

Conversion of saline water and dissolved carbon dioxide into value-added chemicals by electro dialysis



Saad Dara^a, Michael Lindstrom^b, Joseph English^a, Arman Bonakdarpour^a, Brian Wetton^b, David P. Wilkinson^{a,*}

^a Department of Chemical and Biological Engineering and Clean Energy Research Center, 2360 East Mall, University of British Columbia, Vancouver, BC V6T 1Z3, Canada

^b Department of Mathematics, 1986 Mathematics Road, University of British Columbia, Vancouver, BC V6T 1Z2, Canada

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ABSTRACT

We demonstrate a new approach that simultaneously converts dissolved carbon dioxide and high salinity waste-water to desalinated water and value-added chemicals in the form of inorganic acids and carbonate salts. A four compartment electro dialysis cell module using anion exchange and cation exchange membranes, and a Pt/Ir-coated Ti anode and Ti mesh cathode was used to produce HCl and NaHCO₃ products from CO₂ and NaCl feed. Cell operation consisted of feeding the cell with carbonic acid (CO₂ dissolved in water) and a saline solution of sodium chloride. Under an applied voltage clear production of inorganic carbon salts and acids was demonstrated. The product fluxes for HCl and NaHCO₃ were $\sim 0.05 \text{ mM cm}^{-2} \text{ h}^{-1}$ on average. A mathematical model for this electro dialysis cell configuration was developed to better understand limitations within the cell which are not available from experimental data including concentration profiles within the intra-membrane channels.

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1. Introduction

Management of carbon dioxide emissions and wastewater generated through industrial processes is a significant challenge faced by the industrial sector. Increasingly limited water supplies, tightening regulations around carbon emissions and disposal of wastewater have driven industry to look at innovations that address these environmental concerns. Several technological options exist for both carbon dioxide sequestration and mitigation [1–18], and waste-water treatment [19–23], but these approaches remain unattractive for different reasons. While these technologies have attempted to address the issues of both carbon dioxide emissions and water treatment separately, a coupled process that simultaneously addresses both carbon dioxide removal and water treatment has only had limited success. In this paper, electro dialysis is used to convert carbon dioxide provided in the form of dissolved carbon dioxide and saline waste-water into value-added chemicals and desalinated water.

Electrodialysis cells have been shown to be cost effective for desalination purposes [24] for many applications including

industrial saline water treatment [25], brackish water treatment [26], and acid and alkali production [27] with high removal rates for different ions [28]. A number of studies have shown that the energy costs of electro dialysis are lower than other conventional methods like ion resin exchange [28–30].

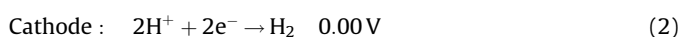
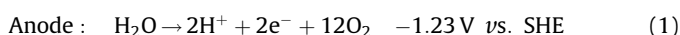
Bipolar membrane electro dialysis has also been shown to produce an acid and base where the base is used to sequester the carbon dioxide. The success of this technique is, however, based on the use of expensive and complex bipolar membranes capable of splitting water within the membrane to produce the conjugate acid and base. Furthermore, the base/caustic solution produced is limited to a low concentration due to the instability of bipolar membranes at high pH (pH > 12) and limited current densities [31,32] which limit the amount of CO₂ that can be removed. Electro dialysis cells can also recover caustic scrubbing solvents used to capture carbon dioxide from a flue gas stream [33]. Here, the caustic solution is regenerated with the use of bipolar membranes and carbon dioxide separated into a purified stream. In another application, bipolar membrane electro dialysis was used to remove carbon dioxide from sea-water into a purified stream, but not for the conversion of the carbon dioxide to a mineralized species [34].

A multi-compartment electro dialysis system with a strategic pattern of anion exchange membranes (AEM) and cation exchange membranes (CEM) to simultaneously remove carbon dioxide in a

* Corresponding author.

E-mail address: dwillkinson@chbe.ubc.ca (D.P. Wilkinson).

mineralized form and desalinate waste-water is demonstrated. The demonstration is further supported with the development of a mathematical model of this concept. A feed water stream (e.g., brine, seawater or waste-water) containing ionic species, which are to be removed, are provided to one feed compartment and carbon dioxide dissolved in water or other high carbon dioxide solubility solvents is provided in the form of carbonates (e.g., CO₃²⁻, HCO₃⁻) to a second feed compartment. The ion exchange membranes are configured in such a manner that the transport of cations and anions from the feed chambers produces carbonate (e.g., NaHCO₃) and acids (e.g., HCl or other acid) or other products in separate chambers. The basic operation of this modified electro dialysis approach is shown in a schematic in Fig. 1. The electrochemical reactions at the electrodes are the electrolytic water decomposition reactions (oxygen evolution reaction) and the reduction reaction to hydrogen, which can be summarized as follows:



The membranes (AEM and CEM) are separated with the use of a spacer which allows the solutions to flow between them and create turbulence in the stream to improve mass transfer.

The approach proposed in this article could have significant benefits as it would remove carbon dioxide and desalinate waste-water to produce value-added chemicals in addition to allowing significantly more reclamation and re-use of water for relevant industries.

