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Influence of solution concentration and salt types on the performance of reverse electrodialysis cells

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Abstract

The influence of salt concentrations on the performance of reverse electrodialysis (RED) stacks has rarely been investigated using thermolytic salts such as NH_4HCO_3 , that can be regenerated using waste heat and can be set at any desired concentration below saturation limits. Here, power densities produced by a RED stack were first investigated using different NaCl concentrations, and then tested using NH_4HCO_3 . The power produced by the RED stack increased with NaCl concentrations from 0.6 M to 3.6 M in the HC (high concentration) solution, but it did not increase at higher salt concentrations due to limited ion exchange membrane capacity. NaCl concentrations larger than 0.14 M in the LC (low concentration) solution decreased power primarily as a result of lower salinity ratios (<25). However, LC concentrations up to 0.14 M NaCl did not appreciably affect power output due to a trade-off between decreased internal resistances with higher solution conductivities and lower salinity ratios. Power densities using NH_4HCO_3 solutions were slightly lower on the basis of identical molar concentrations, but similar on the basis of matched solution conductivities.

1. Introduction

The development of novel technologies for carbon-neutral and renewable energy generation has become increasingly important to avoid further climate change due to the release of CO₂ into the atmosphere from fossil fuels [1, 2]. Salinity gradient energy, released when river water and seawater mix, could provide a large and renewable resource for clean energy production. The theoretical energy of mixing 1 m³ of river water with a large amount of seawater is about 2.5 MJ, equivalent to the energy released when water flows over a dam 250 m high [3, 4]. The possible power production from globally estuarial salinity gradients, estimated to be 1.4–2.6 TW, could potentially generate electricity for over half a billion people [5-7].

Several technologies have been proposed to capture salinity gradient energy, including pressure-retarded osmosis (PRO) [8], reverse electrodialysis (RED) [9], capacitive mixing (CapMix) [10, 11], and hydrogel expansion (HEX) [12]. Each technology uses a uniquely different approach for energy conversion, but one main advantage of RED is that it can be used for continuous and direct electrical current generation from a single reactor. The RED process is based on using a stack of alternating cation (CEMs) and anion exchange membranes (AEMs) [13-15]. When solutions with salinities similar to those of seawater and river water flow through channels separated by CEMs and AEMs, a voltage of approximately 0.1 to 0.2 V is generated across each membrane pair due to the ion flux driven by the differences in salt concentrations [16, 17]. Cations are driven from high concentration (HC) to low concentration (LC) channels through CEMs, while anions are transferred from HC to LC compartments through AEMs. The overall potential can be raised by increasing the number of membranes [18, 19]. The ionic flux in the stack is converted into electrical current through oxidation–reduction reactions on the end electrodes, such as water splitting on the anode and hydrogen evolution on the cathodes [20, 21].

Most studies on RED have used NaCl solutions at concentrations that mimic those of naturally occurring river water and seawater [9, 14, 16], and therefore there are only a few studies on the effect of variable salt concentrations on power production in RED systems in narrow ranges of 0.5-1.8 M for HC and 0-0.15 M for LC solutions [22, 23]. However, engineered salinity gradients can be created using different salts, enabling a range of salt concentrations to be used. For example, it was recently demonstrated that power could be generated with RED using thermolytic salts, such as ammonium bicarbonate (NH_4HCO_3 , AmB), that can be regenerated using waste heat [23-25]. AmB has a low temperature decomposition point, which makes it an excellent chemical for creating salinity gradients from waste heat. At low temperature (40-60 °C), LC solution can be regenerated due to AmB decomposition to CO_2 and NH_4 gases, and HC solution can be obtained by dissolving CO_2 and NH_4 gases into HC effluent [23, 24]. Approximately one-third of the energy consumed at industrial sites is lost as waste heat, and worldwide about 9400 TWh of thermal energy could be annually recaptured for useful work production [25, 26]. The use of closed-loop ammonia bicarbonate RED systems could enable capture of this waste heat directly as electricity [23, 27].

