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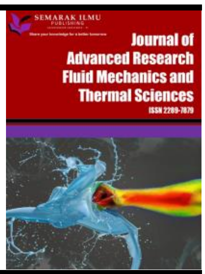

Energy from Salinity Gradient of Wetland Saline Water Using Reverse Electrodialysis Membrane

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ABSTRACT

Reverse electrodialysis (RED) membrane is an emerging renewable energy which harvest electricity from mixing two streams on different salinities. In real practice is still not clearly defined for the effectiveness of salinity gradient power (SGP)-RED due to the limitation of artificial saline water. Generally, South Kalimantan Indonesia is rich by wetland and coastal area. Due to that wetland saline water is potential as saline water sources for collection the SGP. This experiment aims to investigate the impact of natural feed stream wetland saline water was collected from Muara Halayung South Kalimantan-Indonesia to demineralized (WSW),artificial brackish (ABW) (0.35 wt %NaCl), and artificial seawater (ASW) (3.5 wt % NaCl) in terms of power density measured on a lab-scale RED membrane stack prototype 12 x 12 cm. Ion exchange membrane (IEM) was used in this work consisted from cellulose modified Anion Exchange Membrane (AEM) by EDTA-quaternization (EDC) process. Whereas, Cation Exchange Membrane (CEM) was employing Nafion NR-212 with thickness 0.002 in. Lab-scale RED tests operated 3 hours into area system has 121cm^2; AEM and CEM area have 100cm^2; uses spacer nylon a pair between membranes; and two electrodes by stainless-steal and copper. Feed water was into RED system then measured power density as an electrical energy potential that from separate ions by AEM and CEM. The result among of this experiment, characteristic of wetland saline water naturally had electro conductivity (EC) 135.6 µS/cm; TOM 15.2 ppm; and has ionic compound higher in Na^+ and Cl^- which is Cl^- compound as one of potentially formed salinity on that wetland water and become potential uses one of natural feed water in RED system. The highest energy power density on RED process obtained by mixed WSW:ASW was 1.43 W/m^2. While ratio of the gradient salinity WSW:ABW to ABW:ASW were increasing 86% which has 1:5 to 1:30 as the effect of stream mixing by two natural feed water.

1. Introduction

Energy is one of the primary needs forevery human to fulfill life in this word. Nowadays, the most use energy from CO_2 emission has affected on climate change issue. There is need growing to reduce

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or no CO_2 emissions as an alternative which uses salinity gradient power (SGP) [1]. Salinity gradient power is a new non-pollutant source of energy from difference salt concentrations by two solutions. Two concentrations were combined by fresh water and seawater as interpretation of low concentration and high concentration, respectively. The application to harvest SGP is reverse electrodialysis (RED). The RED system produces SGP by ionic membrane exchange separating ions by two feed solutions particularly [2]. RED is one of membrane application to produced energy, beside it is widely applicated as water purification. Commonly, membrane polymer, inorganic such as silica, silica-carbon are used for water and wastewater treatment [3-37]. Moreover, previous works have been mentioned silica membranes are affordable for natural wetland saline water treatment [3,4, 26,33,34,38].

One RED stack or system has each membrane namely Cation Exchange Membrane (CEM) and Anion Exchange Membrane (AEM) as selective ion exchange membranes (IEMs). The principal of CEM and AEM in RED stack for salinity gradient over become voltage difference accumulated and driving force by flux ions process. The ionic flux when pass on beyond by interlayer membranes is contact with couple re-circulating electrodes and transform into electrical current by redox process [39]. As the result from co-ionic flux, the RED harvests presents the energy Gibbs (ΔG_{mix}) by mixing two feed water formed conversely to determine energy efficiency membrane used under natural fresh water-seawater conditions [40]. Where the power density is determined by the resulted electric energy [41]. A Moreover, Veerman, et al. [42] was reported that the power density which has given quantity from river water as a feed water in RED system. The power density of RED system depends voltage and current from open-circuit voltage by mixed two streams feed water [43]. *Guo, et al. [44]* reported that the power density also depends on the chemical compounds feed water. The maximum power density from seawater and freshwater natural (Yangtze River) was 0.15 W/m. Then, the characterization by natural feed water must to investigate include the chemical compound input and output as affect to result maximum power density.

