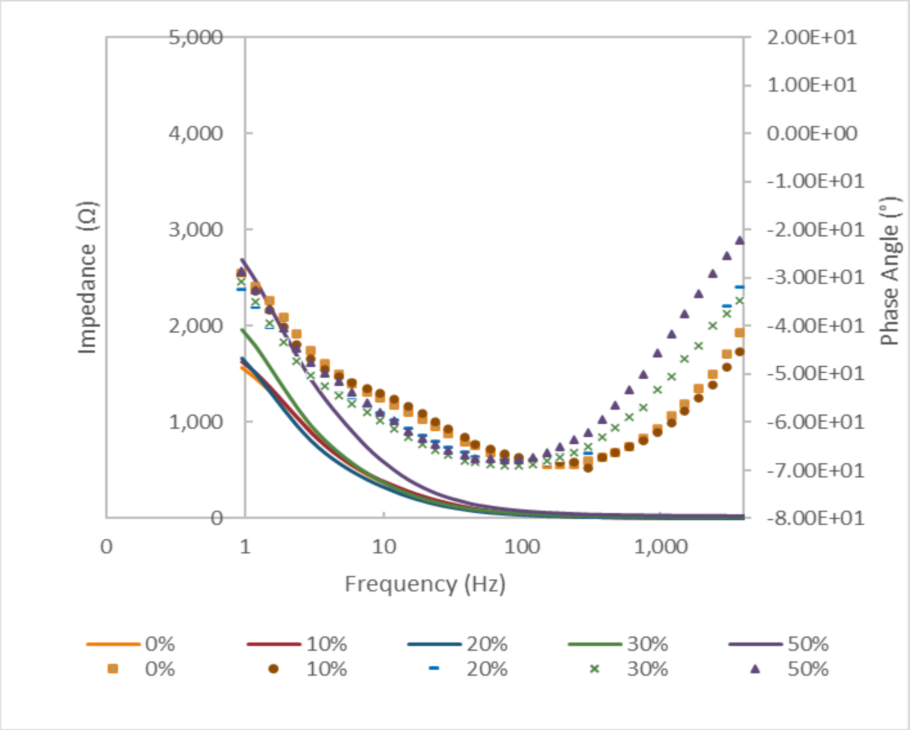

Figure 14. Bode plot of Al 5052 in 3.5% NaCl containing various concentrations of ethylene glycol at 60°C

The electrochemical impedance spectroscopy (EIS) curves of both Bode and Nyquist plot in the high frequency relate to the resistance between solution and Al 5052, while that in the low frequency suggests the charge transfer resistance of the corrosion process. To obtain the ideal charge transfer resistance, the Nyquist plot was analyzed with the equivalent electrical circuit shown in Figure 15. Element Rs refers to uncompensated solution resistance, and Rct corresponds to the charge transfer resistance. Meanwhile, Y_0 and alpha are the parameters related to the constant phase element (CPE) [24]–[26].

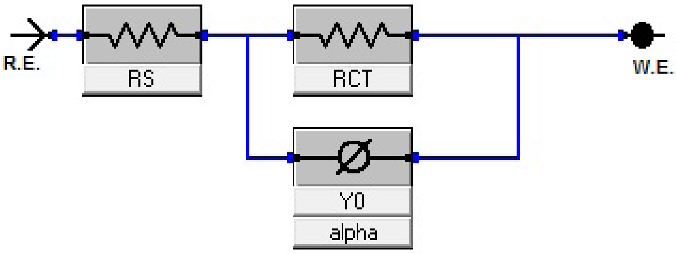

Figure 15. Equivalent electrical circuit of Al 5052 in 3.5% NaCl with various ethylene glycol

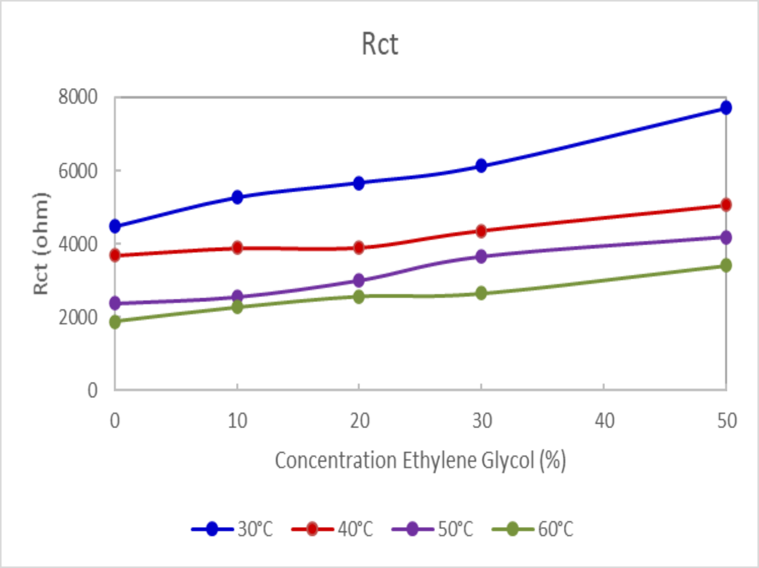

Figure 16. Rct values of Al 5052 in 3.5% NaCl with various concentrations of ethylene glycol at different solution temperatures

Figure 16 shows the Rct value of Al 5052 in 3.5% NaCl with and without ethylene glycol at different temperatures. The results from the Rct parameter (Figure 16) clearly show that the Rct is directly proportional to the concentration of ethylene glycol while inversely proportional to the temperature. The greater values of charge transfer resistance (Rct) correlated with the slower corrosion process. Thus, ethylene glycol can inhibit the corrosion process of Al 5052, according to the EIS results.

