



## Desalination at overlimiting currents: State-of-the-art and perspectives



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### HIGHLIGHTS

- Overlimiting current electro dialysis possibilities
- Understanding of the origin of electroconvection
- How current-induced convection enhances mass transfer
- Water splitting and its effects on electroconvection
- A way to enhance water desalination process

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### ABSTRACT

For about 60 years, it is known that "limiting current" density does not really limit the current density in electro dialysis (ED). The use of intensive currents allows one to decrease the area of expensive ion-exchange membranes, hence to obtain more compact ED plants with lower investment costs. However, in practice overlimiting currents are applied solely in electrodeionization, and not in conventional ED devices. In literature (H. Strathmann [1]), overlimiting current density operation ED is considered as being under ongoing research and development. Where is the research today? What is the actual understanding of overlimiting mass transfer processes in membrane systems? As far as electroconvection is involved as a major mechanism of overlimiting transfer, what could bring specific knowledge and practice acquired in nano- and microfluidics into desalination? Can overlimiting ED be economically advantageous? Is there an alternative for classical ED devices applied? This review is intended to give some responses to these questions.

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### 1. Introduction

While electro dialysis (ED) has been used industrially since the 50s of the last century [2] and considered as a mature process [1], there are a number of outstanding issues. Despite long-term development, ion-exchange membranes (IEMs) still have insufficient permselectivity, especially in concentrated solutions: too high resistance, particularly in solutions containing biomolecules/large organic anions; too high water splitting at anion-exchange membranes. Moreover, IEMs, especially anion-exchange ones, are not stable in food industry applications (dairy, wine, separation of amino acids, etc.). The membranes suffer from scaling and fouling during ED. The cost of IEMs is generally

high, and there are no low-cost and quality bipolar membranes, no membranes for high temperature ED.

It is sufficiently clear that ED operation with intensive current reduces the area of IEMs, hence, the investment costs of desalination. Less evident are other possibilities, which can be eventually realized when applying overlimiting currents. In particular, it can be imagined to help the control of water splitting as well as scaling/fouling by electrokinetic phenomena. Special design of membrane devices and the use of specific knowledge and practice acquired in nano- and microfluidics could revolutionize the practice of desalination. In this review, we will consider the fundamentals of concentration polarization phenomenon and the state-of-the-art in the field. Together with modern understanding of overlimiting transfer mechanisms, some special devices from microfluidics will also be reviewed. We will discuss as well some operational efficiencies as dependent on the current density and show that not all the characteristics of ED are improved with increasing current.

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### List of symbols

#### Abbreviations

CEDI	continuous electrodeionization
ICP	ion concentration polarization
LEN	local electroneutrality
NPP	Nernst–Planck and Poisson (equations)
NS	Navier–Stokes (equations)
QCD	Quasi-uniform charge density distribution
SCR	space charge region

