



Modeling cell pair resistance and spacer shadow factors in electro-separation processes



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ABSTRACT

The electrical resistance of the stack is one of the most important parameters that affects the performance of reverse electrodialysis (RED¹) and other electro-separation processes. The stack is made up of many cell pairs, each cell pair consisting of alternating anion and cation exchange membranes separated by spacer-filled channels which allow fluid to flow across the membrane surfaces, provide mechanical support and maintain inter-membrane distance. The presence of non-conducting mesh in the spacer-filled channels increases the electrical resistance of the cell pairs; this so-called shadow effect can be characterized by the shadow factor (β) of the mesh. The shadow factor has not been determined experimentally so far. Furthermore, the extent of the shadow effect on the flow channels and membranes has not been investigated in much detail and the proposed analytical models in the literature are based only on assumptions. In this paper, we used a non-contact resistance (NCR²) method to measure the resistances of mesh filled spacer channels, membranes and sandwiched unit cells separately in high (1 M) and low (0.1 M) salinity NaCl solutions. The shadow factors of four different spacer mesh samples were determined experimentally. The existing methods of modeling the cell pair resistance were evaluated and the most suitable functional form for the resistances in series (RIS³) model was determined. While the mesh shadow factors were found to be independent of solution concentration, the membrane resistances were found to decrease with an increase in solution concentration. The mesh shadow factor was found to have little influence on the active area of the membranes. The NCR method could expedite the process of down selecting appropriate low resistance spacer meshes and membranes for the RED process. The validated RIS model can be used in techno-economic models for designing electro-separation processes for commercial scale plants.

1. Introduction

Electro-membrane or electro-separation processes are employed in a wide range of applications such as brackish water desalination, waste water treatment, food processing, heavy metal ion removal and recovery of acids/bases from salts [1,2]. The mechanism of mass transfer in many of these processes is by movement of ions across ion exchange membranes. Reverse electrodialysis (RED) is one such electro-membrane process that has been developed to produce energy by reversible mixing of two streams that have different salinities. This form of energy is called salinity gradient power (SGP⁴). Theoretical calculations have estimated the global potential of SGP due to the mixing of all rivers

with sea water to be around 2.6 TW [3]. Newer applications of the RED process include removal of contaminants like Cr(VI) [4] and improving the recovery of sea water desalination by reverse osmosis (SWRO⁵) process [5]. The Environment and Water Industry Programme (EWI⁶) 1 project was started in 2013 to investigate the feasibility of using RED, as part of a hybrid process for improving the recovery and reducing the cost of seawater desalination in Singapore. The high salinity brine and low salinity treated used water streams from co-located SWRO and waste water treatment plants respectively can be passed through RED stacks in a multi-stage operation to partially desalt the SWRO brine and recycle it in the SWRO plant for achieving nearly 100% recovery [6,7]. Renewable energy can also be produced as a by-product.

Abbreviations: RED, Reverse electrodialysis; NCR, Non contact resistance; RIS, Resistances in series; SGP, Salinity gradient power; SWRO, Sea water reverse osmosis; EWI, Environment and Water Industry Programme; CEM, Cation exchange membranes; AEM, Anion exchange membranes; EMF, Electromotive force; OCV, Open circuit voltage; MPD, Maximum power density; ED, Electrodialysis; EDI, Electrodeionization

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The RED process was first proposed by Pattle [8] in 1954. The so called hydroelectric pile (or stack) assembled with 47 cell pairs produced 15 mW power on passing 0.5 M NaCl and tap water through it. A typical RED stack is assembled using alternating cation (CEM⁺) and anion exchange membranes (AEM⁻) as well as mesh filled spacers for maintaining the inter-membrane gap. The role of cation and anion exchange membranes is to allow only the transfer of cations and anions respectively, while restricting the movement of oppositely charged ions. The function of spacers is to provide mechanical support to the membranes, form the flow channels for the concentrate and dilute streams and also promote the mixing of solutions through inertial forces imposed by the mesh filaments. When the concentrate and dilute streams flow in contact with the ion exchange membranes in alternate compartments of the cell, the difference in their chemical potential generates an electromotive force (EMF⁹) or open circuit voltage (OCV¹⁰) across the cell. When the circuit is closed, the cations and anions spontaneously move from the concentrate to the dilute streams through the cation and anion exchange membranes respectively. The stack consists of 1 set of cathode and anode which are separated from the cell pairs by electrode streams. The oxidation and reduction redox reactions of the electrode stream at the anode and cathode respectively convert the ionic current of the cell into electric current for the external circuit. When concentrate, dilute and electrode streams are passing through the stack, a resistor/load can be connected across the electrode terminals to draw a continuous supply of DC power.