Visual Observation and pH test

The visual observation of Al 5052 after electrochemical measurement is presented in Figure 17. It is obvious that the surface of Al 5052 in aquadest is the cleanest of the others because there is no chloride ion. Moreover, the surface of 50% is brighter than the surface of a lower concentration of ethylene glycol. This result confirms the inhibition effect of ethylene glycol.

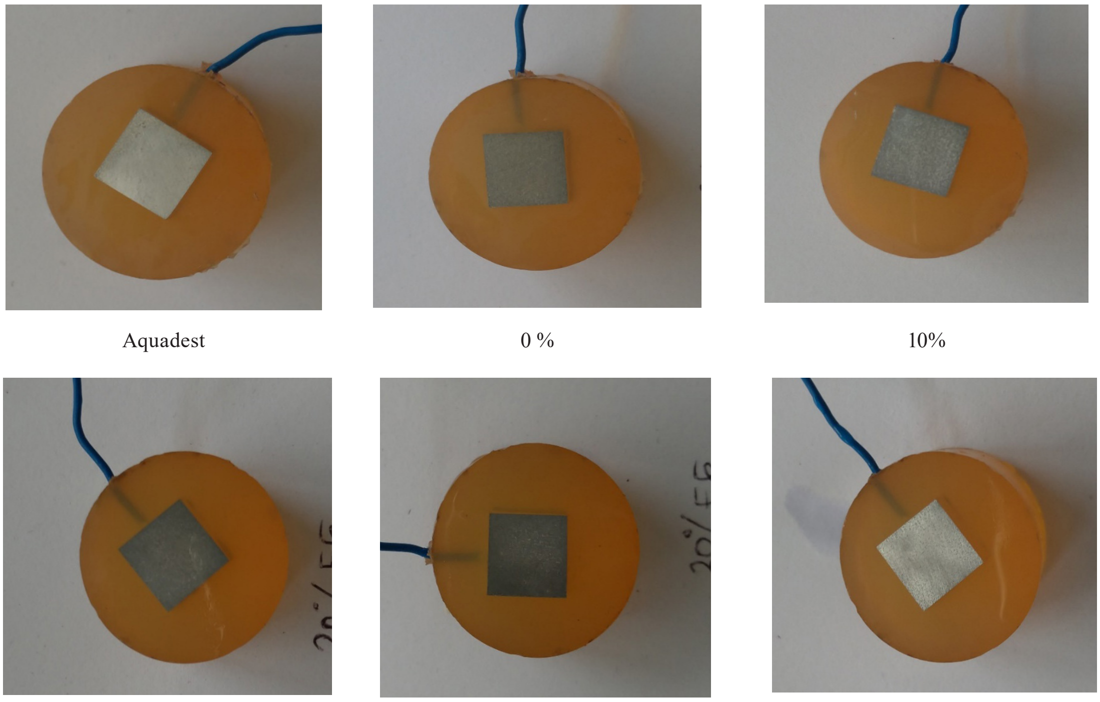
Aquadest
0 %
10%
20%
30%
50%

Figure 17. Visual observation after electrochemical test

The solution containing 50% ethylene glycol was heated and tested using a pH meter and pH test paper to emphasize that in the range of the battery coolant temperature, ethylene glycol can act as a corrosion inhibitor. Figure 18 shows the pH test paper results of the solution at various temperatures. There was no change in the pH until the temperature was 110°C. Therefore, ethylene glycol does not oxidize to the mixture of corrosive acid and acts as a corrosion inhibitor at temperatures 30°C–60°C.

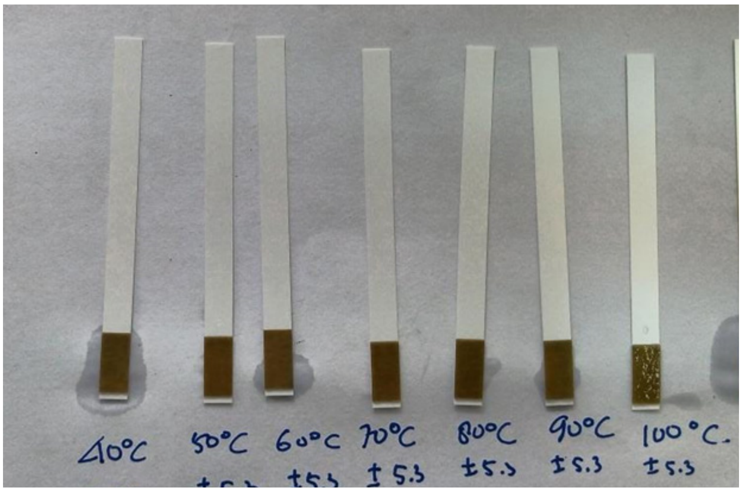

Figure 18. pH test result of 50% ethylene glycol at various temperature

CONCLUSION

The inhibition effect of ethylene glycol on the corrosion behavior of Aluminum (Al) 5052 at 30°C–60°C has been investigated using electrochemical measurement. The inhibition effect of ethylene glycol on the corrosion behavior of Al 5052 depends on the concentration and temperature.

The inhibition efficiency of ethylene glycol increases with the increase in ethylene glycol concentration and temperatures. The corrosion rate of Al 5052 is remarkably influenced by chloride ions. Therefore, the corrosion rate in the blank is lower than in the 3.5% NaCl solution. However, ethylene glycol can inhibit the corrosion rate of Al 5052 in 3.5% NaCl.

Ethylene glycol effectively inhibited the corrosion process of Al 5052 in chloride solution by reacting with aluminum alloy and forming a protective layer on the aluminum surface. Ethylene glycol's role as a corrosion inhibitor of Al 5052 in battery cooling systems has been confirmed by potentiodynamic polarization, electrochemical impedance spectroscopy, and visual observation results.

ACKNOWLEDGEMENT

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