The effect compound of feed water determines quantity pressure drop, power density, and ohmic resistance RED stacks which especially the presence of Na^+ and Cl^-. The other side, the multivalent ions would result complex situation where it has more twice generated by the voltage than monovalent ions of the salinity gradient from natural seawater and river water which would decrease power density [45]. Moreover, the presence of Natural Organic Matter (NOM) and Total Organic Matter (TOC) have significant decrease voltage and inhibit power density because of counter ions or co-ions by both feed water. Where, as long as time organic foulant adhere into the surface IEM, it will close interlayer membranes automatically. The excessive by foulant will literally low effected to measure especially for calculation of electrical current and power density [46]. The observation of fouling of RED system determines the driving force of ions, especially NaCl as primarily to gradient salinity although inlet or outlet RED stacks. The determination of driving force ions as the sum of current and co-ion transport will result by calculation flux overall salinity [47].

Reverse electrodialysis stack is to generate electricity from mixing natural feeds water successfully. However, RED system would never far about fouling problems depended on feeds water. Vermaas, et al. [45] proved that every kind of membranes on RED (CEM and AEM) gradually have impacted on fouling. CEM is plastic material could least sensitive to fouling. While AEM is membrane of diatoms that could easily organic factor was observed. It is suggested to use spacer that will reduce fouling by stream mixing between two feed waters. Thus, this experiment uses wetland saline water as natural feed water and NaCl solution. Wetland saline water is natural water especially find in wetland areas in South Kalimantan, Indonesia [48]. That natural feed stream water mix within NaCl solution as it is an interpretation of artificial brackish and seawater. These would investigate into RED system as a direct conversion technique by energy power density for harvesting electrical by high and low concentration of flows.

2. Materials and Methods

2.1 Membrane Preparation

One stack RED system containing 1 AEM and 2 CEM reported by Ramon, *et al.,* [1]. The stack was built 3 flats ion exchange membranes and spacers in between. Two CEM was NR-212 perfluorosulfonic membrane (CAS. 31175-20-9, Sigma-Aldrich) had dimension 121 cm^2. Physicochemical of NR-212 perfluorosulfonic membrane described on Table 1.

Table 1
Physicochemical characteristic of NR-212

Membrane	NR-212
Formula of Chemical	(C7HF1305S.C2F4)x
Membrane Thickness	0.002 in
Water Content	8%
Acid Capacity	0.92 meq/g
Basis Weight	1100 mg/dm^2
Profile Thickness	53.8 micron
Initial Area Surface	930.25 cm^2

While the one AEM was fabricated from EDTA-quaternization process in cellulose media fiber (ROFA Laboratorium Centre Store, Bandung, Surface Area 58x58cm^2, semipermeable filter, pore size 20 nm). This process based on experiment reported by Igawa, *et al.,* [49] and US Patent 3714010. First, the cellulose fiber was immersed into 0,1M EDTA-2Na (Ethylene Diamine Tetra Acid) (BASF, Germany) for 3 hours with agitated process and after that it is dried at 25^{0}C. Second, the cellulose-EDTA was immersed into mixed solution of 5% glutaraldehyde and 0.3% formaldehyde for 17 hours as cross-linking process and after that it is dried at 25^{0}C. Third, as the quaternization process cellulose-EDTA membrane as AEM was immersed 1:1 methyl Chloride and ethanol for 24 hours, then it is dried at 25^{0}C. AEM by EDTA-quaternization process has ready to use.

2.2 RED Description

A laboratory-scale experiment energy salinity from reverse electrodialysis (RED) stack system with dimension used 121 cm^2 from two square-formed acrylics (Fabricated by PT. Jaya Alam Persada, Tangerang) with thickness 3 cm, it is according to the designed reported by Kwon, *et al.,* [50] and Dong, *et al.,* [51]. Two materials electrode-based on Volta's chain principal had consists stainless steel (Fabricated by PT. Surya Logam Universal, Jakarta) as anode material, while copper (Distributed by FIndustry, Tengerang) as cathode material which have every dimension square 100 cm^2 inside. After acrylic had took gasket from silicon rubber to preserve and reduce leaking on RED system with dimension 121cm^2 [52]. While the spacer used nylon, which has surface area of 165 x 50 cm^2, thickness 0.16 mm, pore size 51 micron with dimension 100 cm^2 (Specialist Filter Technique Filterman Product, Jakarta). One of two nylon spacer had taken between CEM and AEM shown in Figure 1. Each one stack RED had two streams flow for feed water that would pair silicon tube 0.5 inch (Mitra Sarana Tekindo Product, Jakarta).

