

Electrochemical Performance of Galvanic Cell with Silver Coated Cathode in One Compartment System Using Seawater as Electrolyte

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Abstract

This research was carried out to evaluate the electrochemical performance of a galvanic cell using seawater as an electrolyte. The cell was designed to have a volume of 200 mL and equipped with Zn as an anode and Ag-coated Cu as a cathode (Cu(Ag)-Zn system) in order to suppress the corrosion of Cu. As a comparison, the same experiment with the use of uncoated Cu as a cathode (Cu-Zn system) was also conducted. To conduct the experiment, a system was assembled by connecting 20 cells in series and placed in a closed container filled with seawater. The experiment was run for 72 hours, divided into three 24-hour cycles, by replacing the seawater every 24 hours. The performance of the system was evaluated in terms of open circuit voltage, close circuit voltage, current, light intensity, internal resistance, and power. The experimental results show that the corrosion rate of Cu coated with silver was smaller than that of uncoated Cu. Compared to the performance of the Cu-Zn system, it was also found that the Cu(Ag)-Zn system produced higher power and light intensity, which is in accordance with its smaller internal resistance. The overall experimental results indicate better performance of Cu(Ag)-Zn system and this better performance is attributed to the significantly lower corrosion rate of Cu(Ag) cathode which signifies the role of Ag layer to protect the Cu from attack by seawater. As a result, the Cu(Ag)-Zn system maintained the cathode corrosion rate with a ratio of 0.19. The percentage decrease of the OCV of the Cu(Ag)-Zn system was 6.14%, the CCV on the third day was 0.99%, the current was 36.68%, and the power was 37.83%.

Keywords

Seawater, Silver-coated Cathode, Renewable Energy

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1. INTRODUCTION

From an electrochemical point of view, seawater is a very potent natural resource as a source of electrical energy. This potential is related to the high electrolyte content of seawater (Stefano et al., 2002), which means that seawater contains a large amount of chemical energy (Chave, 1971). In principle, this chemical energy can be converted into electrical energy through an electrochemical process in a galvanic cell (Nithya Sivakami et al., 2019; La Mantia et al., 2011). However, the development of galvanic cells with seawater as an electrolyte is still very limited, primarily due to two essential characteristics of seawater that are not easy to control. The sodium as an anode creates problems with safety and cost due to the reactivity and the dry assembly process. The modification of the use of sodium as an electrode was done by the open cathode system method. This is modifying a Li-ion battery into a hybrid battery/fuel

cell system with seawater using a ceramic membrane permeable NASICON (Kim et al., 2014; Kim et al., 2022). For this requirement, it is necessary to provide electricity from the outside through the charge and discharge process, as in the case of rechargeable batteries, so that it does not explicitly act as a source of energy.

In classic electrochemistry, galvanic cell using Cu-Zn pair as electrodes is the most well-known system. As an anode, Zn can be oxidized easily and has no surface passivation during the oxidation, therefore, the system has high efficiency in producing current (Poulin et al., 2022). In addition, Zn is non-toxic and relatively cheap metal, and for these advantageous characteristics Zn has been used in Zinc-Air Batteries (ZAB) with seawater as an electrolyte (Yu et al., 2020). The problem with Cu-Zn galvanic cells with the use of seawater is the corrosive nature of seawater (Thierry et al., 2016; Nejneru et al., 2019), leading to passivation of the Cu electrode due to the production

of CuCl₂ deposit on the surface of the metal. The consequence is that the cell will lose its performance. To reduce passivation, one effective way is to coat the metal with another metal that is more resistant to corrosion (Reda et al., 2020; Ameen et al., 2010), simple and low-cost (Hakim and Pangestu, 2022; Giurlani et al., 2018).

In this research, the Cu coated with Ag was used as a cathode and paired with Zn metal as an anode in the galvanic cell using seawater as an electrolyte. Silver was used as a coating because this metal is more resistant to corrosion and the AgCl that might be formed on the surface is also conductive. To investigate the effect of coating, the performance of a galvanic cell with Cu (Ag)-Zn electrode pair was compared to that of galvanic cell with electrode pair of Cu-Zn. The experiments were run for 72 hours and divided into three cycles by replacing the seawater every 24 hours. The performance of the systems was evaluated in terms of corrosion rate, open circuit voltage, closed circuit voltage, current, light intensity, power, and internal resistance. The novelty of this research is the new method for low-cost electrodes by a silver-coated electrode implemented in seawater and the investigation of electric variables by direct load devices.

