



Selective separation of mono- and di-valent cations in electro dialysis during brackish water desalination: Bench and pilot-scale studies

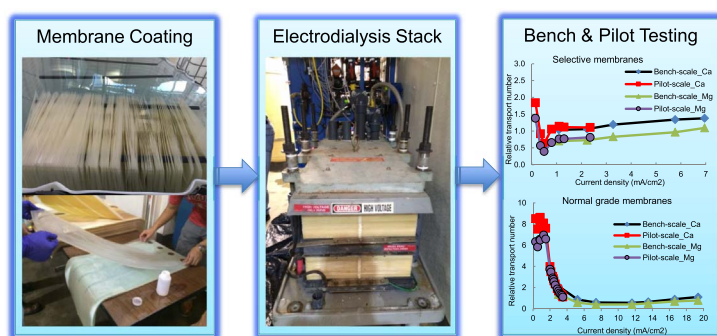


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GRAPHICAL ABSTRACT



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ABSTRACT

Selective separation of mono- and multi-valent ions has important applications in water reuse, desalination, and salt production. Innovative monovalent permselective cation-exchange membrane CR671 was developed by modifying the normal grade membrane CR67 with polyethyleneimine coating. Bench- and pilot-scale electro dialysis experiments were conducted at a brackish groundwater desalination facility to investigate desalination performance and ion selectivity using the CR671 and CR67 under different operating conditions. Both normal grade (CR67 and AR204) and selective (CR671 and AR112B) membranes achieved same desalting efficiency. Na-selectivity in terms of relative transport number using sodium as the standard ion was affected significantly by current density and linear velocity for the normal grade membranes while the selective membranes exhibited stable good Na-selectivity. The Na-selectivity of the CR671 was demonstrated up to 9 and 5 times better than the CR67 during pilot- and bench-scale electro dialysis, respectively. Hydraulic retention time and electro dialysis stack staging had minor impact on Na-selectivity at high current density. With the same hydraulic conditions, overall desalination behavior and ion selectivity were highly comparable between bench- and pilot-scale electro dialysis. It infers that bench-scale testing results can be used to simulate and project desalination performance and ion selectivity of pilot- and potentially full-scale electro dialysis applications.

1. Introduction

With the continuous depletion of fresh water resources, increasing

water demand, and frequently occurring droughts, desalination of brackish groundwater is critical to expand traditional water supplies and to alleviate water shortage. Membrane processes such as reverse

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osmosis (RO) driven by high hydraulic pressure are principal methods for water desalination [1–3]. Electrodialysis is an alternative desalination technology that uses ion-exchange membranes (IEMs) to separate salt ions in water under the influence of electrical potential. Electrodialysis has been utilized for over 60 years for desalting brackish water, treating municipal and industrial wastewater, producing salt from seawater, and being applied in chemical processes, food and drug industries [4–7].

Electrodialysis has high tolerance of silica, hardness, chlorine residual, and organic matter; therefore, it could achieve higher water recovery compared to RO during treatment of impaired waters [5,8–11]. By adjusting applied voltage and using different stages of IEMs, electrodialysis can control salt concentration in product water and modify salt composition to meet specific water quality requirements of end uses [12,13]. Unlike RO that produces high purity water and requires blending and post-treatment, using electrodialysis for desalination can avoid the complexity of product water blending and stabilization.

Another unique advantage of electrodialysis is the selective separation of monovalent ions (such as Na^+ , K^+ , NH_4^+ , Cl^- , and NO_3^-) versus multivalent ions (e.g., Ca^{2+} , Mg^{2+} , SO_4^{2-} , PO_4^{3-}), which can be achieved by using monovalent permselective IEMs [5,7,14–22]. Selective removal of Na^+ over Ca^{2+} and Mg^{2+} is especially important if the treated water is used for irrigation. For example, in southwestern United States (e.g., Arizona, California, New Mexico, and Texas), high sodium adsorption ratio (SAR), defined as the equivalent concentration ratio between sodium and calcium and magnesium, has inhibited the use of reclaimed water and groundwater for irrigation [23–25]. Irrigation water with high SAR associated with relatively high Na^+ concentration can decrease water infiltration and permeability of soil, thereby reducing crop yield [25].

Selective separation of ions in solution through IEMs is governed by the affinity of ions with membranes and their specific migration rate in membranes. The permselectivity of an IEM for a specific ion can be determined by three mechanisms: i) control of the permselectivity of ions with the same charge on the basis of their hydrated ion size; ii) rejection of certain ions by a thin surface layer on the IEM with the same charge as the ions; and iii) specific interactions between the ion-exchange functional groups of the membrane and the mobile ions [17,26]. Monovalent cation and anion permselective IEMs have been industrially used to increase the efficiency of producing NaCl from seawater [7]. However, the selective membranes currently in the market are costly and challenged during desalination of unconventional waters with high fouling and scaling potential. There is a pressing need to develop low-cost robust monovalent permselective IEMs.