The impact of salt concentration was examined here for the purpose of using AmB as a method for converting waste heat into electricity using RED. Changes in solution concentrations and salinity ratios between the LC and HC chambers affect the energy input into the RED stack, as well as solution resistances and diffusion boundary layer resistances [28-30]. Additionally, membrane properties such as permselectivity and resistance are also affected by the solution concentration [25, 31]. Thus, it is important to know how the HC and LC solutions concentration influence the power output and energy recovery of RED processes. While both NaCl and AmB have been examined separately, there have been no direct comparisons of these two different

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4 salts in the same RED stack. In this study, NaCl solutions were initially used in tests on variable
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6 salt concentrations with a commercially available RED stack to allow comparison of the results
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8 with many previous tests using fixed NaCl concentrations. The HC solution concentrations were
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10 selected to range from 0.6 M to saturation, and the LC solution concentrations were varied from
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12 ~0 M (deionized water) to 3 M. Once the optimum conditions were identified, the performance
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14 of the RED stack was examined using AmB on the basis of either the same molar concentrations
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16 or the same solution conductivities as the NaCl solutions.
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23 **2. Materials and methods**

24 *2.1. Reverse electrodialysis stack*

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26 A commercially available, 10-cell-pair electrodialysis stack was used in all tests (PCCell
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28 GmbH, ED 64002-020, Heusweiler Germany). Both electrodes were titanium mesh coated with
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30 platinum and iridium (Ti/Pt-Ir), with a projected area of 64 cm² (8 cm × 8 cm). The membrane
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32 stack was assembled with 11 standard CEMs (PC-SK) and 10 standard AEMs (PC-SA) supplied
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34 by the manufacturer, each with an active membrane area of 64 cm² (8 cm × 8 cm), for a total
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36 active membrane area of 0.13 m². The thickness of spacers between the membranes was 0.5 mm.
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38 Three Masterflex® L/S pumps were separately used to pump HC, LC, and electrolyte solutions
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40 through the RED cell. A fixed concentration of NaCl (35 g/L, 1 L) was recycled through both the
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42 anode and the cathode chambers at 100 mL min⁻¹ to avoid large pH changes in the anolyte or
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44 catholyte. The HC and LC solutions separately flowed through the HC and LC channels of the
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46 stack in a single pass mode at previously established optimum flow rates of 10 mL/min for the
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48 HC solution, and 20 mL/min for the LC solution [32].
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Tests on salt concentration were examined by separately varying NaCl concentrations in HC and LC solutions. The HC solution concentration was increased from 0.6 M NaCl to saturation (0.6, 1.2, 1.8, 2.4, 3, 3.6, 4.2, 4.8 M, and saturated NaCl) with the LC solution fixed at 0.006 M NaCl. When the influence of LC solution concentration was examined, the LC solution concentration was changed from 0 M to 3 M NaCl (0, 0.006, 0.01, 0.02, 0.03, 0.04, 0.05, 0.06, 0.08, 0.1, 0.12, 0.14, 0.15, 0.2, 0.5, 1, 2, and 3 M) with the HC solution concentration set at 3.6 M NaCl. The impact of the type of salts was examined using NH_4HCO_3 solutions on the basis of two different conditions: by matching the molar concentrations to those of NaCl, with a 0.6 M HC solution and a 0.006 M LC solution (HC0.6M/LC0.006M), or 1.5 M HC and 0.015 M LC solutions (HC1.5M/LC0.015M); or by matching the solution conductivities to those of NaCl with different molar concentrations (HC54 mS cm^{-1} /LC0.72 mS cm^{-1} , or HC95 mS cm^{-1} /LC1.62 mS cm^{-1}).

Four digital pressure gauges (DG25, Ashcroft Inc., Stratford, CT) were installed at the inlets and outlets of the HC and LC channels to monitor pressure changes. Two pressure regulators (Hoffman open jaw screw compressor clamp, Humboldt Scientific Inc., Raleigh, NC) were added at the outlets of the HC and LC channels to adjust and match the average pressures in HC and LC chambers of the RED cells to avoid membrane damage from pressure differences.

2.2. Performance tests

The electrochemical performance of the RED stack, in terms of open circuit voltage, maximum power density, and maximum current, was obtained from polarization tests using a potentiostat (model 1470E, Solatron Analytical, Hampshire, England). Current was scanned from 0 to the maximum current (when the voltage of the membrane stack became reversed) at a rate of

0.2 mA/s. Ag/AgCl reference electrodes (BASi, West Lafayette, IN) were placed horizontally on either side of the membrane stack in the anolyte and catholyte through stoppers to record the stack voltage during each sweep. At least three polarization curves were obtained for each set condition. The open circuit voltage was determined from the vertical axis intercept of the polarization curves, and the maximum current was obtained from the horizontal axis intercept in the polarization curves. The power density of the membrane stack, which excluded electrode overpotentials, was calculated as [27]:

$$P_{\text{stack}} = \frac{U_{\text{stack}} \cdot I_{\text{stack}}}{A_{\text{mem}}} \quad (1)$$

where P_{stack} is the power density of the membrane stack (W/m^2 membrane), U_{stack} is the voltage of the membrane stack (V), I_{stack} is the scanned current (A), and A_{mem} is the total active membrane area of the stack (m^2).