#### Symbols

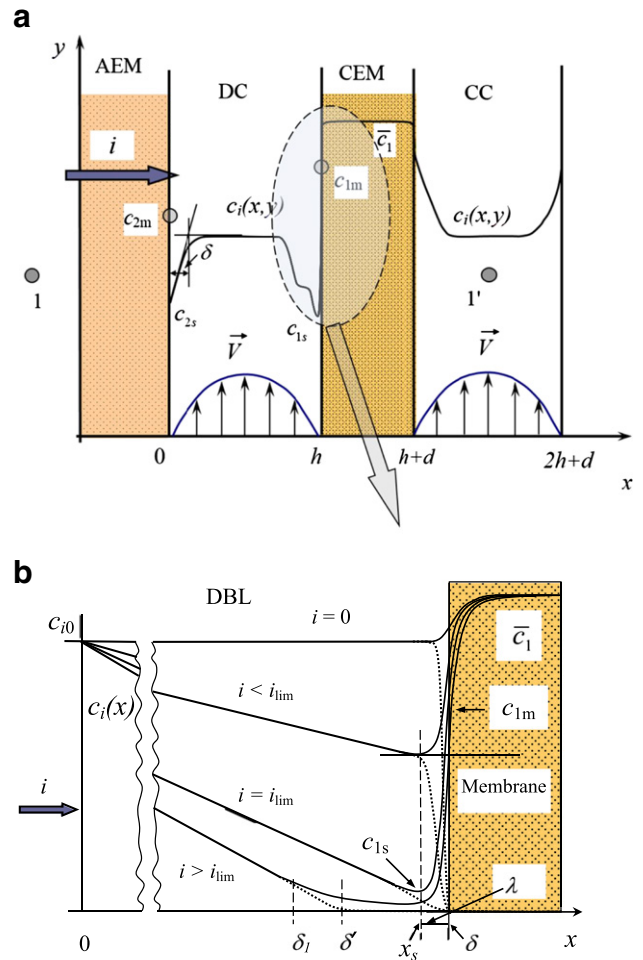
$c_i$	molar concentration of ion $i$
$c_{1s}^I, c_{1s}^{II}$	counterion concentrations at the left-hand and right-hand membrane surfaces
$c_{1m}$	counterion concentration at the inner boundary of solution electric double layer
$C_1$	dimensionless counterion concentration normalized by $c_{10}$
$d$	thickness of the membrane
$D$	electrolyte diffusion coefficient
$D_i$	diffusion coefficient of ion $i$
$E$	electric field intensity
$F$	Faraday constant
$h$	spacing between the membranes
$i$	current density
$i_{lim}$	limiting current density
$J_i$	ionic flux density
$L_D$	Debye length
$t$	time
$t_i$	transport number of ion $i$
$T$	absolute temperature
$T_i$	effective transport number of ion $i$ in membrane, Eq. (8)
$U =  \Delta\varphi $	absolute value of potential difference, voltage
$V$	fluid velocity
$x$	normal to membrane coordinate in ED cell
$y$	tangential coordinate in ED cell
$z_i$	charge number of ion $i$

#### Greek symbols

$\delta$	Nernst's diffusion layer thickness
$\delta'$	effective thickness of the diffusion boundary layer
$\nu$	fluid viscosity
$\varphi$	electric potential
$\Delta c$	concentration drop in the diffusion boundary layer

#### Indices

$i$	ionic species
$w$	species produced by water splitting
1	counterion



**Fig. 1.** a: Scheme of an electrodesalination cell with an anion-exchange (AEM) and a cation-exchange (CEM) membranes; DC and CC are the desalting and the concentrating compartments, respectively. The points 1 and 1' show where the tips of Luggin's capillaries (used for measuring the potential difference) are placed. Deformation of concentration profile at the CEM due to current induced convection is shown. b: Schematic view of salt counterion (solid line) and co-ion (dashed line) concentration profiles (in the framework of Rubinstein's 1D model) in the diffusion boundary layer of thickness  $\delta$  near a CEM at different current densities. The diffusion boundary layer is divided in an electroneutral ( $0 \leq x \leq \delta_1$ ) and a space charge ( $\delta_1 \leq x \leq \delta$ ) regions. The intersection of the straight line extending the linear concentration profile in the electroneutral region with the  $x$ -axis gives the effective thickness of diffusion boundary layer ( $\delta'$ );  $\lambda$  is the thickness of quasi-equilibrium boundary layer.

one membrane side and increases at the other due to selective transport of counterions in the membrane.

Generally, ion transport in solution or membrane is described by the extended Nernst–Planck equation with a convective term:

$$\vec{J}_i = -D_i \left( \nabla c_i + z_i c_i \frac{F}{RT} \nabla \varphi \right) + c_i \vec{V}, \quad i = +, -, H, OH \quad (1)$$

where  $\vec{J}_i$ ,  $D_i$ ,  $z_i$  and  $c_i$  are the flux density, diffusion coefficient, charge number and concentration of ionic species  $i$ , respectively;  $\varphi$  is the electric potential,  $\vec{V}$  the fluid velocity vector;  $F$ ,  $R$  and  $T$  have their usual meanings. In this paper the subscript  $i$  may be attributed to the salt cation (+) or anion (−) as well to  $H^+$  (H) or  $OH^-$  (OH) ions. The first term in the right-hand side of Eq. (1) represents diffusion, the second, migration, and the third, convection.

## 2. Concentration polarization of ion-exchange membranes. Brief theoretical aspects

Schematically, a conventional ED elementary cell pair is shown in Fig. 1. It includes an anion-exchange membrane (AEM), a cation-exchange membrane (CEM) and solutions flowing between them. When an electric current passes through an ion-selective membrane separating two electrolyte solutions, salt concentration decreases at

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