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Time dependence of transport number ratio during electrodialysis process

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ABSTRACT

The transport number ratio between anion and chloride ion P^a_{Cl} was measured to discuss the anion permselectivity in an electrodialysis (ED). It was shown that P^a_{Cl} changed with the progress of ED. In this paper, a mechanism of anion permselectivity in ED was theoretically discussed with an equivalent circuit to find the reason why P^a_{Cl} changed with the progress of ED. It was found that P^a_{Cl} depended on the total electrolyte concentration of the diluted compartment C_B. Then, P^a_{Cl} changes with the progress of ED, since C_B decreases with the progress of ED. In addition, the simulation of ED with the equivalent circuit was successfully performed using the electric resistances of the equivalent circuit obtained from the analysis of P^a_{Cl}.

Keywords: Electrodialysis; Permselectivity; Transport number; Equivalent circuit; Simulation; Desalination

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1. Introduction

Recently, water shortage problem has become more and more serious in the world [1]. The desire to make a drinking water by treating a ground water, a surface water, a sea water and so on has been increasing. An electrodialysis (ED) is one of the useful methods and has applied to make the drinking water as well as RO and NF membranes. Especially ED is utilized to deionize harmful ions like F^- and NO_3^- [2–5].

ED is an electrical system utilizing ion exchange membranes. Ion exchange membranes have ion permselectivity and deionization rate depends on ionic species in ED [6]. It is very important to understand the mechanism of permselectivity of ion exchange membranes to design ED system. The transport number ratio between anion and chloride ion has been treated to discuss the permselectivity of anion exchange membrane in ED [7]. It is considered that the transport number ratio is constant determined by ionic species and a membrane. The Gibbs hydration energy is considered to be a main factor that determines the transport number ratio [8]. Recently, it has been found that the ratio of transport number between anion and chloride ion changes with the progress of ED. In this paper, a mechanism of anion permselectivity in ED is theoretically discussed with an equivalent circuit to make clear the reason why the transport number ratio changes with the progress of ED. In addition, the simple way to simulate ED is proposed.

2. Theoretical

2.1. Model of electrodialysis system

The transport number ratio between anion *a* and chloride ion P^a_{Cl} is defined by Eq.(1) [7]:

$$P^a_{Cl} = (t_a/[a]_B)/(t_{Cl}/[Cl]_B) = ([J_a]/[J_{Cl}])/([Cl]_B/[a]_B) \quad (1)$$

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where t_a and t_{Cl} denotes the transport number of anion a and Cl ion, respectively. $[a]_B$ and $[Cl]_B$ are the concentration (eq/m^3) of anion a and Cl in the diluted compartment, respectively. $[J_a]$ and $[J_{Cl}]$ denotes the flux (eq/m^{2}s) of anion a and Cl, respectively.

In ED, the ion flux is determined by the electric current that flows through the system. Fig. 1 shows the fundamental system of ED. When the voltage is applied to the system where the feed solution is divided by the pair of anion exchange membrane (AEM) and cation exchange membrane (CEM), anions are migrated to anode through AEM while cations are migrated to cathode through CEM. However, anions cannot permeate CEM and cations cannot permeate AEM because of an electric repulsion between ions and membrane charge. Consequently, the ion concentration of the central compartment between AEM and CEM is decreased, since ions are removed from the compartment. The central compartment is called as a diluted compartment. The several pairs of AEM and CEM are used in the practical ED system. So, the ion concentration is increased during ED in the compartment next to the diluted compartment. This compartment is called as a concentrated compartment. The electric current that flow through the system is the same at everywhere within the system. Thus, in the constant current system, it is possible to calculate the flux based on the system shown in Fig. 1. The calculated result is converted to the practical system by multiplying the number of pairs of AEM and CEM.

According to the Faraday's law of electrolysis, the amount of desalinated ions is determined by the total amount of current that flows through the system. Then, the equivalent circuit shown in Fig. 2 is proposed assuming the system contains n kinds of electrolytes. All electrolytes are assumed to be univalent for theoretical simplification. In addition, it is assumed that the concentration distribution in the diluted compartment is homogeneous and the membrane charge density of AEM and CEM is extremely higher than the electrolyte concentration of concentrated and diluted compartments. In Fig. 2, $R^K{}_i$ denotes the electric resistance of CEM for cation i, $R^A{}_a$ the electric resistance of AEM for anion a and R_{dilu} the electric resistance of diluted compartment. Subscript n (figures) denotes nth electrolyte. Each of 1st to nth circuits corresponds to 1st to nth electrolytes, respectively.