2.3. Energy input and energy recovery

Energy input to the system (X^{in} , in W) changes with the HC and LC solutions concentration, which was determined from the change in the free energy due to complete mixing of the HC and LC solutions as [12, 33]:

$$X^{\text{in}} = RT \sum_i (Q_{\text{HC}} c_{i,\text{HC}}^{\text{in}} \ln \frac{a_{i,\text{HC}}^{\text{in}}}{a_{i,\text{M}}} + Q_{\text{LC}} c_{i,\text{LC}}^{\text{in}} \ln \frac{a_{i,\text{LC}}^{\text{in}}}{a_{i,\text{M}}}) \quad (2)$$

where R ($8.314 \text{ J mol}^{-1} \text{ K}^{-1}$) is the universal gas constant, T (298 K) the absolute temperature, Q (L s^{-1}) the flow rate of the solutions, c (M) the molar concentration of ionic species i in the solution, a the activity of ionic species i in the solutions obtained using OLI Analyzer Studio software (OLI Systems Inc., Cedar Knolls, NJ), and the subscripts HC, LC, and M indicates the high concentration, low concentration, and mixed solutions.

Energy recovery (E_r) was calculated as the ratio of electric power output of the stack relative to the energy input to the system [9, 34]. The electric power output was obtained based on the measured maximum power density of stack and the total membrane area.

3. Results and discussion

3.1. Influence of HC solution concentration

When NaCl concentration in the HC solution was changed from 0.6 M to saturation with a fixed LC concentration of 0.006 M, the open circuit voltage (OCV) was constant at 1.58 ± 0.03 V (Fig. 1A). The absence of an increase indicated that concentration of the HC stream was not a limiting factor in the OCV. However, both the maximum power densities (Fig. 1B) and the maximum currents (Fig. 1C) increased with HC concentration up to 3.6 M, and then became relatively stable above 3.6 M. These increases were mainly due to the increased salinity ratio driving increases in current. The power was also improved due to decreases in ohmic solution and non-ohmic diffusion boundary layer resistances in the HC channels due to the added higher salt concentrations, and decreases in these resistances in the LC channels due to the increased salt crossover from the HC into the LC channels [28]. A lack of further increases in power at HC concentrations above 3.6 M was likely limited by ionic transport capability in the ion exchange membranes. It has been demonstrated that high solution concentrations suppress the Donnan exclusion capacity of the ion exchange membranes, severely reducing the permselectivity and diminishing energy conversion efficiency in a RED stack [25, 35]. Increase of the charge groups in the membranes may improve the ion exchange capacity, but other factors (e.g, electrical resistance, swelling degree, permselectivity, and mechanical strength) should also be taken into account [13, 36, 37].

Fig. 1. (A) Open circuit voltage, (B) maximum power density, and (C) maximum current of the RED stack when HC solution concentration increased from 0.6 M to saturated NaCl and LC solution concentration was 0.006 M NaCl.

The change of HC solution concentration had no impact on the hydrodynamic power loss (pumping energy) as demonstrated by similar pressure drops in the HC and LC channels (Fig. 2A). The energy input increased almost linearly with HC concentration due to the higher salinity ratios (Fig. 2B). However, energy recovery decreased inversely with the HC concentration due to inefficient capture of mixing energy, and limitations in ion transport through the membrane (Fig. 2B). In order to obtain a maximum energy output and relatively higher energy recovery, the

optimum HC solution concentration was considered here to be ~3.6 M. Under this condition, the open circuit voltage was 1.56 ± 0.01 V, the maximum power density was 0.78 ± 0.03 W/m²-membrane (total area), the maximum current was 274 ± 8 mA, and the overall energy recovery was 3.0%. The energy recovery was low due to the use of a single pass of the solutions through the membranes, but it could be enhanced by passing the effluents through additional units or recycling through the same unit [33, 38].

Fig. 2. (A) Pressure drops of HC and LC channels and (B) energy input and energy recovery of the RED stack when HC solution concentration increased from 0.6 M to saturated NaCl and LC solution concentration was 0.006 M NaCl.