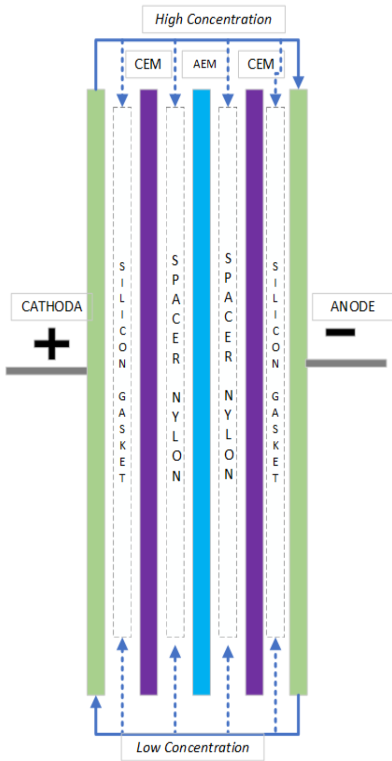

Fig. 1. Schematic of RED System

2.3 Feed Water

Two feeds water are employed in this experiment. Wetland saline water was obtained in nearby Barito River (Desa Muara Halayung, Banjar District, South Kalimantan, Indonesia). While saline water is artificial NaCl solution of 0.35 and 3.50 wt.% (EMSURE® Ph Eur MERCK :1.06404.1000). These sources of water were interpretated as river water, brackish water, and seawater. Wetland saline water naturally was stored in a buffer tank for 14 days sedimentation before processes on RED system. As the natural river water, wetland saline water was measured physical properties Total Suspended Solid (TSS) (DR 2700 Spectrophotometer Potable, HACH), Total Dissolve Solid (TDS) and Electroconductivity (EC) (Conductivity Portable Starter300C, OHAUS). While chemical properties inorganic compound used Agilent 5110 ICP OES to inorganic analyser, Metrohm-877 Titrandro Titrator to Chloride compound, Spectrophotometer SO_3, Total Organic Carbon (TOC) with TOC analyser, and Titrimetric Winkler to total organic matter (TOM) analyse.

Multiples of feed water we used two variables which low salinity (LS) and high salinity (HS). Two salinities concentration as interpretation by natural water has given

Table 2
Interpretation of Multiples Feed Water

High Salinity	Low Salinity	Interpretation
Wetland Saline Water (0.02 wt% NaCl)	Demineralize water	WSW
3.50 wt% NaCl	Wetland Saline Water	ASW
0.35 wt% NaCl	Wetland Saline Water	ABW

2.4 Operation Process

To measure power density in this experiment was employed a digital multimeter (Avometer Digital Zotek ZT102 Portable Auto-Ranging, China). The power density, on this experiment, depended voltage and resistance which was measured in the multimeter. OVC and resistance were measured every 5 minutes in overall times 3 hours previously research by Ouyang, et al. [53] continuously with flowrate 0.80 L/minute. First, the gross power density calculated on the formula by Simões, et al. [41].

$$P_{gross} = V.I \tag{1}$$

where V (Voltage) was the voltage measured by experimental data, while I was currently extracted (Ampere) by the stack RED system. Before that, in order to calculate energy efficiency (%) considered energy Gibbs inlet from mixed feed water

$$\Delta G = T.\Delta S \tag{2}$$

$$\Delta S = -Rn_{tot} \sum_i x_i \ln\left(\gamma_i x_i\right) \tag{3}$$

where S in entropy (J/K), and n_{tot} was total number (mol), and x_i was fraction of elements salinity Na^+, Cl^-, and H_2O. Then, the energy efficiency (%) was given

$$\eta_{energy} = \frac{P_{gross}}{\Delta G}.100\% \tag{4}$$

And another side, the energy of pump (J/K) ought to observe by calculation Eq. (5).