The gross maximum power density (MPD¹¹) achieved during the RED process is a vital indicator of the stack performance. Experimental and modeling work on the RED process has revealed that the MPD of the stack depends on the salinity gradient between the concentrate and dilute streams, the membrane permselectivity and the cell pair resistance [9,10]. The ohmic resistance of the stack is equal to the sum of the resistances of individual spacer-filled channels and membranes by the resistances in series (RIS) concept, as the same current passes through all the stack components [11–13]. The flow channel resistance is proportional to the spacer thickness and inversely proportional to the solution conductivity. Experimental studies concluded that the dilute stream dominates the cell pair resistance when low salinity dilute solutions (~ 0.017 M NaCl solutions or river water) are used. Veerman et al. [14] and Danidilis et al. [15] performed RED experiments with 0.017 M and 0.01 M NaCl solutions respectively in the dilute stream and 0.5 M NaCl solutions in the concentrate stream. The contributions of the spacer-filled dilute channel resistances to the cell pair resistances in these two experiments were estimated to be 45% and 66% respectively. The impact of the dilute channel resistance can be reduced by using thinner spacers. Vermaas et al. [16] compared the performance of RED stacks assembled with four different types of spacers and observed that the ohmic resistance of the stack assembled with 0.1 mm thick spacers was 70% lower than the stack assembled with 0.485 mm thick spacers, for the same solutions in the feed streams (0.5 M/0.017 M NaCl solutions). As a result, the measured gross MPD of the 0.1 mm thick spacer stack was 300% higher than the other stack. However, the stack assembled with even thinner 0.06 mm spacers in the same set of experiments failed to produce desired results due to high pressure drop in the flow channels, which restricted the flow rate and affected the net power density.

The mesh used in conventional spacers is generally made of non-conducting materials like polymers which are known to cause additional stack resistance. This is referred to as the shadow effect and the evidence of this phenomenon was found in several experiments [16,17]. Belfort and Guter [18] investigated the resistance ratios of spacer-filled channels to that of the empty channel for 21 different spacer mesh samples in 0.05 N NaCl solution and observed resistance ratios between 1 and 5.7. Many authors have quantified the shadow effect in their process models using a shadow factor in the RIS equation. At least two different functional forms of RIS equation were proposed to incorporate the shadow effect of the spacer mesh, as there is no clear consensus on the nature of the shadow effect. Tanaka et al. [19]

presumed that the tortuous movement of ions in the spacer-filled channels results in an increase in the resistances of only the respective flow compartments. In contrast, Post et al. [20] and Lee et al. [21] assumed that the shadow effect also acts on the membranes by reducing the effective area available for the movement of ions. Some authors anticipated only a minor contribution of the shadow effect to the spacer-filled channel resistance and ignored it in their RIS equations [5,22,23]. Although the existence of the shadow effect has been acknowledged and extensively discussed in the literature, the relationship between the spacer shadow effect, mesh geometry and the cell pair resistance remains a grey area for electro-membrane process modeling. Researchers have made estimates on the shadow factor values based on the mesh geometries but these methods have not been validated so far [13,14]. As the spacer mesh is in direct contact with the membranes, the influence of shadow effect on the membrane resistance is also an interesting research topic.

Membrane conductivity determination is a popular research topic and detailed studies that co-relate membrane properties with membrane conductivity have been published [24–26]. Although thicker mesh spacers are known to dominate the cell pair resistance at very low concentrations of dilute stream, very little work has been done on developing characterization methods for spacer-filled channel resistance and shadow factor determination. This paper evaluates the relationship between spacer mesh, membrane and cell pair resistances in stacks assembled with mesh spacers. Four spacer mesh samples with different thickness and filament densities were used in this study. The spacer-filled channel and membrane resistances were experimentally measured in high and low salinity NaCl solutions separately (1 M and 0.1 M), using the non-contact resistance (NCR) method with alternating current (AC). The shadow factors of the four mesh samples were determined from the measured flow channel resistances. The dependence of ion exchange membrane resistances on NaCl solution conductivities was qualitatively analyzed. Furthermore, the RIS equation was validated and the functional form of cell pair resistance equation for stacks consisting of spacer-filled channels was determined by comparing the model values with the measured resistances of the sandwiched cell pairs. The method enables us to measure pure ohmic resistances of cell components as the boundary layer formation on the membrane surface is minimized by high frequency AC. In addition, salt and osmotic water transport phenomena are eliminated, as the same solution is present on both sides of the membrane. The NCR method facilitates quick evaluation and down selection of suitable low resistance spacer meshes and membranes for optimizing the RED process. The findings of this study lay the foundation for process models for predicting the stack performance and designing industrial scale electro-membrane processes.

2. Theory

2.1. Resistance in series (RIS) model

Fig. 1 illustrates the sectional view of a RED unit cell consisting of concentrate (C_C) and dilute (C_D) streams filled with oval shaped spacer mesh filaments (oriented at 45° with respect to the flow) separated by cation (C) and anion (A) exchange membranes. The flow channels and the ion exchange membranes in an assembled cell function as resistances in series for the ionic current. The equivalent electrical circuit of a cell pair is shown in the lower part of Fig. 1. Therefore, the resistance of the cell pair (R_{CP}) can be expressed as the sum of resistances-in-series (RIS) as shown in Eq. (1) [11,27,28].

$$R_{CP} = R_{CM} + R_{AM} + R_D + R_C \quad (1)$$

where R_{CM} and R_{AM} are the cation and anion exchange membrane resistances respectively, R_D and R_C are the dilute and concentrate spacer-filled channel resistances. The combined resistance of the solution and spacer mesh in spacer-filled channels is referred to as spacer resistance in the rest of this paper.

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