3.2. Influence of LC solution concentration

An increase of NaCl concentration in the LC solution from 0 to 3 M NaCl, with a fixed HC solution concentration of 3.6 M, consistently decreased the OCV (Fig. 3A and 3B) due to the lower salinity ratio (energy input). However, the maximum power densities and maximum currents were nearly constant in the low LC solution concentration range of 0 to 0.14 M (Fig. 3D and 3F), and they only rapidly decreased at LC solution concentrations above 0.14 M (Fig. 3C and 3E). The stable power output below 0.14 M was likely a result of the trade-off between the reduction of ohmic solution resistance achieved by using an LC solution with a higher concentration of salt (greater ion conductivity), and the decrease in the salinity ratio which would reduce the driving force for power generation. At LC solution concentrations lower than 0.14 M, changes in these two properties were relatively balanced, resulting in little change in power production with low LC solution concentrations. However, at LC solution concentrations larger than 0.14 M, the influence of solution resistance on power generation was less important, and the reduction of the salinity ratio (<25) was the main factor affecting power production. Thus, the power output greatly declined with LC solution concentrations > 0.14 M.

Fig. 3. Open circuit voltage for a whole range (A) and a narrow range (B), maximum power density for a whole range (C) and a narrow range (D), and maximum current for a whole range (E) and a narrow range (F) of the RED stack when LC solution concentration increased from 0 M to 3 M NaCl and HC solution concentration was 3.6 M NaCl.

The pressure drops through HC and LC channels were also quite similar over the range of LC solution concentrations (Fig. 4A and 4B), which indicated that the hydrodynamic power losses (pumping energy) were unaffected by the LC concentration. The energy input almost linearly

decreased inversely with the LC solution concentration due to decreasing salinity ratios (Fig. 4C and 4D). Because the power output was constant with LC solution concentrations lower than 0.14 M, and it greatly decreased for LC solution concentrations larger than 0.14 M, the energy recovery was relatively constant at LC solution concentrations below 0.14 M, with large decreases above 0.14 M. Under closed-loop conditions using thermolytic salts (e.g., AmB), the LC and HC solutions are regenerated using waste heat. AmB in the LC effluent is decomposed to CO_2 and NH_3 gases at low temperature (40-60 °C), which are dissolved into the HC effluent. Thus, a higher LC solution concentration would be preferred because less regeneration energy was needed to distill CO_2 and NH_3 out. Taking into account the impact of the LC concentration on power generation and energy recovery, these results suggest that the optimum LC solution concentration was ~ 0.14 M. Under this condition, the open circuit voltage was 1.08 ± 0.01 V, the maximum power density was 0.62 ± 0.02 W/m^2 -membrane, the maximum current was 302 ± 2 mA, and the energy recovery of 2.6 % was again limited due to a single-pass system.

Fig. 4. Pressure drops of HC and LC channels for a whole range (A) and a narrow range (B), and energy input and energy recovery for a whole range (C) and a narrow range (D) of the RED stack when LC solution concentration increased from 0 M to 3 M NaCl and HC solution concentration was 3.6 M NaCl.

The maximum power density of 0.62 W/m²-membrane here with HC of 3.6 M and LC of 0.14 M was within the range of previous results of 0.4 to 1.4 W/m²-membrane, although within the lower range of those power densities [14, 18, 19, 36]. Differences in the studied conditions could all be a factor, including the relatively low flow rates (HC of 10 mL/min, linear velocity of 2.5 cm/min, LC of 20 mL/min, linear velocity of 5 cm/min) used here to reduce pumping energy as previously reported [32], the intermembrane distance of 0.5 mm which could be reduced using different spacers [39], and the permselectivity of the ion exchange membranes compared to those used by others [13, 40]. Although the optimum HC and LC solution concentrations for other RED cells at different operating conditions might not be the same as those here, the general trends of RED performances with the different HC and LC solution concentrations would be similar to those observed here. The most important factor is the membrane. In order to make RED application practical, the electrochemical properties of ion exchange membranes used in these systems need to be further improved, and the price of the membranes needs to be reduced.

3.3. Comparison between NaCl and NH₄HCO₃ solutions

Based on the above results using NaCl, the performance of the RED stack was examined using NH₄HCO₃ at two different ratios of salt concentrations. The first comparison was made at NH₄HCO₃ molar concentrations that matched the NaCl concentrations used to represent typical river water and seawater (HC0.6M/LC0.006M). The second comparison was made at a very high molar concentration based on upper limits for the solubility of NH₄HCO₃ (HC1.5M/LC0.015M).