2. EXPERIMENTAL SECTION

2.1 Materials

The materials used in this research are seawater, Cu plate 7×4×0.05 cm, Zn plate 7×4×0.02 cm, 12-volt 3 watts DC LED lamp, water filter, AgNO₃ 0.02 M solution, 1% HNO₃ solution, and ethanol 96%. The tools used in this research are a multimeter, TL series digital scales, lux meter, glass baker, power supply, and water filtration unit.

2.2 Methods

2.2.1 Preparation of Cu(Ag) Electrode

The Cu(Ag) electrode was prepared by electroplating process using AgNO₃ solution in an electrochemical cell with Cu metal as the cathode and a carbon rod as the anode. Prior to electroplating, the Cu metal surface was cleaned with a 1% HNO₃ solution to remove any dirt from the surface, followed by cleaning with 96% ethanol to remove the HNO₃ adsorbed on the surface. Electroplating was run with a voltage of 2 volts for 5 minutes. The electroplated Cu was removed from the electrochemical cell, carefully rinsed with deionized water, and dried.

2.2.2 Fabrication of Electrochemical Cell

The electrochemical cell was fabricated from acrylic material with the dimension of 8×4×7 cm as shown in Figure 1a. To conduct experiment, 20 cells were assembled in a closed container and connected in series as shown in Figure 1b.

2.2.3 Data Acquisition Circuit

Data acquisition was carried out in a circuit as depicted in Figure 2. The circuit was used to determine electrical characteristics including open-circuit voltage (OCV) and close-circuit voltage (CCV), the current flowing in a 12-volt DC 3-watt Led

load. The light intensity (lux) was measured at a distance of 10 cm from LED load. at a distance of 10 cm. In this study, the experiments were performed for 72 hours with data acquisition at every hour and the seawater was replaced every 24 hours. The electrical characteristics generated were then used to calculate internal resistance (R_{in}) using Equation 1.

$$R_{in} = \frac{V_{ocv} - V_{ccv}}{I} \tag{1}$$

where R_{in} = internal resistance (kΩ), V_{ocv} = no-load voltage (open circuit voltage) in volt, V_{ccv} = voltage with load (close circuit voltage) in Volt, I = current in Ampere.

2.2.4 Corrosion Rate Measurement

In this study, the corrosion rate of the electrodes was determined using the weight-loss method and calculated using Equation 2 (ASTM, 1999).

$$r = \frac{Km}{\rho AT} \tag{2}$$

where r = corrosion rate (mm /year), K = constant (8.76 x 10⁴), ρ = density (gr/cm³), $m = m_0 - m_1$ = initial mass (gr) - final mass (gr), A = surface area of the electrode (cm²).

3. RESULT AND DISCUSSION

3.0.1 The Corrosion Rate of the Electrode

The corrosion rate of the electrodes was determined based on the weight loss after the electrodes were utilized for 72 hours, and the results are presented in Table 1.

Table 1. Corrosion Rates of the Electrodes After 72 Hour

Cu-Zn		Cu(Ag)-Zn	
Cu (mm/year)	Zn (mm/year)	Cu (mm/year)	Zn (mm/year)
0.331	0.372	0.062	0.486

The experimental results in Table 1 show that all electrodes are corroded with different corrosion rates including Cu and silver-coated Cu or Cu(Ag) used as cathodes. As can be seen in Table 1, the corrosion rate of Cu metal was much higher than that of silver-coated Cu. The calculation of the corrosion rate ratio of Cu(Ag) to Cu was 0.19, indicating a higher ability of Ag metal to resist corrosion by seawater. In this case, the Ag layer protected the Cu metal surface, but seawater could still penetrate the protective metal layer.

