



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



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ABSTRACT

The influence of salt concentrations on the performance of reverse electro dialysis (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.

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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 CO_2 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 m^3 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 electro dialysis (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 0.1–0.2 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 temperatures decomposition point, which makes it an excellent chemical

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for creating salinity gradients from waste heat. At low temperatures (40–60 °C), the LC solution can be regenerated due to AmB decomposition to CO₂ and NH₃ gases, and the HC solution can be obtained by dissolving CO₂ and NH₃ gases into the 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 concentrations 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 solution concentrations 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 salts in the same RED stack. In this study, NaCl solutions were initially used in tests on variable salt concentrations with a commercially available RED stack to allow comparison of the results with many previous tests using fixed NaCl concentrations. The HC solution concentrations were selected to range from 0.6 M to saturation, and the LC solution concentrations were varied from ~0 M (deionized water) to 3 M. Once the optimum conditions were identified, the performance of the RED stack was examined using AmB on the basis of either the same molar concentrations or the same solution conductivities as the NaCl solutions.

2. Materials and methods

2.1. Reverse electro dialysis stack

A commercially available, 10-cell-pair electro dialysis stack was used in all tests (PCCell GmbH, ED 64002-020, Heusweiler Germany). Both electrodes were titanium mesh coated with platinum and iridium (Ti/Pt-Ir), with a projected area of 64 cm² (8 cm × 8 cm). The membrane stack was assembled with 11 standard CEMs (PC-SK) and 10 standard AEMs (PC-SA) supplied by the manufacturer, each with an active membrane area of 64 cm² (8 cm × 8 cm), for a total active membrane area of 0.13 m². The thickness of spacers between the membranes was 0.5 mm. Three Masterflex[®] L/S pumps were separately used to pump HC, LC, and electrolyte solutions through the RED cell. A fixed concentration of NaCl (35 g/L, 1 L) was recycled through both the anode and the cathode chambers at 100 mL min⁻¹ to avoid large pH changes in the anolyte or catholyte. The HC and LC solutions separately flowed through the HC and LC channels of the stack in a single pass mode at previously established optimum flow rates of 10 mL/min for the HC solution, and 20 mL/min for the LC solution [32].

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 NH₄HCO₃ 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 (HC0.6 M/LC0.006 M), or 1.5 M HC and 0.015 M LC solutions (HC1.5 M/LC0.015 M); 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 follows [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/m² 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 (m²).

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 J mol⁻¹ K⁻¹) is the universal gas constant, T (298 K) is the absolute temperature, Q (L s⁻¹) is the flow rate of the solutions, c (M) is the molar concentration of ionic species i in the solution, a is 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 indicate 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.

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