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Continuous ion-exchange membrane electrodialysis for drinking water production: Numerical analysis

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ABSTRACT

Computer simulation program of a continuous electrodialysis process is developed for saline water desalination. By inputting operating parameters into the program, the electrodialyzer is operated applying constant voltage to produce drinking water. Changing voltage step by step, the performance such as; ion and solution flux across a membrane pair; salt concentration in desalting and concentrating cells; current efficiency; water recovery; energy consumption; limiting current density are calculated. Electrodialysis conditions are discussed for producing drinking water meeting the WHO drinking water standard at under limiting current density stably. Standard deviation of solution velocity ratio between desalting cells functions as a safety factor in operating an electrodialyzer.

1. INTRODUCTION

Ion-exchange membranes are applied in electrodialysis to desalinate and concentrate salt solutions. The membranes are also applicable to separate ionic species in the solutions. Desalination is a fundamental technology in ion-exchange membrane electrodialysis, and its industrial applications extend in many fields at present. Electrodialysis processes are classified to a continuous, a batch and a feed-and-bleed process. Among these processes, the continuous process is the most fundamental one and it is applied to a large-scale electrodialysis. The performance of the continuous process is discussed from various points so far (Belfort 1970), (Avriel 1972), (Lee 2002), (Moon 2004), (Fidaleo 2005), (Sadradeh 2007), (Nikonenko 2008), (Brauns 2009).

In the previous investigation, the electrodialysis program of the continuous process was established for discussing the performance of saline water desalination processes (Tanaka 2009), (Tanaka 2010a). The program was applied to evaluate the performance of sea water concentrating plants (Tanaka 2010b). The influence of temperature on the performance of an

electrodialyzer was discussed (Tanaka 2011a). Data obtained by computation were compared to data observed by operating a practical-scale electrodialyzer for seawater concentration (Tanaka 2011b).

From the above investigations, the reasonability of the electrodialysis program is assumed to be generally demonstrated. The program consists of many steps describing many unit processes and it was established based on many fundamental principles and experiments. Precision of the program have been improved every time in the course of the program development. Consequently, the description of the program in the previous articles becomes partially insufficient, so the description becomes necessary to be supplemented. This article describes the supplemented program in which the principle of the continuous process is briefly explained. Further, an electrodialyzer is operated by applying constant voltage and by supplying 1000 mg/l and 25 $^{\circ}$ C raw saline water. Changing voltage applied step by step, the performance of the electrodialyzer such as; ion and solution flux across a membrane pair; salt concentration in desalting and concentrating cells; current efficiency; water recovery; energy consumption; limiting current density are computed. Electrodialyzer operating conditions are discussed for producing drinking water meeting the WHO drinking water standard at under limiting current density.

2. CONTINUOUS ELECTRODIALYSIS PROCESS

An electrodialyzer is operated with a constant electric current *I* as illustrated in Fig. 1. A salt solution (raw salt solution, concentration: C'_{in}) is supplied to the inlets of desalting cells (De) at average linear velocity of u'_{in} . For preventing scale formation in concentrating cells, a part of a raw salt solution is supplied also to the inlets of concentrating cells (Con) at the average linear velocity of u'_{in} . Ions and solutions transfer from desalting cells to concentrating cells across an ion-exchange membrane pair and their flux is defined by J_S and J_V respectively. In desalting (concentrating) cells, salt concentration is decreased (increased) from C'_{in} ($C''_{in} = C'_{in}$) under applied average current density I/S (I; electric current, S; membrane area) and reaches average salt concentration C'_{out} (C''_{out}) at the outlets of desalting (concentrating) cells. Salt concentration density change along the flow-pass from i_{in} at the inlets to i_{out} at the outlets. The current density becomes i at x distant from the inlets of desalting cells. I/S, J_S , J_V , C'_p , C''_p , u'_p and u''_p are altogether the values at x = pl distant from the inlets of desalting cells. V_{in} , V_{out} and V_p are voltage difference between electrodes respectively at the inlets (x = 0), the outlets (x = l) and x = pl of desalting cells ($V_{in} = V_p = V_{out}$).



