



## Electrodialysis with spacers: Effects of variation and correlation of boundary layer thickness

Younggy Kim<sup>1</sup>, W. Shane Walker<sup>2</sup>, Desmond F. Lawler<sup>\*</sup>

Department of Civil, Architectural and Environmental Engineering, The University of Texas at Austin, Austin, TX 78712, USA

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### ABSTRACT

The performance of electro dialysis is strongly dependent on the boundary layer thickness near an ion-exchange membrane. While a thinner boundary layer is known to enhance the ionic separation, the effects of statistical properties of the boundary layer thickness, such as the variation of the thickness and the correlation between the two boundary layers facing across the spacer, have not been elucidated. These effects were estimated by the Monte Carlo method incorporated into an analytical model. The analytical model simulates the binary ionic transport in four distinct regions of an electro dialysis cell pair: the bulk solution, boundary layer, ion-exchange membrane, and interface between the aqueous solution and ion-exchange membrane. The model current and potential relationships found that a greater variation or more positive correlation improves the ionic separation in the non-Ohmic regime. A bench-scale electro dialyzer was operated in a batch recycle system to develop steady-state current and potential relationships. Comparison between the model and experimental results found that the mean boundary layer thickness was tens of micrometers and the standard deviation of the thickness was similar to or greater than the mean thickness with the sheet-flow type mesh spacer.

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### 1. Introduction

For the treatment of brackish groundwater, electro dialysis or electro dialysis reversal is known to be versatile with respect to avoiding scaling and fouling problems [1]. This characteristic makes the technology an attractive complement of reverse osmosis to reduce the concentrate waste from an inland brackish groundwater desalination plant [2,3]. In addition, electro dialysis has extended its application to energy production from the mixing of saline and fresh waters [4,5] and to microbial desalination [6,7].

An electro dialyzer consists of two electrodes (anode and cathode), alternating anion-exchange membranes (AEMs) and cation-exchange membranes (CEMs), and spacers. The electrodes generate an electric field which drives the ionic transport in the electro dialyzer. An AEM contains such a high concentration of cations fixed in its polymeric textile that only anions can pass through it, and vice versa with a CEM. The spacers between the ion-exchange membranes (IEMs) provide space for the so-called cell. As salty water flows parallel to the IEMs in the cell, ions are driven by the electric field perpendicular to the IEMs. As ions are transported through the IEMs, one cell loses ions and the

adjacent cell collects ions. Together, they form a “cell pair” of a diluate and a concentrate cell, respectively, and an electro dialysis “stack” is a repetitive series of cell pairs.

Another important role of the spacers is to mix the aqueous solution in the cell. However, since the mixing is not complete within tens of micrometers from the IEM surface, one can assume a thin boundary layer where the hydrodynamic condition can be considered laminar (i.e., more precisely, the advection flux normal to the IEM surface is negligible). Without the advection flux in the boundary layer of the diluate cell, the ionic concentration decreases from the bulk solution to the IEM surface, so that the diffusion flux of co-ions (excluded ions from the IEM) cancels their electromigration flux. In the concentrate boundary layer, the concentration decreases from the membrane surface to the bulk solution due to the flux cancellation of co-ions. This phenomenon is called concentration polarization near IEMs. With a greater potential drop across the diluate boundary layer, the concentration gradient becomes steeper until the ionic concentration approaches zero at the IEM surface. The current that would cause zero concentration at the IEM surface is called the limiting current; it is equivalent to the maximum ionic separation rate that could be achieved in an electro dialyzer while maintaining electroneutrality in the boundary layer. Consequently, the electro dialyzer performance is directly controlled by the boundary layer thickness (i.e., a thinner boundary layer allows a steeper concentration gradient and a greater limiting current).

Because of the important role of the boundary layer in determining the maximum rate of ionic separation, many previous studies focused on characterizing the boundary layer in relatively simple IEM systems without spacers (e.g., a single IEM dividing an electrochemical cell into

\* Corresponding author. Tel.: +1 512 471 4595; fax: +1 512 471 5870.