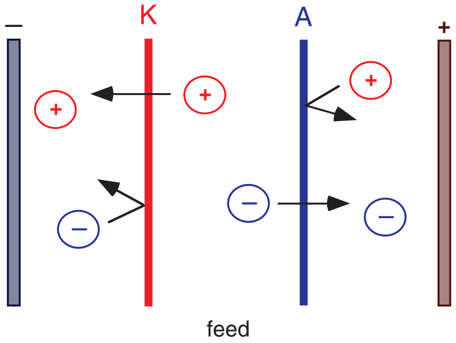

Fig. 1. Fundamental system of ED.K: cation exchange membarane (CEM), A: anion exchange membrane (AEM).

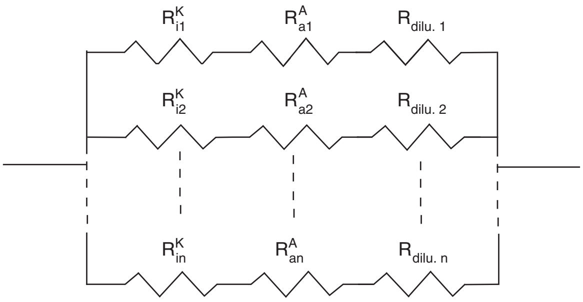

Fig. 2. Equivalent circuit of ED. $R^K{}_{in}$ and $R^A{}_{an}$ denote the electric resistance of cation and anion exchange membranes for ion transport, respectively. $R_{dillu.n}$ is the electric resistance of the diluted compartment due to nth electrolyte.

2.2. Transport number ratio

According to Fig. 2, the flux of n^{th} electrolyte is obtained from the electric current that flows through the nth circuit. The externally applied voltage E (V) is the common for all circuits from 1st to nth circuit. Hereafter, ian denotes nth electrolyte. in, and an denote cation and anion of ian, respectively. $[in]$, $[an]$ and $[ian]$ show the concentration (eq/m^3) of in, an and ian, respectively. The Faraday's law gives the relation between a quantity of electricity and an electric current. The Ohm's law gives the relation between an electric current and a resistance. These laws are given by Eq. (2), putting the quantity of electricity as Q (C):

$$Q(C) = I(A)\,t(s), \qquad I(A) = E(V)\ /\ R(\Omega) \tag{2}$$

Thus, the electric current I_n (A) carried by n^{th} electrolyte through n^{th} circuit is given by:

$$I_n = F[J_n]\,M_s = E\ /\ (R^K{}_{in} + R^A{}_{an} + R_{dilu.n}) = E\ /\ R_n \tag{3}$$

where F is the Faraday constant (sA/eq), $[J_n]$ the flux of nth electrolyte and M_s the membrane surface area (m^2).

In Eq.(3), $R_{dilu.n}$, the resistance of diluted compartment (Ω) due to nth electrolyte, is determined by the equivalent conductivity of nth electrolyte and is given by:

$$\begin{aligned} R_{dilu.n} &= \{1\ /\ (\lambda_{in}[in]_B + \lambda_{an}[an]_B)\}(l_{dilu}\ /M_s) \\ &= \{1\ /\ (\lambda_{in} + \lambda_{an})[ian]_B\}(l_{dilu}\ /M_s) \end{aligned} \tag{4}$$

where λ_{in} and λ_{an} denote the equivalent conductivity (S m^2/eq) of ion *in* and *an* respectively. l_{dilu} is the thickness of diluted compartment (m).

The membrane resistance is determined from the ion flux through the membrane. In ED, the ion flux through the membrane is mainly determined by the ion flux induced by the externally applied voltage [9]. Thus, assuming that k denotes all ionic species including cation i and anion a, the flux of ion k through the membrane $[J_k]$ is given by:

$$[J_k] = -z_k \alpha_k U_k [k]_M \left(\frac{\partial E_{AE}}{\partial x} \right) \tag{5}$$