$$P_{pump} = (\phi_{SW} + \phi_{RW})dP \tag{5}$$

where dP was calculated by specification pump experiment. Because peristaltic pump used into two of one system dP was the same. Difference calculation between gross power density which measured by multimeter and power pump was determined as P_{net}.

$$P_{net} = P_{gross} - P_{pump} \tag{6}$$

From energy efficiency by net power density (%) is determined by Eq. (7).

$$\eta_{net} = \frac{P_{net}}{\Delta G}.100\% \tag{7}$$

Finally, overall power density (W/m^2) was determined by P_{tpump} with P_{gross} divided total membrane area at Eq. (8).

$$P_d = \frac{P_{values}}{2.A} \tag{8}$$

where P_{values} was overall power density while A was membrane area (m^2).

3. Results and Discussion

3.1 Characterization of Wetland Saline Water

Wetland saline water was taken at Desa Muara Halayung, Banjar District, South Kalimantan-Indonesia is characterized by color, odor, salinity, TSS, TDS, EC, TOM, TOC, and ion compounds as shown in Table 3. A typical wetland saline water naturally has species of Cl^- as largest concentration than Na^+. Then, after output on the RED system, that two components (Na^+ and Cl^-) still larger than multivalent ions (Ca^{2+}, Mg^{2+}, and SO_4^{2-}). The presence multivalent ions Ca^{2+}, Mg^{2+}, and SO_4^{2-} that may reduce voltage [45].

Table 3
Physicochemical characteristic of wetland saline water before (A) and overall after (B) input to the RED System

Physical Parameters	Wetland Saline Water	RED Effluent (WSW-ABW-ASW)
Color	Brown Yellow	Brown Yellow-Less
Odor	Odorless	Odorless
Salinity (ppt)	0.10	15.3
TSS (ppm)	1350	1008
TDS (ppm)	986	1506
EC (µS/cm)	135.60	2936
TOM (ppm)	15.20	15.20
TOC (ppm)	10.20	<10
Ion Compounds (ppm)	A	B
Al	4.458	0,959
Na	4.083	433,40
Cl	50.00	194,94
Ca	3.671	7,631
Mg	6.364	4,264
SO_4^{2-}	17.90	25,30
Total Hardness ($CaCO_3$)	35.37	18,56

This experiment would mix by demineralized water (WSW) and also NaCl artificial (0.3% and 3.5%) as interpretation of artificial brackish water (ABW) and seawater (ASW) respectively. NaCl artificial increased monovalent ion Na^+ and Cl^- that increased voltage generate [54] and reduce negative effected by TOC and NOM that potentially effected on generate power density [55]. Moreover, the spacer nylon used to eliminate the shadow effect from membrane modification and fouling effect also reduce turbulent flow [56] by two streams feed water. The overall result characterization of effluent of RED system was not only concentrations of Na^+ and Cl^- dramatically increased but also physically properties were increased. Although, multivalent ions (Ca^{2+}, Mg^{2+}, and SO_4^{2-}) were unsignificant increased than monovalent ion (Na^+ and Cl^-). This experiment has successful reduce the presence of multivalent ion and increasing monovalent ion that potentially reached optimal energy power density.

3.2 Characterization Membrane NR-212 and Anion Exchange Membrane (AEM) EDTA

Among this experiment uses two types Nafion NR-212 as CEM while EDTA's-Cellulose (EDC) as AEM with characteristic every membrane has shown in Figure 2. In RED system, two kind of membranes (CEM and AEM) is a function in order to split ions depends of membranes properties

[54]. The CEM has function to absorb amount of positive ions as negatively charged, while the AEM has function to absorb amount of negative ions as positively charged [57].