E-mail addresses: [kimygy@gmail.com](mailto:kimygy@gmail.com) (Y. Kim), [wswalker2@utep.edu](mailto:wswalker2@utep.edu) (W.S. Walker), [dlawler@mail.utexas.edu](mailto:dlawler@mail.utexas.edu) (D.F. Lawler).

<sup>1</sup> Current address: Department of Civil and Environmental Engineering, The Pennsylvania University, University Park, PA 16802, USA.

<sup>2</sup> Current address: Department of Civil Engineering, The University of Texas at El Paso, El Paso, TX 79968, USA.

the cathode and anode sides). With the hydrostatic condition in those simplified IEM systems, the boundary layer thickness was found to be hundreds of micrometers [8–16]. On the other hand, Taky and coauthors [17,18] found that the boundary layer thickness was about 80 μm under a vigorously mixing condition. These results imply that the boundary layer thickness can vary by an order of magnitude depending on the hydrodynamic conditions.

Some studies were performed with an electro dialyzer stack including spacers to correlate the water flow rate and the boundary layer thickness. While the mean thickness was reported to be proportional to a reciprocal function of the water flow rate [19–22], the micro-scale variation in the thickness around the spacer geometry has not been addressed. Recently, the boundary layer was visualized by laser interferometry, and the thickness was found to vary significantly between 100 and 1000 μm along the rectangular spacer geometry [23,24]. Based on the substantial variation in the boundary layer thickness depending on the spacer geometry as well as the hydrodynamic condition, we suggest that the boundary layer thickness be considered as a statistical variable rather than a single mean value to properly simulate the electro dialysis performance.

In addition to the thickness variation, another important statistical property is the correlation between the two boundary layers facing across the spacer. Fig. 1 conceptually shows how the spacer geometry coupled with the water flow governs the correlation between the two boundary layer thicknesses. In the left cell of the figure, the asymmetric spacer geometry induces a negative correlation (i.e., one is thick, while the other is thin); in contrast, the boundary layers are positively correlated in the right cell due to the symmetric spacer geometry.

Almost all electro dialysis systems are built with spacers between the ion-exchange membranes. Given the important roles of the spacer geometry in governing the correlation as well as the variation of the boundary layer thickness, the effects of these statistical properties should be delineated. Thus, the objectives of this study are (1) to suggest that the boundary layer thickness should be represented by a random variable (not a uniform thickness) for model simulations of the ionic separation in electro dialysis, (2) to investigate how the current and potential relationship is influenced by both the standard deviation of the boundary layer thickness and the correlation coefficient between the two boundary layers facing across the spacer, and (3) to evaluate an approximate range of the mean and standard deviation of the boundary

layer thickness associated with a sheet-flow type mesh spacer in a bench-scale electro dialyzer.

## 2. Model development

### 2.1. One-dimensional model domain

The one-dimensional model built in Sections 2.2 through 2.5 simulates the steady-state ionic transport in an electro dialysis cell pair. As shown in Fig. 2, a cell pair includes an AEM, a CEM, two bulk solutions, and four boundary layers. A cell pair is a repetitive unit of an electro dialyzer; hence, the ionic transport in a cell pair can represent a whole electro dialysis system (except for the electrode reactions and the ionic transport in the electrode rinse cells).

The thickness of the boundary layer was assumed to be controlled by the hydrodynamic condition. According to several previous studies performed without spacers, the boundary layer thickness is considered as the extent of the concentration polarization from the IEM surface [25,26]. In this view, the boundary layer thickness is dependent on the current density. However, with the substantial mixing condition imposed by the spacer geometry and the flow velocity, we assume in this study that the boundary layer thickness is defined by the hydrodynamics, and the concentration polarization occurs within the defined thickness of the boundary layer.