where z_k denotes the ionic valence of ion k, (1 or –1), U_k the apparent mobility within the membrane (m^2/sV), $[k]_M$ the concentration (eq/m^3) within the membrane and $(\partial E_{AE}/\partial x)$ the externally applied electric potential gradient across the membrane (V/m). α_k is the parameter that shows the sieving effect of the membrane for ion k [10]. The concentration of ion partitioned into the membrane is obtained from the electroneutrality condition and the Donnan equilibrium condition. The concentration of ion k within the membrane is affected by the concentration at the both sides of membrane. However, in ED, the ion flux is mainly determined by the concentration of the diluted compartment. Thus, the concentration of cation and anion of nth electrolyte are given by Eq. (6), considering that the membrane charge density is extremely higher than the electrolyte concentration of diluted compartments C_B [11]:

$$\begin{aligned} [in]_M &= \theta^K{}_M\,[in]_B/C_B = \theta^K{}_M [ian]_B/C_B, \\ [an]_M &= \theta^A{}_M\,[an]_B/C_B = \theta^A{}_M\,[ian]_B/C_B \end{aligned} \tag{6}$$

where $\theta^K{}_M$ and $\theta^A{}_M$ denote the membrane charge density of CEM and AEM, respectively.

The flux is obtained by integrating Eq. (5) over the membrane thickness. The electric current carried by electrolyte is obtained from the flux by means of the Faraday's law. The membrane resistance, $R^K{}_{in}$ and $R^A{}_{an}$ are obtained from the electric current by means of the Ohm's law and are given by Eq. (7) from Eqs. (5) and (6):

$$\begin{aligned} R^K{}_{in} &= C_B l_M{}^K/\alpha_{in} U_{in} F M_s \theta^K{}_M\,[ian]_B, \\ R^A{}_{an} &= C_B l_M{}^A/\alpha_{an} U_{an} F M_s \theta^A{}_M\,[ian]_B \end{aligned} \tag{7}$$

where $l_M{}^K$ and $l_M{}^A$ indicates the membrane thickness (m) of CEM and AEM, respectively.

From Eqs. (1), (3), (4) and (7), $P^{am}{}_{an}$, the transport number ratio between *am* and *an*, is given by:

$$P^{am}_{an} = (a + bC_B) \,/\, (c + dC_B) \tag{8}$$

where

$$a = \{1/(\lambda_{in} + \lambda_{an})\}(l_{dilu}/M_S) \tag{9}$$

$$b = l_M{}^K/(\alpha_{in} U_{in} F M_s \theta^K{}_M) + l_M{}^A/(\alpha_{an} U_{an} F M_s \theta^A{}_M) \tag{10}$$

$$c = \{1 \,/\, (\lambda_{im} + \lambda_{am})\}(l_{dilu/}M_S) \tag{11}$$

$$\begin{aligned} d = {} & l_M{}^K \,/\, (\alpha_{im} U_{im} F M_s \theta^K{}_M) \\ & + l_M{}^A \,/\, (\alpha_{am} U_{am} F M_s \theta^A{}_M) \end{aligned} \tag{12}$$

In Eq. (8), a and c are the parameters determined by the equivalent conductivity of n^{th} and m^{th} electrolyte in the diluted compartment, respectively. b and d correspond to the sum of membrane resistance of CEM and AEM for n^{th} and m^{th} electrolyte, respectively.

It is found from Eq. (8) that the transport number ratio depends on the total electrolyte concentration of the diluted compartment C_B. If C_B is sufficiently high, $P^{am}{}_{an}$ is mainly determined by the membrane resistance b and d. On the contrary, if C_B is sufficiently low, $P^{am}{}_{an}$ is mainly determined by the equivalent conductivity of ions a and c. Thus, the transport number ratio changes with time, since the electrolyte concentration of the diluted compartment decreases with the progress of ED. The value of a and c are determined by the equivalent conductivity of ions. The values of b and d are determined from the analysis of the transport number ratio. After a, b, c and d are determined, it will be possible to simulate the change of ion fluxes as a function of time, that is the desalination during the ED, since all resistance values in the equivalent circuit shown in Fig. 2 are known.