When the NH_4HCO_3 and NaCl solutions are used at identical molar concentrations, the ionic conductivities of the solutions are different due to the different ion activities (Table 1). Therefore, the concentrations of the NH_4HCO_3 solutions were increased using additional salt to match the conductivities of the NaCl solutions with molar concentrations of 0.6 M for HC and 0.006 M for LC, which was indicated as HC54 mS cm^{-1} /LC0.72 mS cm^{-1} . The concentrations of the NaCl solutions were decreased to match the conductivities of the NH_4HCO_3 solutions at molar concentrations of 1.5 M for HC and 0.015 M for LC, which was indicated as HC95 mS cm^{-1} /LC1.62 mS cm^{-1} (Table 1).

Table 1. Concentration (C , in M) and conductivity (S , in mS cm^{-1}) of NaCl and NH_4HCO_3

NaCl				NH_4HCO_3 (AmB)			
HC		LC		HC		LC	
C/M	$S/\text{mS cm}^{-1}$	C/M	$S/\text{mS cm}^{-1}$	C/M	$S/\text{mS cm}^{-1}$	C/M	$S/\text{mS cm}^{-1}$
0.6	54	0.006	0.72	0.6	45	0.006	0.80
1.5	119	0.015	1.85	1.5	95	0.015	1.62
1.13	95	0.013	1.62	0.74	54	0.005	0.72

When the RED stack performance was examined on the basis of matching the molar concentrations of the NH_4HCO_3 to NaCl solutions, power production was reduced (Fig. 5A). Part of this reduction in power was due to the lower permselectivities of NH_4HCO_3 in these membranes compared to NaCl [40]. However, the main reason was the different solution conductivities. When the conductivities of the NH_4HCO_3 were adjusted to match those of the NaCl solutions, the maximum power densities of the RED stack was similar for both NaCl and NH_4HCO_3 solutions (Fig. 5B).

Fig. 5. Polarization curves of the RED stack with NaCl and NH_4HCO_3 solutions (A) at the same molar concentrations (HC0.6M/LC0.006M or HC1.5M/LC0.015M), or (B) at the same conductivities (HC54 mS cm^{-1} /LC0.72 mS cm^{-1} or HC95 mS cm^{-1} /LC1.62 mS cm^{-1}).

The energy input was higher using NaCl solutions than NH_4HCO_3 on the basis of the same molar concentrations due to the higher activities of Na^+ and Cl^- ions, especially for the high concentration pair (HC1.5M/LC0.015M) (Fig. 6), in agreement with the better performance of the RED stack with NaCl solutions. The energy recovery was also larger for NaCl when the two solutions were compared at the same molar concentrations at the lower salt concentrations (HC0.6M/LC0.006M), but it was smaller at the higher salt concentrations (HC1.5M/LC0.015M) (Fig. 6). The larger reduction in energy recovery at the higher NaCl concentrations reflected greater losses of mixing energy, which could have resulted from easier Na^+ and Cl^- ions

transport (permeability) through the membranes under these conditions. The energy inputs were similar when solutions conductivities were matched (Fig. 6), but the energy recovery was larger for NaCl solution for the low conductivity conditions pair (HC54 mS cm⁻¹/LC0.72 mS cm⁻¹), and similar for high conductivity pair (HC95 mS cm⁻¹/LC1.62 mS cm⁻¹) (Fig. 6). These results indicate that previous results for RED tests using NaCl solutions can be used for NH₄HCO₃ solutions, but only when the solution conductivities are matched.

Fig. 6. Energy input and energy recovery of the RED stack with NaCl and NH₄HCO₃ solutions at the same molar concentrations (HC0.6M/LC0.006M or HC1.5M/LC0.015M), or at the same conductivities (HC54 mS cm⁻¹/LC0.72 mS cm⁻¹ or HC95 mS cm⁻¹/LC1.62 mS cm⁻¹).

4. Conclusions

The performance of the RED stack significantly affected by the HC and LC solution concentrations. The power output of the RED stack increased with the HC solution concentration in the range of 0.6 M to 3.6 M NaCl, but it was constant above 3.6 M as further increase in power was limited by the ion exchange capacity of membranes. The power output of the RED stack was relatively constant for LC solution concentrations of 0 to 0.14 M NaCl due to the trade-off between decreased salinity ratios and solution resistances, while it greatly decreased

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4 above 0.14 M due to the much lower salinity ratio. The performance of the RED stack with
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6 NH_4HCO_3 salt was lower than that with NaCl salt at the same molar concentrations due to lower
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8 ion activities, but they were similar when the solution conductivities were matched. These results
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10 therefore indicated that previous conclusions regarding RED tests using NaCl solutions are
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12 suitable for NH_4HCO_3 solutions when tests are conducted at the same solution conductivities.
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Figure-1

Figure-2

Figure-3

Figure-4

Figure-5
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