De: Desalting cell, Con: Concentrating cell K: Cation-exchage membrane, A: Anion-exchange membrane J_s, J_v : Fluxes of ions and solutions across membrane pairs at x = pl C'_p, C''_p : Electrolyte concentration in desalting and concentrating cells at x = pl u'_p, u''_p : Linear velocity in desalting cells and concentrating cells at x = pl

Fig. 1. Continuous process

3. ELECTRODIALYSIS PROGRAM

Main equations describing the phenomena in the continuous process are enumerated with brief explanation.

3.1. Overall mass transport equation

Flux of ions J_S and a solution J_V across an ion-exchange membrane pair at x = pl are expressed by the following overall mass transport equation (Tanaka 2006).

$$J_{S} = \lambda \left(I/S \right) - \mu \left(C^{"}_{p} - C^{'}_{p} \right) = \eta \left(I/S \right)/F$$
(1)

$$J_V = \phi(I/S) + \rho(C''_p - C'_p)$$
⁽²⁾

Membrane pair characteristics λ , μ , ϕ and $2r_{alter} = r_K + r_A$ are expressed by the functions of ρ :

$$\lambda = 9.208 \times 10^{-6} + 1.914 \times 10^{-5} \,\rho \tag{3}$$

$$\mu = 2.005 \times 10^{-4} \,\rho \tag{4}$$

$$\phi = 3.768 \times 10^{-3} \rho^{0.2} - 1.019 \times 10^{-2} \rho$$
(5)

$$2r_{alter} = r_{alter,K} + r_{alter,A} = 1.2323 \ \rho^{-(1/3)}$$
(6)

 ρ is expressed by the function of temperature *T* (Tanaka 2011a):

$$\rho = 3.421 \times 10^{-3} + 3.333 \times 10^{-4} T \tag{7}$$

3.2. Salt concentration and linear velocity in desalting cells

The following equations are introduced.

$$u'_{out} = Q'_{out}/ab = (Q'_{in} - blJ_V)/ab$$
(8)

$$u'_{p} = u'_{in} - p(u'_{in} - u'_{out}) = u'_{in} - p(q/ab)$$
(9)

$$C'_{out} = C'_{in} - \left(\frac{\eta l}{aF}\right) \left(\frac{1}{u'_p}\right) \left(\frac{I}{S}\right)$$
(10)

Frequencies Y_j of the linear velocity ratio ξ_j in desalting cell group j is defined as;

$$\xi_j = \frac{u_j - u}{u'} \tag{11}$$

 u'_{j} : linear velocity in the desalting cells in group j, u': average linear velocity in desalting cells.

Frequencies Y_j of ξ in group j; ξ_j is expressed by the normal distribution (standard deviation: σ) (Tanaka 2000), (Tanaka 2005). Many desalting cells are integrated in a stack in an electrodialyzer. ξ_j is divided among j = 0 - n according to Eq. (12).

$$\xi_j = -3\sigma \left(1 - \frac{j}{n/2}\right), \qquad j = 0 - n \tag{12}$$

Linear velocity at x = pl distant from the inlets of j^{th} desalting cells is

$$u'_{pj} = u'_{p}(\xi_{j} + 1)$$
(13)

Accordingly

$$C'_{out,j} = C'_{in,j} - \left(\frac{\eta l}{aF}\right) \frac{1}{u'_p(\xi_j + 1)} \left(\frac{I}{S}\right)$$
(14)

Salt concentration at x = pl is:

$$C'_{p} = \frac{1}{N} \sum_{j=0}^{n} Y_{j} C'_{p,j}$$
(15)

$$C'_{p,j} = C'_{in} - \left(\frac{\eta l}{aF}\right) \left\{ \frac{1}{u'_{p/2}(\xi_j + 1)} \right\} \left\{ a_1 p + \left(\frac{a_2}{2}\right) p^2 + \left(\frac{a_3}{3}\right) p^3 \right\}$$
(16)

3.3. Salt concentration and linear velocity in concentrating cells

Saline water supplied to desalting cells is also supplied to concentrating cells.