3. Experimental

The ED experiment was done with the lab-scale ED system, Micro Acilyzer S3 (Astom Corp.). Fig. 3 shows the experimental set up. CMX (Astom Corp.) was used as CEM and AMX (Astom Corp.) as AEM. The ion exchange capacity of CMX was 2 ~ 3 meq/dry-g and that of AMX was 1.5 ~ 2.5 meq/dry-g. The membrane area of CMX and AMX was 5.5×10^{-3} m^2 each. This system contains 10 pairs of CMX and AMX. The gap between membranes was 7.5×10^{-4} m. Concentrated, diluted and electrode solutions were stocked in each reservoir and supplied to each compartment by circulating with pump. The volume of each reservoir was

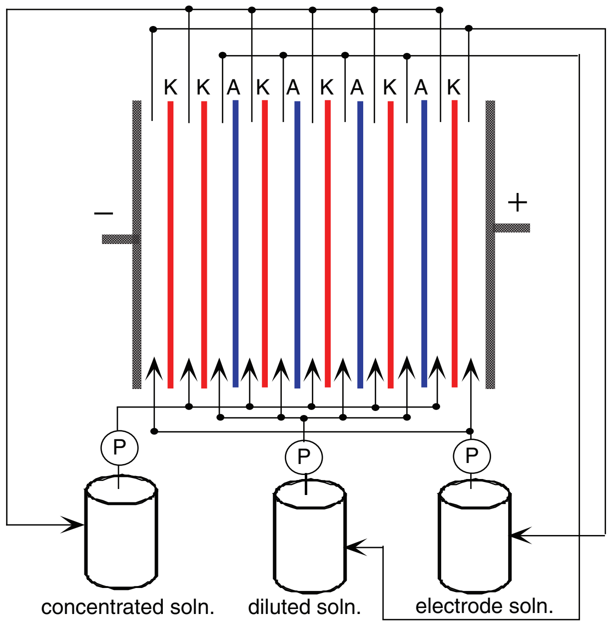

Fig. 3. Experimental set up K: CMX, A: AMX, P: pump.

500 ml and the flow rate of the solution was 1.2 l/min. 0.4% of $NaNO_3$ solution was used as an electrode solution. The current density was constant at 20 A/m^2 and the temperature was 25°C. NaF, $NaNO_3$ and NaCl were used as electrolytes. The initial concentrations of these electrolytes were changed in each experiment. The initial composition of the diluted solution and the concentrated solution was the same. The sample of 1 ml of diluted solution was collected from the reservoir every 5 min and was analyzed with an ion chromatograph apparatus (IA-300, DKK-TOA Corp.).

4. Results and discussion

4.1. Change of anion concentration of the diluted compartment during ED

Fig. 4(a) shows the change of total amount of anions deionized from the diluted compartment in three systems with different initial concentrations of NaF, $NaNO_3$ and NaCl. Fig. 4(b) shows the change of concentration of anions during ED. In Fig. 4(b), the initial NaF, $NaNO_3$ and NaCl concentrations were 22.6, 21.3 and 21.3 eq/m^3, respectively. As shown in Fig. 4(a), the total amounts of deionized anions are collinear and does not depend on the initial ion compositions. However, it is clear from Fig. 4(b) that the deionization rate depends on ionic species. The order of the deionization rate of AMX is NO_3^-> Cl^- >> F^-.

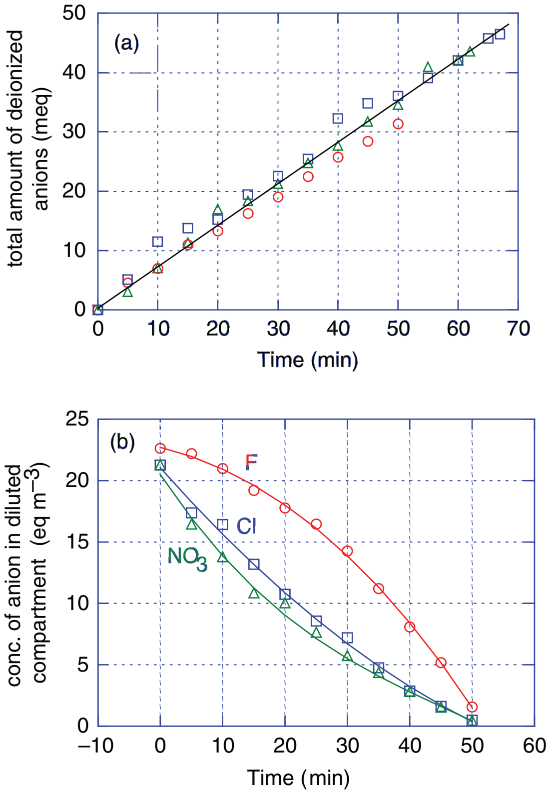

Fig.4. (a) Change of total amount of deionized anions during ED. Initial NaF, $NaNO_3$ and NaCl concentrations were□: 22.3, 22.7 and 50.4 eq/m^3, ○: 22.6, 21.3 and 21.3 eq/m^3, Δ: 23.3, 44.5 and 20.5 eq/m^3 (b) Change of concentration of anion in the diluted compartment during ED. Initial NaF, $NaNO_3$ and NaCl concentrations were 22.6, 21.3 and 21.3 eq/m^3, respectively. Symbols show the experimental data.