In the redox reaction concept of the electrochemical cell, the cathode functions as the site for reduction, and therefore theoretically no corrosion (oxidation) occurs at this electrode. However, in the case of seawater, in a previous study, it was reported that copper used as cathode was found to react with chloride ions to produce CuCl (Li et al., 2012; Kear et al.,

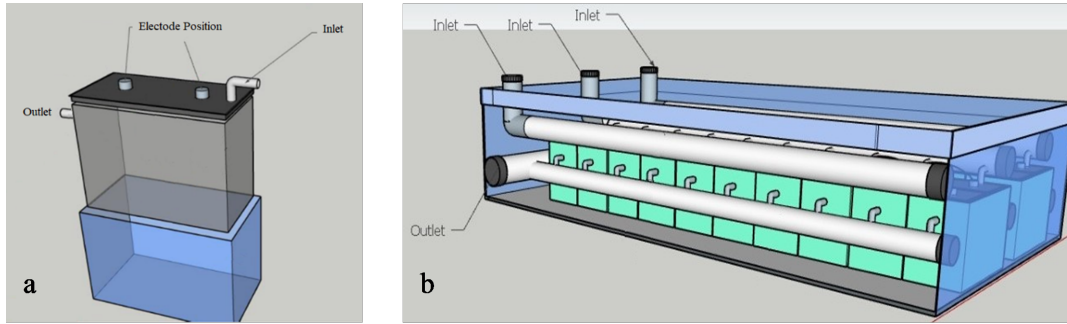


Figure 1. Electrochemical Cell Used in This Study: Single Cell (a) and Assembly of 20 Cell (b)

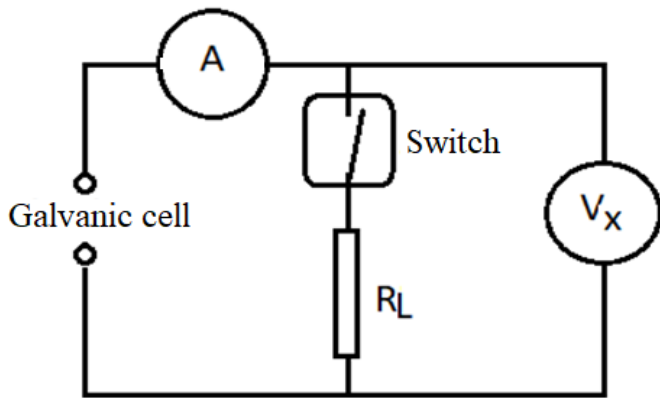


Figure 2. The Electronic Circuit Electrical Characteristics Measurement

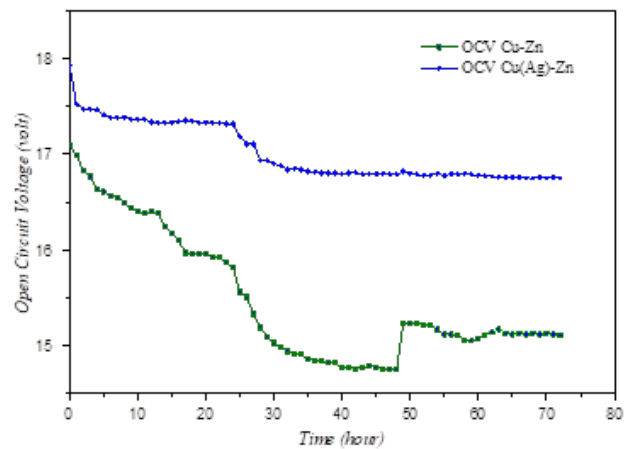


Figure 3. Open Circuit Voltage as a Function of Time for Cu-Zn and Cu(Ag)-Zn Systems

2004). Similarly, formation of AgCl was reported by Jin et al. (2003) and Sharif and Dorrnian (2015) in their study using Ag metal as a cathode in an electrochemical cell with seawater as electrolyte. Other investigators Shi et al. (2018) conducted a study to compare corrosion behavior of pure Cu and pure Cu-Ni-Zn alloy in 3.5% NaCl solution and artificial seawater with the electrochemical impedance spectroscopy and potentiodynamic polarization technique and reported that Cu experienced more severe corrosion of than that of Cu-Ni-Zn alloy. It was suggested that the reduced corrosion rate of alloyed Cu was attributed to the existence of a protective layer, while on the surface of Cu no such layer formed.