In this study, we collaborated with General Electric Water & Process Technologies (GE) to develop a cost-effective monovalent permselective cation-exchange membrane (CEM) CR671 by modifying the normal grade CEM CR67 with polyethyleneimine. The CR67 is a homogeneous CEM comprised of cross-linked copolymer of vinyl compounds, containing sulfonic acid functional groups, which allows the cations to pass through. Coating a thin layer of polyethyleneimine conductive cationic polymer on the CR671 surface causes stronger electric repulsion of the cationic layer to multivalent cations (e.g., Ca^{2+} , Mg^{2+}) than to the monovalent cations (e.g., Na^+ , K^+); and the increased density of the surface structure in the polyethyleneimine layer leads to sieving of multivalent cations with relatively larger hydrated ion radius, resulting in higher permselectivity of CR671 for monovalent cations in electrodialysis of salt solutions. As a positively charged polyelectrolyte, the structure of polyethyleneimine is highly branched [27], so most of its amino groups remain on the CR671 membrane surface without ion-exchange with sulfonic acid groups of the membrane. Thin film formed by adsorption or electrodeposition of polyethyleneimine onto CEMs was reported to fade away during a long-term continuous operation [7,28]. Hence, it is desirable to form a stable chemical bond of the cationic polyethyleneimine layer with the membrane. The CR671 has active

epoxy groups on the membrane surface that react with the amino groups of polyethyleneimine to form a thin layer with stable acid-amide bonding to improve coating durability.

In view of the important applications of monovalent permselective IEMs in water reuse, desalination, and salt production, this study aimed to demonstrate the desalination performance and ion selectivity of the newly developed CR671 for electrodialysis of brackish groundwater. Continuous-flow experiments using bench- and pilot-scale electrodialysis systems were conducted at a desalination plant to treat brackish groundwater. The specific objectives of the study are threefold: i) compare the electrodialysis performance and ion selectivity of the CR67 and the CR671; ii) evaluate the impact of operating conditions on desalination and selective separation of cations in electrodialysis, including applied current density, linear velocity, hydraulic retention time (HRT), and stack staging; and iii) compare the electrodialysis performance at bench- and pilot-scale for system scale-up and optimization.

2. Materials and methods

2.1. Groundwater quality and analysis

The bench- and pilot-scale electrodialysis experiments were conducted from December 2015 to July 2016 at the Kay Bailey Hutchison Desalination Plant in El Paso, Texas, treating brackish groundwater. During the testing period, the total dissolved solids (TDS) concentration of the groundwater was 2736 ± 50 mg/L with electrical conductivity of 4626 ± 263 $\mu\text{S}/\text{cm}$. The major ions in the groundwater included Na^+ (721 ± 35 mg/L), Ca^{2+} (168 ± 19 mg/L), Mg^{2+} (40 ± 4 mg/L), K^+ (18 ± 0.7 mg/L), Cl^- (1370 ± 74 mg/L), SO_4^{2-} (297 ± 10 mg/L), and SiO_2 (28 ± 2 mg/L). The concentrations of other minor ions such as arsenite/arsenate, phosphate, iron, manganese, nitrate, nitrite, bromide, and fluoride, were negligible to impact electrodialysis performance or below detection limits. The pH of the groundwater was 7.78 ± 0.05 and the alkalinity was 90 ± 29 mg/L as CaCO_3 . The total organic carbon (TOC) concentration in the groundwater varied between 0.3 and 1.5 mg/L, and the water temperature was 26–27 °C. The high TDS and SAR (13–14) make the groundwater unsuitable for drinking and agricultural irrigation.

For each electrodialysis testing condition, water samples were collected from the feed-in, final product, inter-stage diluate-out and concentrate streams, and concentrate blowdown for analysis. Electrical conductivity and pH of the water samples were measured using a conductivity and pH meter (Model 431-61, Cole-Parmer, Vernon Hills, IL, USA). TOC was quantified using a carbon analyzer (Shimadzu TOC-L, Kyoto, Japan). Common cations and anions including sodium, calcium, magnesium, potassium, chloride, phosphate, sulfate, nitrate and nitrite were measured using an ion chromatograph (IC, ICS-2100, Dionex, Sunnyvale, CA, USA). The concentrations of other elements including aluminum, arsenic, boron, calcium, chromium, copper, iron, potassium, magnesium, manganese, lead, and selenium were quantified using an inductively coupled plasma mass spectrometry (ICP-MS, Elan DRC-e, PerkinElmer, Waltham, MA, USA). Alkalinity was measured using a digital titrator (Hach, CO, USA) and 1.6 N sulfuric acid standard solution. The TDS concentration was measured following the evaporation method at 180 °C after filtering the groundwater samples using a 0.45 μm cellulose acetate membrane filter (Toyo Roshi Kaisha, Ltd., Japan).

2.2. Bench- and pilot-scale electrodialysis systems

Bench- and pilot-scale electrodialysis systems were manufactured by GE. The electrodialysis pilot system (AQ3-1-4) is a scaled-down version of a full-scale electrodialysis reversal (EDR) water treatment plant, used to demonstrate treated water quality and to collect operational data for full-scale design. The pilot system could produce 3 to 13 gal per minute

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