4.2. Transport number ratio

In order to discuss the permselectivity of anion exchange membrane, the transport number of F^- and NO_3^- relative to Cl^- were calculated. The ion flux was obtained from the gradient of ion concentration curve shown in Fig. 4(b). The transport number ratio was obtained by inserting the ion flux and ion concentration at certain time into Eq. (1). The gradient of ion concentration has some uncertainty at initial and final points of the experimental data. Thus, the transport number ratio obtained at initial and final experimental point is neglected.

Fig. 5(a) shows the transport number ratios P^{a}_{Cl} for several electrolyte compositions including the data shown in Fig. 4(b) as a function of time and Fig. 5(b) shows those as a function of the total electrolyte concentration of the diluted compartment. It is found from Fig. 5(a) that the transport number ratios are scattered depending on the initial electrolyte compositions.

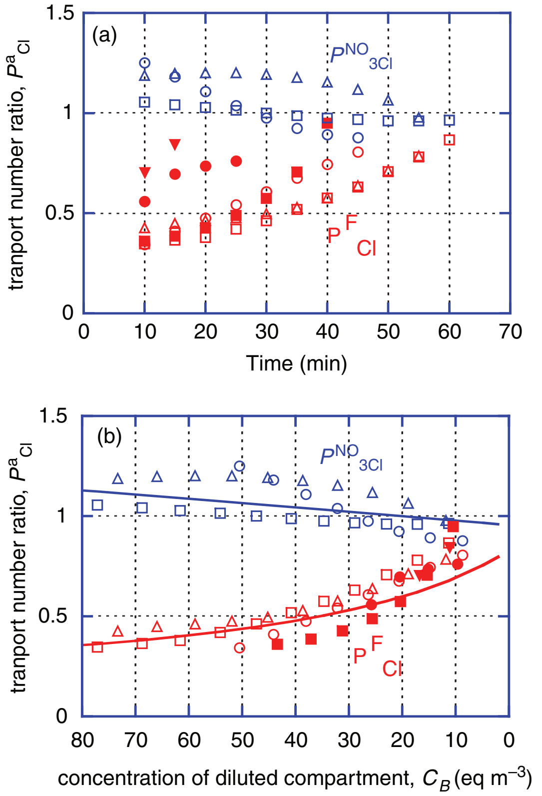

Fig. 5. Transport number ratio as a function of time (a) and as a function of the total electrolyte concentration of the diluted compartment C_B (b). Solid lines in Fig. 5(b) show the theoretical curves calculated with eq.(8). Initial NaF, $NaNO_3$ and NaCl concentrations were □:22.3, 22.7 and 50.4 eq/m^3, ○: 22.6, 21.3 and 21.3 eq/m^3, Δ: 23.3, 44.5 and 20.5 eq/m^3, ■: 18.4, 0 and 40.6 eq/m^3, ▼:19.7, 0 and 9.4 eq/m^3, •: 19.9, 0 and 18.8 eq/m^3.

On the contrary, the data shown in Fig. 5(b) approximately lie on the lines. This means that the transport number ratio depends on the total electrolyte concentration of the diluted compartment. This is in agreement with the prediction by Eq. (8).

The data shown in Fig. 5(b) were analyzed with Eq.(8). In Eq.(8), a and c are the parameters determined by both the equivalent conductivity of electrolyte in the diluted compartment and ED system as shown in Eqs. (9) and (11), respectively. Thus, a and c were obtained by inserting the equivalent conductivity of electrolyte, the membrane area (5.5×10^{-3} m^2) and the gap between membranes (7.5×10^{-4} m) into Eqs. (9) and (11), respectively. The equivalent conductivity of Na^+, F^-, NO_3^- and Cl^- are 50.1×10^{-4}, 55.4×10^{-4}, 71.4×10^{-4} and 76.3×10^{-4} Sm^2/eq, respectively. b and d are the parameters that show the membrane resistance for ion transport through the ion exchange membranes and were determined from the best fit of the theoretical curve with the experimental data. Solid lines in Fig. 5(b) show the theoretical curves calculated with Eq. (8). It is clear from Fig. 5(b) that the theoretical curves fit with the experimental data well. The value of parameters used to calculate the theoretical curves are shown in Table 1. From Eq. (8), the transport number ratio is mainly determined by b/d in the system where the total electrolyte concentration of the diluted compartment C_B is sufficiently high. On the contrary, in the system where C_B is sufficiently low, the transport number ratio is mainly determined by a/c. Thus, the transport number ratio changes with the progress of ED, since C_B decreases with time.