$$C''_{out} = \frac{\sqrt{M^2 + 4LN} - M}{2L}$$
(17)

$$C''_{p} = C''_{in} + p(C''_{out} - C'_{in})$$
(18)

$$u''_{out} = \frac{1}{2} \left(\sqrt{M^2 + 4LN} + M \right)$$
(19)

$$u''_{p} = u''_{in} + p(u''_{out} - u''_{in})$$
⁽²⁰⁾

in which

$$L = \frac{l}{a} \rho p \tag{21}$$

$$M = \frac{l}{a} \left[\phi \left(\frac{I}{S} \right) - \rho \left\{ C'_{p} - (1 - p) C'_{in} \right\} \right] + u''_{in}$$
(22)

$$N = C''_{in} u''_{in} + \frac{l}{a} \left\{ \lambda \left(\frac{I}{S} \right) - \mu \left(C''_{p} - C'_{p} \right) \right\}$$
(23)

3.4. Electric resistance of an ion-exchange membrane pair and solutions in desalting and concentrating cells

Direct current electric resistance of an ion exchange membrane r_{mem} is calculated using the following equations (Tanaka 2003).

$$\log\left(\frac{r_{dire}^{*}}{r_{alter}}\right) = 0.3380 + 0.6386\kappa' + 0.2961(\log\kappa')^{2}$$
(24)

$$\frac{r_{dire}}{r_{dire}^{*}} = 1.000 - 0.1359 \left(\frac{\kappa''}{\kappa'}\right)$$
(25)

$$r_{memb} = \left(\frac{r_{dire}^{*}}{r_{alter}}\right) \times \left(\frac{r_{dire}}{r_{dire}^{*}}\right) \times 2r_{alter} = 2r_{dire}$$
(26)

- r_{alter} ; alternating current electric resistance of a membrane.
- r^*_{dire} ; direct current electric resistance of a membrane measured in a diluate/diluate solution system.
- r_{dire} ; direct current electric resistance of a membrane measured in a diluate/concentrate solution system.

Electric resistance of a desalting cell r' and of a concentrating cell r'' in an electrodialyzer are;

$$r' = \frac{a}{(1-\varepsilon)\kappa'} \tag{27}$$

$$r'' = \frac{a}{(1-\varepsilon)\kappa''} \tag{28}$$

 ε is the volume ratio of a diagonal net spacer defined by;

$$\varepsilon = \frac{\pi a}{8\chi \sin \theta} \tag{29}$$

 χ ; distance between spacer rods. θ ; crossing angle of the rods.

3.5. Current density distribution

Current density distribution is assumed to be approximated by the following quadratic equation expressed at x/l distant from the inlet of a desalting cell (Tanaka 2000).

$$i = a_1 + a_2 \left(\frac{x}{l}\right) + a_3 \left(\frac{x}{l}\right)^2 \tag{30}$$

To determine a_1 , a_2 and a_3 in Eq. (30), the following three-simultaneous equations are set up taking account of voltage difference between electrodes.

$$V_{in} = V_{out} \tag{31}$$

$$V_{in} = V_p \tag{32}$$

$$\zeta_{inout} = \zeta_{inp} \tag{33}$$

 ζ_{inout} is introduced from Eq. (31) and expressed by Eq. (34), which is equals to ζ_{out} (outlet electric current non-uniformity coefficient).

$$\zeta_{inout} = \frac{\alpha_1 + \alpha_2 p + \alpha_3 p^2}{\beta_1 + \beta_2 p + \beta_3 p^2} = \zeta_{out} = \frac{i_{out}}{I/S}$$
(34)

 ζ_{inp} is introduced from Eq. (32) and expressed by Eq. (35), which is also equals to ζ_{out} .

$$\zeta_{inp} = \frac{\gamma_1 + \gamma_2 p + \gamma_3 p^2}{\left(2p - 3p^2\right)\left(I/S\right)} = \zeta_{out}$$
(35)

We determine a_1 , a_2 , a_3 , ζ_{in} (inlet electric current non-uniformity coefficient), ζ_{out} , i_{in} and i_{out} defined in the following equations.

$$\zeta_{in} = \frac{i_{in}}{I/S} = \frac{a_1}{I/S} \tag{36}$$

$$\zeta_{out} = \frac{i_{out}}{I/S} = \frac{a_1 + a_2 + a_3}{I/S}$$
(37)