The model for a binary ionic system consists of a closed-form mathematical expression that calculates the potential drop of each region of a cell pair (the bulk solution, boundary layer, AEM, CEM, and interface between the aqueous solution and IEM) as a function of a specified current density. By summing the potential drop in each region as in Eq. (1), the model finds the cell pair potential drop ( $\Delta\phi_{CP}$ ) at a given current density.

$$\Delta\phi_{CP} = \left[ \begin{matrix} \Delta\phi_{AEM} + \Delta\phi_{Donnan-1} + \Delta\phi_{BL-1} + \Delta\phi_{DB} + \Delta\phi_{BL-2} + \Delta\phi_{Donnan-2} \\ + \Delta\phi_{CEM} + \Delta\phi_{Donnan-3} + \Delta\phi_{BL-3} + \Delta\phi_{CB} + \Delta\phi_{BL-4} + \Delta\phi_{Donnan-4} \end{matrix} \right] \quad (1)$$

$\Delta\phi$  is the potential drop in volts. The subscripts *DB* and *CB* denote the diluate and concentrate bulk solutions, respectively. The subscript *Donnan* represents the Donnan potential drop at the interface between the aqueous solution and IEM, while *BL* abbreviates the boundary layer with the corresponding number in Fig. 2.

### 2.2. Potential drop across the bulk solution

In the bulk solution, the ionic concentration is assumed to be homogenous in the current direction; thus, the Nernst-Planck equation reduces to Eq. (2) with only the electromigration term. In an electrolyte, the current density (*I*) is carried by the ionic molar flux (*J*) as in Eq. (3), where the current density has the same sign as the cationic flux. Introducing Eq. (2) into Eq. (3) and integrating the resulting equation over the bulk width (*w<sub>B</sub>*) give the potential drop in the bulk solution ( $\Delta\phi_B$ ) as in Eq. (4).

$$J_i = -\frac{z_i D_i F}{RT} c_i \frac{d\phi}{dx} \quad (2)$$

$$I = F \sum_i z_i J_i \quad (3)$$

$$\Delta\phi_B = -\frac{IRT w_B}{F^2 \sum_i z_i^2 D_i c_i} \quad (4)$$

*D* is the diffusivity, *z* is the ionic charge, and *c* is the molar concentration for ionic species *i*, which is either *p* or *q* in this binary model indicating the cation and anion, respectively. *F* is the Faraday constant, *R* is the gas constant, and *T* is the absolute temperature. The independent variable *x* is the distance from the interface with the

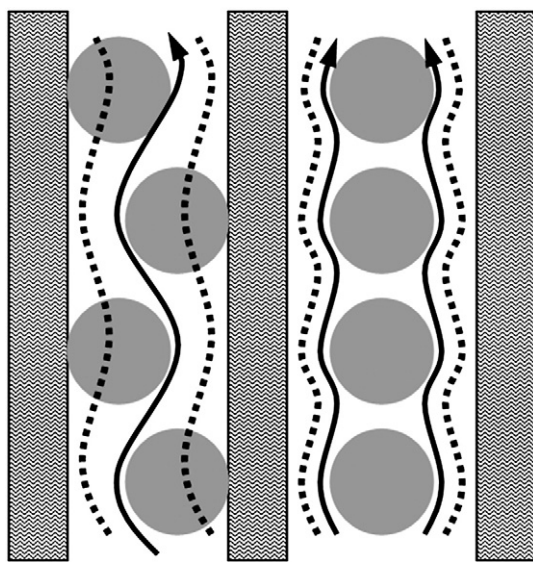


Fig. 1. Conceptual diagram for the correlation between two boundary layers facing across the spacer. (The circles indicate the spacer geometry; the solid lines are the stream lines of water flow; and, the dotted lines visualize the border of the boundary layer. In the right cell, the space between the spacer geometry and membrane for the water flow is maintained by other parts of the spacer that support the inter-membrane distance.)

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