The results obtained also indicate that the use of a Cu(Ag) cathode led to an increased corrosion rate of the Zn electrode compared to the corrosion rate with the use of Cu as a cathode. This is the effect of increased power produced by the cell.

When the metal is covered with a dense dielectric layer with minimal defects, the interface between the metal surface and the electrochemical electrolyte solution generally has a high impedance. The layer acts as a dielectric in the capacitor. Because most of the protective layer has small holes and other permeable defects, this is related to the homogeneity and roughness of the electrode surface and is often used to

study local corrosion of metal layers (Chu et al., 2006). In the measurements that have been carried out, there is a relatively higher corrosion rate value on some Cu (Ag) plates compared to the average value. This shows that the electrolyte solution can still penetrate the layer at the location of the coating defect so that a corrosion reaction occurs on the metal substrate and the area on the layer.

3.0.2 Electrical Characteristic Measurement

As previously described, the first series of experiments were conducted for 72 hours and specified as a short-term experiment. Figure 3 represents the open circuit voltage (OCV) of cell using Cu-Zn and Cu(Ag)-Zn as electrode pairs. During the experiment, seawater was replaced with fresh seawater every 24 hours.

To summarize the results, the initial and final values of OCV in each of the cycles are presented in Table 2. In Table 2, for every 24-hour cycle (one cycle), in the initial measurement, Cu(Ag)-Zn is higher (17.93 volts) compared to the OCV in the Cu-Zn system (17.09 volts). In the first cycle measurement, the initial voltages can withstand an average of 96.8% for the

Table 2. Summary of the Initial and Final Value of OCV

Cycle	OCV of Cu(Ag)-Zn System (volt)		OCV of Cu-Zn System (volt)	
	Initial	Final	Initial	Final
1 st	17.93	17.32	17.09	15.82
2 nd	17.19	16.79	15.56	14.76
3 rd	16.82	16.75	15.24	15.11

Table 3. Summary of CCV and Current of System

Cycle	Cu(Ag)-Zn System				Cu-Zn System			
	CCV (volt)		I (mA)		CCV (volt)		I (mA)	
	Initial	Final	Initial	Final	Initial	Final	Initial	Final
1 st	8.18	8.01	2.32	1.22	7.99	7.88	1.50	0.52
2 nd	8.11	8.00	1.65	1.13	7.98	7.86	1.07	0.46
3 rd	8.06	7.98	1.51	1.04	7.98	7.82	1.03	0.33

Table 4. Summary of Light Intensity and Power of System

Cycle	Cu(Ag)-Zn System				Cu-Zn System			
	Light intensity (lux)		Power (mW)		Light intensity (lux)		Power (mW)	
	Initial	Final	Initial	Final	Initial	Final	Initial	Final
1 st	39	21	19	9.79	27	11	11.98	4.1
2 nd	27	20	13.41	9.01	21	9	8.52	3.63
3 rd	27	16	12.2	8.27	18	6	8.2	2.55

Cu(Ag)-Zn system, while for the Cu-Zn system it is 94.4%. With the progress of the experiment until the third cycle, the OCV in both systems decreased, but the trend observed for the Cu-Zn system was significantly steeper, with decreased percent ratio was 11.59%, than that observed for the Cu(Ag)-Zn system was 6.14%, implying that the Cu(Ag)-Zn was significantly more stable than the Cu-Zn system. This higher stability was attributed to the role of the Ag layer in resisting seawater attack.

Figure 4 indicated the closed-circuit voltage (CCV) value and current (I) observed for the two systems during the 72 hours experimental time. The experiments were conducted with the load 12-volt 3 watt of LED. To summarize the results of CCV and current in each of the cycles are presented in Table 3.