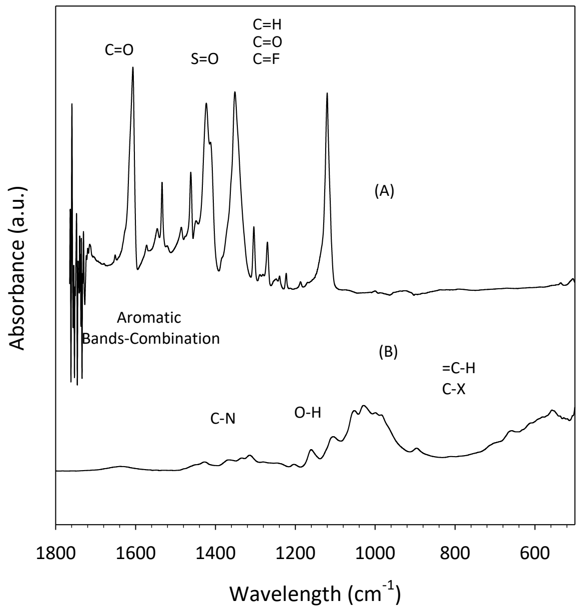

Fig. 2. (A) Chemical Compounds Nafion NR-212 (CEM) and (B) Chemical Compounds EDC (AEM)

The typical of structure chemical compound of NR-212 in Figure 2(A) has aromatic bands-combination found at around 1700 cm^{-1} and aliphatic ketone (C=O) at 1730-1705 cm^{-1} because in Nafion Du Pont NR-212 membrane has derivative aromatic of proton exchange membrane polymers like polyether ketone [58]. The wavelength at peak between 1300-1270 cm^{-1} and 1413 cm^{-1} [59] have compound C-F stretching that indicated the presence of atom halogen fluoride. Every around's of the halogen C-F presence, there are two peaks overlap sharply presence which has compound S=O found at 1425 cm^{-1} and 1353 cm^{-1}. However, the previously experiments Kunimatsu, *et al.,* [60] observed that not only presence of symmetric C-F stretching into membrane NR-212 but also S=O stretching presence into overlaps intensity which described NR-212 has a polymer chain relaxation into C-F as perfluoro groups and has intensity sulfonic acid groups. Another side around that area have peak dramatically indicated by component ether C-O-C at 1121 cm^{-1} and overlaps peak arounds 1130-1080 cm^{-1}. It is indicated the presence of SO_4^{2-} [61] because NR-212 has component complex of polyvinyl ether which polyvinyl indicated on of anhydrate component (C-O) [58].

As the compare, in Figure 2(B) has too shown a typical of EDC membrane which has more low chain chemical than NR-212. The typical of EDC membrane indicated the stretching aromatic in amine (C-N) compound at region absorbed at 1360-1250 cm^{-1} wavelength, it is because of EDTA has dinitrogen compound [62]. Moreover, the aromatic of amine in EDC membrane indicated has stretching nitro (NO_2) symmetries at 1370-1330 cm^{-1} by treated media in EDTA with aqueous solution [63] bends with stretching ester (C-O) aromatic at weak-peak 1315 cm^{-1} because it is indicated the methyl group as wagging or twisting bending [64]. However, O-H was indicated at 1300-1000 cm^{-1} wavelength overlaps with ester aromatic due to the electrooxidation process acetaldehyde from EDTA and formaldehyde compound with ethanol [65] and also aliphatic amine (C-N) has indicated at peak 1031 cm^{-1}. It is indicated interactions electrostatic into cellulose media with adsorption and Van der Walls process [66]. And furthermore, at fingerprint area has indicated the presence of halogen compound (C-X) at 700-558 cm^{-1} because of the quaternization process by methyl chloride as crosslinker and positive charge groups for base-foundation of AEM [67].

3.3 Free Energy Gibbs and Gradient Salinity

Free energy Gibbs was available to convert the electrical energy by harvesting mixed salinity [68]. The free energy Gibbs artificial brackish water and seawater resulted 238 mJ/mg and 294 mJ/mg respectively. While the free energy Gibbs WSW was 230 mJ/mg. As the result the free energy Gibbs artificial brackish water (BSW) 50% and artificial seawater (ASW) 56% more increased than WSW. The salinity resulted artificial brackish water (BSW) and artificial seawater (ASW) respectively 0.50 ppt and 15.3 ppt while WSW found 0.1 ppt which converted on the gradient salinity artificial brackish water (BSW) and artificial seawater (ASW) were 83.0% and 99.3% respectively.