From b and d in Table 1, the sum of membrane resistance of AMX and CMX for ion transport are 0.03 Ω for NaCl, 0.3 Ω for NaF and 0.005 Ω for $NaNO_3$. Na^+ is common as a cation for all electrolytes. Thus, from these resistances, it is clear that the apparent mobility of F^- in AMX is lower than Cl^- and NO_3^-. This low apparent mobility of F^- is the reason why the desalinating rate of F^- is lower than other anions.

It is reported that the transport number ratio of anions with same ionic valence does not depend on the Stokes radius, but depends on the Gibbs hydration energy [8]. This indicates that dehydrated anions can permeate the anion exchange membrane. Table 2 shows the Gibbs hydration energies and the Stokes radii of F^-, Cl^- and NO_3^- [8]. The Gibbs hydration energy of F^- is highest in these three anions. F^- needs higher energy to dehydrate than Cl^- and NO_3^-. This condition makes F^- hard to permeate AMX. The high membrane resistance of F^- reflects this situation. On the other hand, the Gibbs hydration energy of NO_3^- is lowest. NO_3^- is easily dehydrated and permeates AMX easier compared with Cl^- and F^-.

Table 1
Values of parameters obtained from the analysis of Fig. 5(b)

	a/Ω eq m^{-3}	b/Ω	c/Ω eq m^{-3}	d/Ω
P^F_{Cl}	10.7	0.03	12.9	0.3
$P^{NO_3}_{Cl}$	10.7	0.03	11.2	0.005

Table 2
Gibbs hydration energy, $-\Delta G_h^0$ and Stokes radius

Anion	$-\Delta G_h^0$(kJ/mol)	Stokes radius (pm)
F^-	434	166
Cl^-	317	121
NO_3^-	270	129

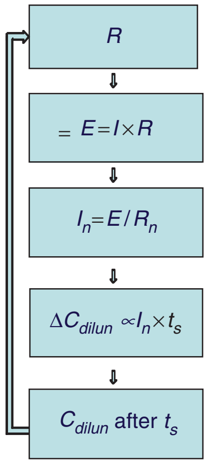

Fig. 6. Flow chart of the simulation of ED with constant current.

4.3. Simulation

The values of resistances in the equivalent circuit shown in Fig. 2 are calculated with Eqs. (4) and (7) using the values of *a*, *b*, *c* and *d* shown in Table 1. This makes possible to calculate the electric current that flows through each circuit connected parallel, which is the desalinated amount of each electrolyte.

Fig. 6 shows the flow chart of simulation of ED in the constant current system. The initial concentration of electrolytes is known from the experimental condition. At first, as shown in Fig. 6, the total electric resistance of circuit R (Ω) is obtained from the sum of the inverse of resistance of each circuit. The voltage applied to circuit E (V) is calculated by means of the Ohm's law, since the current that flows through the system is known. This voltage is common for every circuits connected parallel. Thus, the electric current that flows through each circuit is calculated, since applied voltage and resistance are known. The desalinated amount of electrolyte is calculated by means of the Faraday' law assuming the electrolyte concentration is constant during a short time t_s. The electrolyte concentration after time t_s is obtained by subtracting the desalinated amount from the initial electrolyte concentration. The simulation is done by repeating the same procedure, using the electrolyte concentration after time t_s as a new initial concentration.

The effective current is needed to simulate ED, since a part of electric current is not used to deionize. The effective electric current was obtained as 0.11 A from Fig. 4(a) by means of the Faraday's law. The simulation results and experimental data are shown in Fig. 7. It is found that the simulation results fit with the experimental data very well. Thus, the simulation of ED with the equivalent circuit was successfully performed.