2. Experimental

Experimental demonstration of the treatment and conversion of dissolved carbon dioxide and saline water into chemicals was performed in a commercially available, bench-top, four chamber electro dialysis cell acquired from PC-Cell GmbH. The cell has an active membrane area of 64 cm² and an intra-membrane gap of 0.75 mm. The anode and cathode electrodes were comprised of Pt-coated and Ir-coated titanium mesh, respectively. All

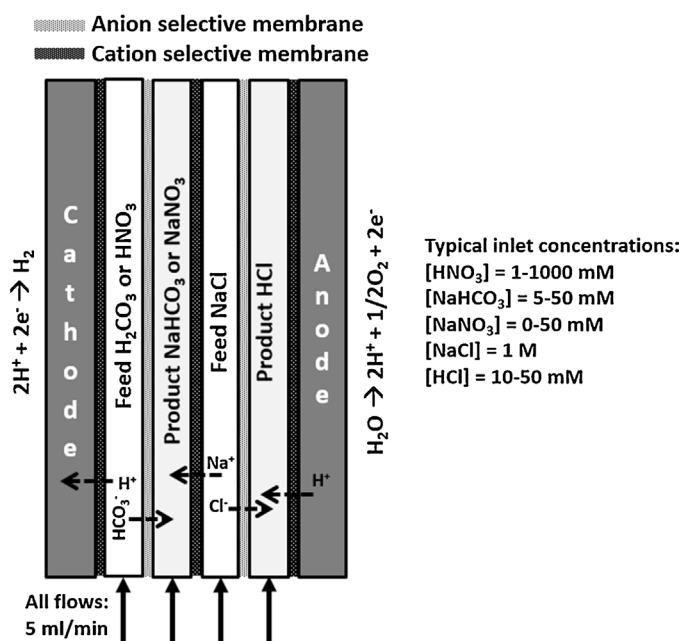


Fig. 1. Schematic showing the basic operation within the electro dialysis cell to simultaneously remove dissolved CO₂ and desalinate saline solutions.

Table 1

Summary of membranes used for the relevant ions and the solutions for pre-treatment.

Ion	Membrane type	Membrane name	Pre-treatment solution
Proton	Acidic sulfonic CEM	PC SK	Deionized water
Sodium	Acidic sulfonic CEM	PC MVK	5 wt.% NaCl
Bicarbonate	Alkaline ammonium AEM	PC MVA	0.5 M NaHCO ₃
Chloride	Alkaline ammonium AEM	PC Acid 60	5 wt.% NaCl
Nitrate	Alkaline ammonium AEM	PC Acid 60	0.5 M NaNO ₃

membranes in the electro dialysis cell used were supplied by PC-Cell. The membranes were pre-treated by storing in a salt solution of the relevant ion for at least 24 h. The membranes used to conduct the different ions are summarized in Table 1. There are numerous membranes available for this purpose. The choice of membranes used (see Table 1) was based on the manufacturer's recommendations for the different ions and solutions. Polarization curves and modeling work used PC-MVK, PC-SK and PC-MVA membranes. For longer-term tests at constant voltage the PC-MVA membrane was replaced with PC-Acid 60 membranes due to their lower proton leakage.

Pure carbon dioxide was bubbled in deionized water for 30 min to produce carbonic acid prior to being fed to the system. Carbon dioxide was continuously bubbled during operation to maintain sufficient ionic species in the carbonic acid stream ("Feed H₂CO₃ or HNO₃" in Fig. 1). The flow rate of all reactants was controlled with the use of peristaltic pumps (Masterflex L/S 600 RPM, flow rates in ml/min). In all cases the saline stream to be treated consisted of 1 M NaCl. The product NaHCO₃ and HCl streams were provided with an initial concentration of 5 mM NaHCO₃ and 10 mM HCl, respectively. The anode and cathode compartments were fed a 1 M sulfuric acid solution to facilitate oxygen and hydrogen generation. All streams were supplied to the cell at a flow rate of 5 ml/min. The addition of sulfuric acid improves conductivity and minimizes voltage losses at the electrodes thus allowing a better understanding of the ion transport within the cell. The cell was operated for periods of time up to 12 h at different applied cell potentials in the range of 0–7 V. The chrono-amperometric measurements were performed using a Biologic SP-150 potentiostat with current and voltage collection frequency of 3 Hz.

The streams fed to the ED system were sampled, analyzed for the relevant ions and pH measurements of the solutions were obtained at regular intervals. Chloride and nitrate ion analysis was performed with the use of a Dionex Ion Chromatograph ICS-900 with an AS22-FAST column. Bicarbonate ion analysis was performed by titration of the solution with 10 mM hydrochloric acid using a pH meter to determine neutralization points. Details of determination of hydrochloric acid by titration can be found in Vogel's "Textbook of Quantitative Chemical Analysis" [35]. Sodium ion analysis was performed by inductively coupled plasma–optical emissions spectroscopy (ICP–OES).

3. Calculations

A mathematical model of some of the experimental aspects was developed to further understand the performance limitations of this approach and to assess future designs. The model was developed by providing a nitric acid feed instead of the carbonic acid feed. Modeling of this feed is simpler and a clearer match to the experimental results in this case can be obtained due to the absence of a dynamic equilibrium that exists with carbonic acid.

3.1. Fit to electrolysis polarization and external resistance

Detailed modeling of the electrolysis in the electrodes of the electro dialysis cell is difficult and not central to the current project.

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