3.6. Cell voltage, energy consumption, water recovery, desalting ratio

Ohmic voltage and membrane voltage at the inlet of the desalting cell ($V_{\Omega,in}$ and $V_{memb,in}$) and those at the outlet of the desalting cell ($V_{\Omega,out}$ and $V_{memb,out}$) are:

$$V_{\Omega,in} = \left(r'_{in} + r_{memb,in} + r''\right)i_{in}$$
(38)

$$V_{memb,in} = 2\left(t_K + t_A - 1\right) \times \left(\frac{RT}{F}\right) \ln \frac{\gamma_{in}^{"}C_{in}^{"}}{\gamma_{in}^{'}C_{in}^{"}}$$
(39)

$$V_{\Omega,out} = \left(\sum_{j=0}^{n} Y_j r'_{out,j} + \sum_{j=0}^{n} Y_j r_{memb,out,j} + r'_{out} N\right) \times i_{out} \times \left(\frac{1}{N}\right)$$
(40)

$$V_{memb,out} = 2\left(t_K + t_A - 1\right) \left(\frac{RT}{F}\right) \sum_{j=0}^n \ln \frac{\gamma_{out}^{"} C_{out}^{"}}{\gamma_{out,j}^{'} C_{out,j}^{'}} \times \left(\frac{1}{N}\right)$$
(41)

Cell voltage V_{cell} is introduced from Eqs. (38) – (41) as:

$$V_{cell} \left(V/pair \right) = V_{\Omega,in} + V_{memb,in} = V_{\Omega,out} + V_{memb,out}$$
(42)

Energy consumption E is expressed by the following equation.

$$E\left(kWh/m^{3}\right) = \frac{V_{cell}I}{Q_{out}} \times 3600 \times 10^{3}$$
(43)

 Q'_{out} is the solution volume; output of the desalted solution.

Water recovery Re is:

$$\operatorname{Re} = \frac{Q'_{out}}{Q'_{in} + Q''_{in}}$$
(44)

Desalting ratio α is:

$$\alpha = \left(1 - \frac{C'_{out}}{C'_{in}}\right) \tag{45}$$

3.7. Limiting current density

Limiting current density of a cation-exchange membrane is less than that of an anion-exchange membrane, because the mobility of counter ions in a solution for a cation-exchange membrane is less than that for an anion-exchange membrane. So the limiting current density of an ion-exchange membrane integrated in an electrodialyzer i_{lim} is given by the following empirical equation established for a cation-exchange membrane (Tanaka 2005).

$$i_{\rm lim} = (m_1 + m_2 u'_{out}) C'_{out}^{n_1 + n_2 u'_{out}}$$
(46)

$$m_1 = 83.50, m_2 = 24.00, n_1 = 0.7846, n_2 = 8.612 \times 10^{-3}$$

When current density reaches the limit of a cation-exchange membrane i_{lim} at the outlet of a desalting cell in which linear velocity becomes the least among u'_{out} : $u'_{out}^{\#}$, the average current

density applied to an electrodialyzer is defined as its limiting current density $(I/S)_{\text{lim}}$ which is expressed by Eq. (47) introduced from Eqs. (37) and (46) putting $u'_{in}^{\#} = u'_{out}^{\#}$.

$$\left(\frac{I}{S}\right)_{\lim} = \frac{i_{\lim}}{\zeta_{out}} = \frac{m_1 + m_2 u_{in}^{\#}}{\zeta_{out}} \left(C_{out}^{\#}\right)^{n_1 + n_2 u_{in}^{\#}}$$
(47)

in which C'_{out} [#] is C'_{out} at $u' = u'_{in}$ [#] which is given by;

$$u'_{in}^{\ \#} = u'_{in}(1 - 3 \sigma)$$
 (48)

The relationship between $(I/S)_{lim}$ and C'_{out} [#] is also introduced as follows

$$\left(\frac{I}{S}\right)_{\lim} = \left(\frac{a}{\lambda l}\right) u_{in}^{*} \left(C_{in}^{*} - C_{out}^{*}\right)$$
(49)

Putting Eq. (47) = Eq. (49):

$$Z_{1} = \frac{\left(C_{out}^{\dagger \#}\right)^{n_{1}+n_{2}u_{in}^{\#}}}{C_{in}^{\dagger}-C_{out}^{\dagger \#}}$$
(50)

$$Z_2 = \left(\frac{a\zeta_{out}}{\lambda l}\right) \left(\frac{u_{in}^{**}}{m_1 + m_2 u_{in}^{**}}\right)$$
(51)

$$Z_1 = Z_2 \tag{52}$$

Limiting current density of an electrodialyzer $(I/S)_{\text{lim}}$ is computed with trial and error calculation by substituting control key $C'_{out}^{\#*}$ for $C'_{out}^{\#}$ in Eq. (50) to realize $Z_1 = Z_2$ (Eq. (52)) for determining $C'_{out}^{\#}$. $(I/S)_{\text{lim}}$ is calculated by substituting $C'_{out}^{\#}$ into Eq. (47).