It is shown in Figure 3, it is observed that the increase of salinity of kind variables mixed WSW, ABW and ASW will increase free energy Gibbs. However, the dramatically at changes ABW to ASW because continuously process and concentration of NaCl artificial that is added more than 10 times of 0.35%. The result of ratio gradient salinity this experiment (WSW: ABW and ABW: ASW) was 1:5 and 1:30 respectively. Moreover, *Golubenko, et al. [69].,* was reported which phenomena effect of concentration NaCl was impacted by conductivity. At this experiment ratio conductivities between WSW: ABW is 2:1, while ABW: ASW is 1:5. This effect increasing diffusion permeability of electrolyte to adsorbed in the membrane [69].

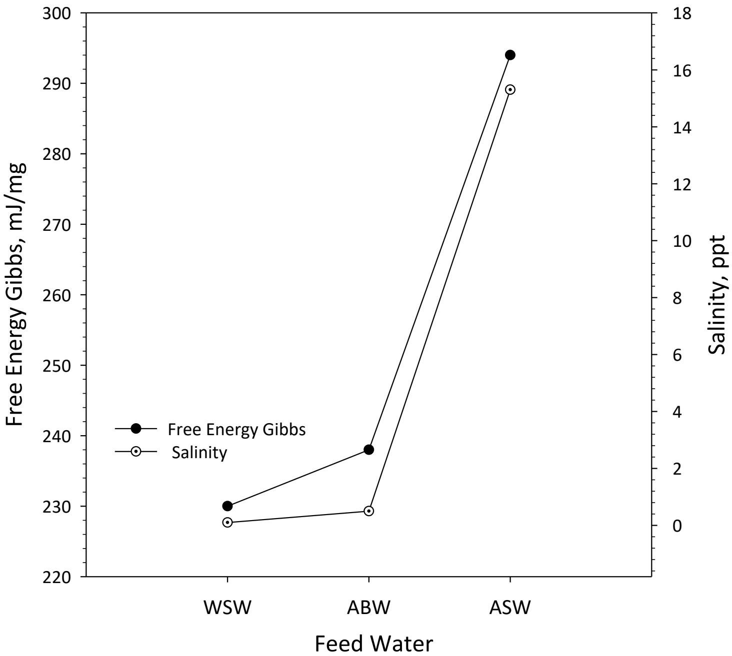

Fig. 3. Correlations Between Free Energy Gibbs with Salinity

3.4 Voltage and Power Density of RED

When the salinity increases of feed water, it shows the maximum power density on RED system by concentrations of feed water. In the other words, the concentrations especially salinity concentration, change process in RED stack to covert electrical energy. Whereas the high salinity concentration decrease voltage and area resistance while increase power density [70] and those correlations shown at Figure 4 and Figure 5 below.