In Table 3, during the first measurement, the initial CCV was 8.18 volts for the Cu(Ag)-Zn system higher than initial CCV for Cu-Zn (7.99 volts), as well as in the second and third cycles. Each initial value for the Cu(Ag)-Zn system was consistently higher than Cu-Zn system. The percentage reduction ratio of CCV of Cu(Ag)-Zn on the third day was 0.99%, while Cu-Zn was 2.00%. The average percentage reduction ratio of the current of Cu(Ag)-Zn was 36.68%, and Cu-Zn was 63.43%. This data indicated that the galvanic cell with silver plating on the cathode could produce tremendous energy than the uncoating cathode. What was unique about the graph in Fig-

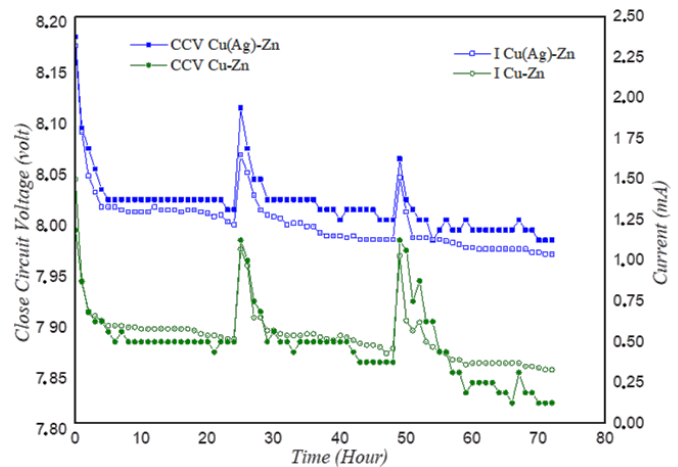


Figure 4. Closed Circuit Voltage and Current as a Function of Time for Cu-Zn and Cu(Ag)-Zn Systems

Table 5. Summary of Internal Resistance of System

Cycle	Internal Resistance of Cu(Ag)-Zn System		Internal Resistance of Cu-Zn System	
	Initial	Final	Initial	Final
1 st	4.27	7.62	5.99	15.25
2 nd	5.51	7.83	7.04	14.89
3 rd	5.73	8.52	7.12	22.4

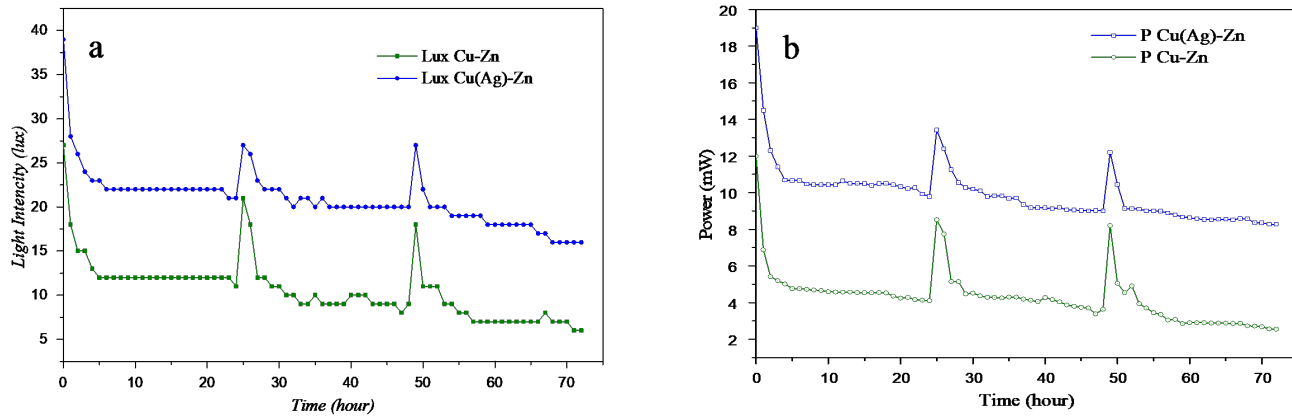


Figure 5. The Experimental Data of Light Intensity (a) and Power System (b)

ure 4, the chart pattern of these two systems was marked by a peak that rises dramatically at the change of seawater in each cycle. A sharp decrease in CCV and current during the first five hours then continued the graph at the end of the previous cycle. The drastic increase in CCV and currents were most likely due to high dissolved oxygen in the fresh seawater (Thierry et al., 2016) and then reduction during galvanic process (Guo et al., 2021). This high oxygen concentration has caused an increase in the rate of the oxidation reaction so that the CCV and current were increased.