4. DRINKING WATER PRODUCTION

4.1. Operating conditions

The computation is carried out according to the electrodialysis program (Fig. 2) by inputting the following parameters into the programs keeping cell voltage V_{cell} constant.



Fig. 2. Electrodialysis program

Flow-pass thickness of a desalting and a concentrating cell a = 0.05 cm

Flow-pass width of a desalting and a concentrating cell b = 100 cm

Flow-pass length of a desalting and a concentrating cell l = 100 cm

Solution temperature $T = 25^{\circ}C$

Salt concentration at the inlets of desalting cells and concentrating cells $C'_{in} = C''_{in} = 1000 \text{ mg/}l$ Linear velocity at the inlets of desalting cells $u'_{in} = 10 \text{ cm/s}$

Linear velocity at the inlets of concentrating cells $u''_{in} = 1$ cm/s

Standard deviation of the normal distribution of solution velocity ratio $\sigma = 0, 0.05, 0.1, 0.15$

In decision point 1 in Fig.2, trial-and-error calculations are carried out to realize $C'_p = C''_p^*$ (control key) and $C''_p = C''_p^*$ (control key). In decision point 2, the calculations go on to realize $\zeta_{inout} = \zeta_{inp}$ (Eq. (33)) by substituting p^* (control key). In decision point 3, the calculations are continued to realize $V_{cell} = V_{cell}^*$ by inputting V_{cell}^* (control key) and I/S^* (control key).

 V_{cell} is changed step by step in the constant voltage operation.

4.2. Results

Fig. 3 shows ions flux J_S and solution flux J_V across a membrane pair. The data are independent of σ , so we recognize that the fluxes are not influenced by σ , *i.e.* the fluxes are not influenced by solution velocity distribution between desalting cells.

Fig. 4 shows salt concentration at the outlets of concentrating cells C''_{out} and salt concentration passing through the membrane pair C''^{\flat} . C''_{out} is drastically decreased comparing to C''^{\flat} . This phenomenon is due to that the feeding solution is supplied to concentrating cells for preventing scale formation in concentrating cells.

Fig.5 shows energy consumption E, which is increased with V_{cell} and not influenced by σ .

I/S and $(I/S)_{lim}$ are plotted against V_{cell} and shown in Fig. 6. Real limiting current density $(I/S)_{lim}$ and real limiting cell voltage $V_{cell,lim}$ at real $(I/S)_{lim}$ of the electrodialyzer is determined from the intersection between the a I/S line and a $(I/S)_{lim}$ line in the figure. The real limiting current density $(I/S)_{lim}$ and real cell voltage $V_{cell,lim}$ depend on σ and they are listed in Table 1.

Salt concentration at the outlets of desalting cells C'_{out} is decreased with the increase of V_{cell} (Fig. 7). We determine salt concentration at the outlets of desalting cells as $C'_{out} = 500$ (WHO drinking water standard), 400, 300 mg/l. Operating cell voltage $V_{cell,ope}$ for producing drinking water ($C'_{out} = 500, 400, 300 \text{ mg/l}$) is given in Fig. 7 and it is listed in Table 1.

An electrodialysis operation is assumed to become unstable ($V_{cell,ope} > V_{cell,lim}$) in the following conditions.

 $\sigma = 0.1, C'_{out} = 300 \text{ mg/}l$ $\sigma = 0.15, C'_{out} = 400, 300 \text{ mg/}l$



Fig. 3. Ion flux and solution flux across a membrane pair.



Fig. 4. Salt concentration of a solution at the outlets of concentrating cells and passing through membrane pairs.







Fig. 6. Current density, limiting voltage and limiting current density.



Fig. 7. Salt concentration at the outlets of desalting cells.

 Table 1. Limiting cell voltage and operating cell voltage.