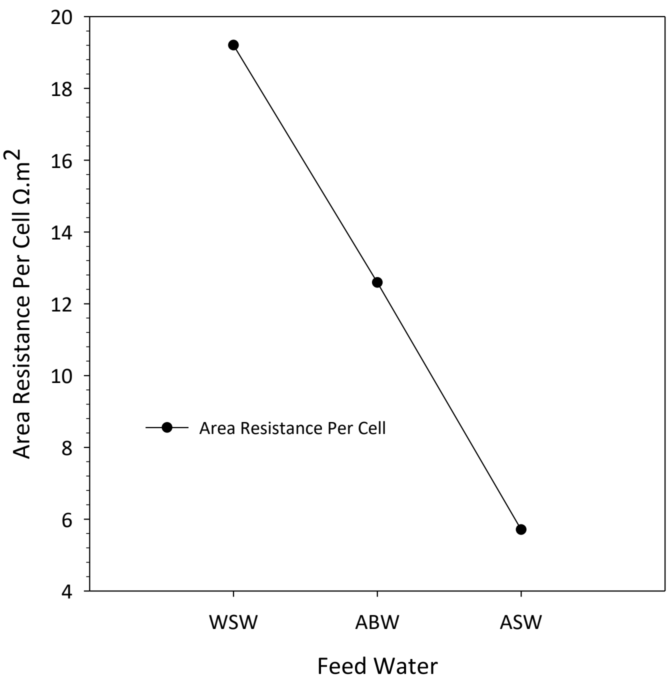

Fig. 4. Area Resistance per Cell Membrane in Feed Water

Figure 4 shown the area resistance per cell membrane. It decreases when ratio salinity of feed water increases during the experiment. The area resistance per cell decreases due to transport ion from feed water as concentrate in order to elevate the dilute solution. Dilute solution rises the area resistance per cell when membrane decreases. Then, the IEMs is dependent by concentration of ion [70]. Moreover, when the concentrate of membrane is different which ratio conductance of low solution WSW are larger than ASW. Area resistance per cell of membrane can be affected by the voltage derived from RED system shown in Figure 5.

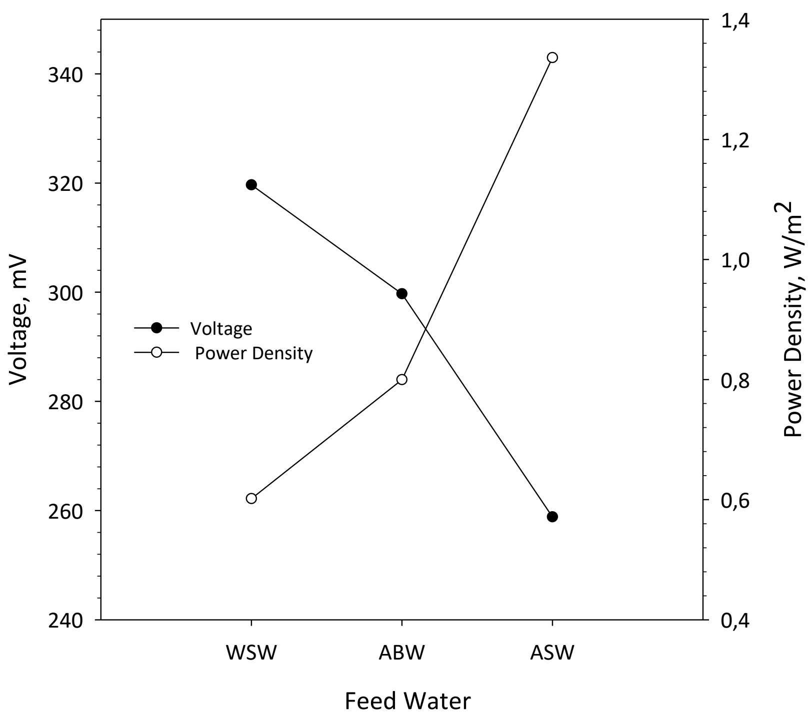

Fig. 5. Correlations Between Voltage with Power Density

By the experiment observe not only resistance is decrease but also voltage is decrease because of the transportation of ion from feed water. However, these phenomena may increase the power density. The result of maximum power density of this experiment was 1.34 W/m^2 used CEM NR-212 and AEM EDTA with feed water used wetland saline water-seawater artificial (ASW). Kim, *et al.,* [71], had observed maximum power density achieved 2.4 W/m^2 used artificial seawater and artificial river water with membranes KIER [71] with RED stack was used lab-scale, similar to this experiment. However, Choi, *et al.,* [46] observed the power density by artificial of seawater and river water used KIER membranes with 10 cell pairs membrane achieved 1.39W/m^2. As a comparation of this experiment has resulted 0.05 W/m^2 which has different of this experiment only used 3 cell pairs of membrane. Thus, this experiment has successfully effectively used less membrane previously which more than that for result of maximum power density.

4. Conclusions

Reverse electrodialysis in this experiment uses feed water WSW, ABW, and ASW to result maximum electrical energy as harvest salinity in free energy Gibbs in lab-scale. Wetland saline water with demineralized water (WSW) and NaCl artificial (ABW and ASW) uses spacer of nylon can reduce multivalent ion which affect negatively on potential energy in RED system which has TOC 10.20 ppm to reduce <10 ppm. And another side, this experiment uses two membranes CEM NR-212 and one membrane EDC has maximum power density 1.43W/m^3 more effective than previously experiment which uses ten pairs membrane. This experiment is an excellent way to investigate the energy potential uses harvesting WSW in RED system.

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