The experimental data of light intensity and power are shown in Figure 5. The summary of both systems could be seen in Table 4. In the initial measurement, the intensity of visible light of LED 12 volt 3 watts was produced by Cu(Ag)-Zn 39 lux and Cu-Zn 27 lux, and in the third cycle, the light intensity on the Cu(Ag)-Zn system was 16 lux, and on Cu-Zn, only 6 lux. The percentage of power loss looks quite different, 37.83% for Cu(Ag)-Zn and 64.02% for Cu-Zn. This intensity graph has been the same as the CCV and current chart patterns in Figure 4. The power was dropped due to the presence of deposits from the reaction of the cathode surface with the seawater electrolyte. The presence of Cl⁻ ions in seawater will cause CuCl deposits to appear on the Cu electrodes and AgCl on the surface of the Ag layer. CuCl has a higher resistance than AgCl, so the power decreased drastically in the Cu-Zn cell.

The magnitude of the current serves to be able to calculate

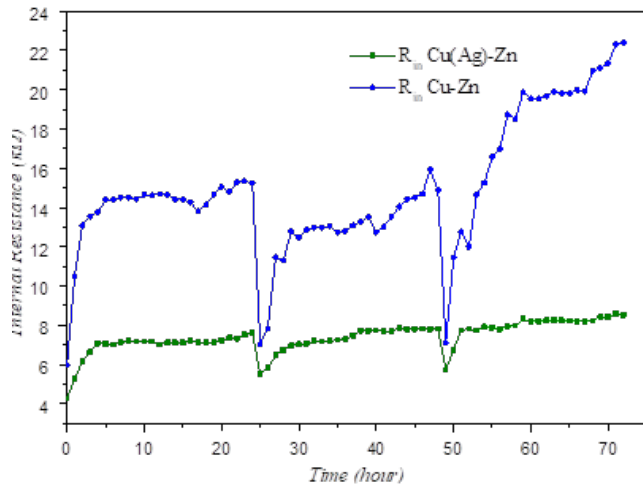


Figure 6. R_{in} in 72 Hours Measurement

the internal resistance (Figure 6) that is available in the galvanized cell. The Summary of internal Resistance can be seen in Table 5. The resistance in Cu(Ag)-Zn galvanic cells has smaller compared to Cu-Zn, with the average percent ratio increasing of resistance from cycle 1st to 3rd was 35.44%, compared with Cu-Zn 60.55%, so the power produced by Cu(Ag)-Zn is greater. When the metal is in the electrolyte, a process occurs which is a combination of the oxidation reaction, and the appearance of an electrical double layer at the interface of the metal and electrolyte surface. This process is usually illustrated with a Randles network. The Randles equivalent network is one of the simplest and most commonly used electrochemical impedance network models to analyze electricity in electrolyte systems (Kumar et al., 2019; Xu et al., 2019; Yavarinasab et al., 2021; Saxena and Srivastava, 2019; Alavi et al., 2016). The internal resistance of galvanized cells is determined by several factors such as electrolyte resistance, resistance on the electrode surface as a result of chemical reactions and crystallization and deposition processes, capacitive double layers, resistance due to polarization between electrodes, and Warburg impedance.

The Cu(Ag) electrode has less plaque and deposition on the surface than Cu, which indicates that Cu immersed in seawater will react faster on the surface. This is possible because the sea contains mineral ions such as Cl⁻, Na⁺, SO₄²⁻, Mg²⁺, Ca²⁺, K⁺, HCO³⁻, Br⁻, Sr²⁺, B³⁺, F⁻, Mo²⁺, and others.

4. CONCLUSION

Silver-coated cathodes showed better performance than uncoated copper on galvanic cells. In silver coated electrodes, the electrical characteristics of OCV, CCV, current, power are higher and internal resistance is lower than in uncoated cathodes, so the system can be used longer to generate electrical energy.

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