σ		0.00	0.05	0.10	0.15
$(I/S)_{lim}$	A/dm^2	undeter-	0.715	0.584	0.462
V _{cell,lim}	V/pair	mined	1.035	0.709	0.510
C'out		V cell, ope			
mg/ <i>l</i>		V/pair			
500		0.496	0.496	0.499	0.505
400		0.636	0.638	0.644	0.660
300		0.817	0.822	0.855	0.917

 σ is influenced extremely by the precision of part dimensions of a stack and skill of stack assembling work of an electrodialyzer. It is desirable to keep σ values as low as possible, but we can not exactly control σ values in an electrodialysis operation. However, if we assume that σ functions as a safety factor in operating an electrodialyzer, it is significant to determine an appropriate σ value for realizing an stable operation of an electrodialyzer. From this view point, we define $\sigma = 0.1$ as the safety standard deviation factor.

5. CONCLUSION

Electrodialysis program for saline water desalination is developed. 1000 mg/l and 25°C saline water is electrodialyzed for producing drinking water stably to meet WHO drinking water standard. Limiting current density of the electrodialyzer is influenced strongly by the standard deviation of the solution velocity ratio between desalting cells σ . $\sigma = 0.1$ is defined as a safety factor for operating an electrodialyzer stably.

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NOMENCLATURES

- *a* flow-pass thickness in a desalting and a concentrating cell (cm)
- *b* flow-pass width in a desalting and a concentrating cell (cm)
- *C* electrolyte concentration (eq cm⁻³, eq dm⁻³, mg l^{-1})
- E energy consumption (kWh m⁻³)
- F Faraday constant (As eq⁻¹)
- *i* current density (Acm⁻², Adm⁻²)
- *I* electric current (A)
- I/S average current density (Acm⁻², Adm⁻²)
- J_S ion flux across a membrane pair (eq cm⁻²s⁻¹)
- J_V solution flux across a membrane pair (cm³cm⁻²s⁻¹)
- *n* divided group number of desalting cells in a stack; j = 0 n
- *N* number of desalting cells in a stack
- *p* dimensionless distance from the inlet of a desalting cell at which current density is equal

to the average current density I/S of an electrodialyzer

- Q solution volume (cm³s⁻¹)
- *r* electric resistance ($\Omega \,\mathrm{cm}^2$)

 r_{memb} direct current electric resistance of a membrane pair ($\Omega \text{ cm}^2$)

R gas constant (JK⁻¹mol⁻¹)

Re water recovery

S ion-exchange membrane area (cm^2)

t transport number of ions in a membrane

T temperature (°C, K)

u linear velocity in desalting and concentrating cells (cm s⁻¹)

 V_{cell} cell voltage (V pair⁻¹)

 V_{memb} membrane potential (V pair⁻¹)

 V_{Ω} Ohmic potential (V pair⁻¹)

x distance from the inlet of a desalting cell (cm)

 Y_j frequency of the linear velocity ratio ξ_j in desalting cell group j

Greek letters

- α desalting ratio
- γ activity coefficient of electrolytes in a solution
- ϵ volume ratio of a spacer in a desalting and a concentrating cell
- ζ current density non-uniformity coefficient
- η current efficiency
- κ specific conductivity of a solution (mS cm⁻¹)
- θ crossing angle of spacer rods (radian)
- λ overall transport number of a membrane pair (eq A⁻¹s⁻¹)
- μ overall solute permeability of a membrane pair (cm s⁻¹)
- ξ linear velocity ratio of solutions in desalting cells
- ρ overall hydraulic permeability of a membrane pair (cm⁴eq⁻¹s⁻¹)
- σ standard deviation of normal distribution of linear velocity ratio ξ
- ϕ overall electro-osmotic permeability of a membrane pair (cm³A⁻¹s⁻¹)
- χ distance between spacer rods (cm)

Subscripts

- A anion-exchange membrane
- alter alternating current electric resistance
- dire direct current electric resistance

- in inlet of a desalting and a concentrating cell
- j group j in the normal distribution within the range of $\xi_j \Delta \xi_j < \xi < \xi_j + \Delta \xi_j$
- K cation-exchange membrane
- lim limiting current density
- out outlet of a desalting and a concentrating cell
- p point x = pl distant from the inlet of a desalting cell

Superscripts

- ' desalting cell
- " concentrating cell
- * control key
- # desalting cell in which solution velocity becomes the least
- b solution passing through a membrane pair