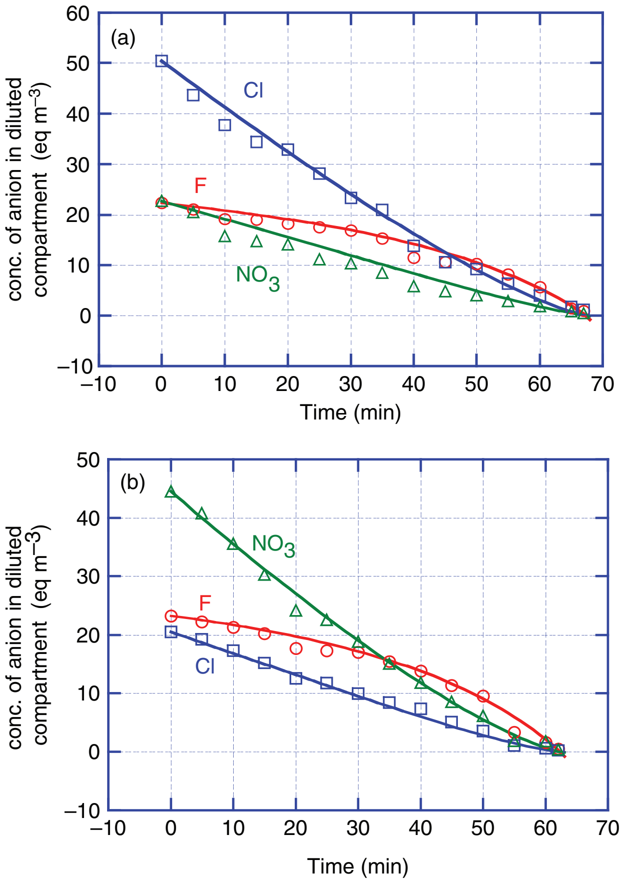

Fig. 7. Simulation of the ED system. Solid lines show the simulation results and symbols the experimental data. Effective electric current is 0.11 A. (a): Initial NaF, $NaNO_3$ and NaCl concentrations were 22.3, 22.7 and 50.4 eq/m^3, respectively. (b): Initial NaF, $NaNO_3$ and NaCl concentrations were 22.3, 44.5 and 20.5 eq/m^3, respectively.

5. Conclusion

In ED, the desalination rate depends on ionic species, since ion exchange membranes have permselectivity. In order to discuss the anion permselectivity, the transport number ratio between anion and Cl^- ion P^a_{Cl} was measured. It was shown that P^a_{Cl} changed with the progress of ED. The equivalent circuit of ED was proposed to discuss the mechanism of permselectivity. According to this model, the transport number ratio depends on the total electrolyte concentration of the diluted compartment C_B. P^a_{Cl} is determined by the membrane resistances of ions transport in the system where C_B is sufficiently high. On the other hand, in the system where C_B is sufficiently low, P^a_{Cl} is determined by the equivalent conductance of electrolytes in the diluted compartment. Thus, P^a_{Cl} changes with the progress of ED, since the electrolyte concentration of the diluted compartment decreases with time. The transport number ratio was analyzed with this model and it was shown that the model explained the experimental results very well.

The simulation of ED with the equivalent circuit was successfully performed using the electric resistances of the equivalent circuit obtained from the analysis of P^a_{Cl}. The simulation results were in good agreement with ED data.

Acknowledgement

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Symbols

C_B — total electrolyte concentration of the diluted compartment (eq/m^3)
I_n — electric current carried by nth electrolyte (A)
$[J_k]$ — flux of ion k (eq/m^{2}s)
M_S — membrane surface area (m^2),
P^k_{Cl} — transport number ratio between ion k and Cl^-
R^K_i, R^A_a, R_{dilu} — electric resistance of cation exchange membrane for cation, i, of anion exchange membrane for anion, a and of diluted compartment, respectively (Ω)
U_k — apparent mobility of ion k within the membrane (m^2/sV)
a, b, c, d — defined by Eqs. (9)~(12), respectively
in, an, ian — nth cation, anion and electrolyte
$[k]$ — concentration of ion k (eq/m^3)
l_{dilu} — thickness of diluted compartment (m)
l_M — membrane thickness (m)
t_k — transport number of ion k, $[J_k]/\Sigma[J_s]$ (-)
α_k — sieving effect of membrane for ion k
λ_k — equivalent conductivity of ion k (S m^2/eq)
θ_M — membrane charge density (eq/m^3)

Subscripts, superscripts

B, M — bulk solution and membrane, respectively
K, A — cation exchange membrane and anion exchange membrane, respectively
i, a, k — cation, anion and ion k, respectively
n, m — nth and mth electrolyte, respectively

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