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K. Kontturi[a]
[a] DEPARTMENT OF CHEMISTRY HELSINKI, UNIVERSITY OF TECHNOLOGY, ESPOO, FINLAND

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Trace Ions in Countercurrent Electrolysis in a Thin, Porous Membrane

K. KONTTURI

DEPARTMENT OF CHEMISTRY
HELSINKI UNIVERSITY OF TECHNOLOGY
SF-02150 ESPOO, FINLAND

Abstract

The transport of trace ions added to a binary electrolyte system through a porous membrane during countercurrent electrolysis has been studied both theoretically and experimentally. Theoretical models based both on the general transport equations and on the Nernst-Planck equations are presented. Experiments and calculations for the binary system NaCl-H_2O with trace ions Li^+ and K^+ were performed. The theoretical model was able to predict the transport phenomena fairly well.

INTRODUCTION

It has been shown that countercurrent electrolysis in a thin porous membrane can be used to separate ions of different mobilities both in systems consisting of strong electrolytes (*1*-*3*) and of weak electrolytes (*4*). In this paper a special case, a binary system with trace ions, is studied. A trace ion is defined as an ion which is added to a system in such small amounts that its effect on the main system—here the binary one—is negligible. Evidently the main system has a great effect on the trace ion.

From a practical point of view, the concept of trace-ion transport is important: consider a separation process that has continued for so long that very high separation efficiency has been reached. Then the ion making the solution impure is a typical example of a trace ion. The case is the same if we start to separate some valuable ion from the solution in

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which it is present in only small amounts. It is clear that if the mathematical solution is easier in the case of a trace ion than in the case of a real ternary system (*1, 3*) it is very reasonable to use the trace-ion approach. We will show that this is really the case, and mathematical modeling can be simplified considerably by using the concept of a trace ion. By modeling the transport of a trace ion in countercurrent electrolysis in a thin porous membrane, it will be shown that the closed form solution of the transport problem can be obtained. Although the solution is not obtainable in the form of elementary functions, it is a step forward for a simpler interpretation of the transport process.

It can be easily imagined that the solution of transport of a trace ion can be used as a first approximation when dealing with cases where the concentration of one ion is low but not so low that the trace-ion assumption (i.e., not effecting the main system) is strictly valid. In the present work the behavior of trace-ions in countercurrent electrolysis will be studied in detail. This approach is also important in the study of mobilities and charge density distributions in polyelectrolyte solution provided that polyelectrolytes are added to a solution containing some salt which forms the "main" binary system.

TRANSPORT EQUATIONS FOR TRACE IONS

The transport of ions in porous membrane in multicomponent system can be represented by

$$J_i = J_i^{DIFF} + J_i^{MIGR} + J_i^{CON} \tag{1}$$

where

$$J_i = c_i v_i \tag{1a}$$

$$J_i^{DIFF} = -\sum_{l=1} \nu_{il} \sum_{j=1} D_{ij} \frac{dc_j}{dx} \tag{1b}$$

$$J_i^{MIGR} = t_i \frac{I}{z_i F} \tag{1c}$$

$$J_i^{CON} = c_i v \tag{1d}$$

The ionic flow is J_i, consisting of the diffusional flow J_i^{DIFF}, the migration flow J_i^{MIGR}, and the convective flow J_i^{CON}. Furthermore, t_i is the transport number, z_i is the charge number, c_i is the concentration of ion i, c_j is the concentration and D_{ij} are the diffusion coefficients of component j, ν_{il} is the stoichiometric coefficient of ion i, F is the Faraday constant, I is the electric current density, and v is the velocity of solvent flow through the membrane (here referred to as convection). Quantities v and v_i, and thus J_i and J_i^{CON}, are expressed with reference to the membrane.

We begin by considering such an aqueous quaternary system which consists of three cations with a common anion. This kind of system is general enough to give information on coupling effects between the main binary system and the trace ions as well as between the different trace ions themselves.

The lack of measured data for transport quantities means that we have to estimate the concentration dependences of these quantities. A simple and efficient way to estimate diffusion coefficients and transport numbers in multicomponent systems is offered by the Nernst-Planck equations (*5*). Thus we can estimate the diffusion coefficients from

$$D_{ij} = \frac{RTt_i}{z_i^2 F^2}\left[\sigma_{ij}\frac{\lambda_i}{t_i} - \left(\frac{z_i\lambda_j}{z_j} - \frac{z_i z_j}{z_4^2}\lambda_4\right)\right]; \qquad i, j = 1, 2, 3 \tag{2}$$

and the transport numbers

$$t_i = \frac{c_i\lambda_i}{\sum_{k=1}^{4} c_k\lambda_k}; \qquad i = 1, 2, 3, 4 \tag{3}$$

where σ_{ij} is Kronecker's delta ($\sigma_{ij} = 1$ when $i \neq j$; $\sigma_{ij} = 0$ when $i \neq j$), and λ_i is the molar conductivity of ion i. Subscripts 1, 2, and 3 denote the cations and subscript 4 denotes the common anion. Furthermore, the subscript 1 denotes the cation of the main binary system and subscripts 2 and 3 denote the trace ions.

The solution of the transport equations is simplified by using the trace-ion assumptions. Since the trace ions have been added to the main binary system in small amounts, it follows that

$$t_2, t_3 \ll t_1 \text{ since } c_2, c_3 \ll c_1 \tag{4}$$

Assumption (4) is not sufficient to guarantee that the trace ions do not have an effect on the main binary system. This is achieved by assuming that the trace ions cannot maintain steep concentration gradients, i.e.,

$$\left|\frac{dc_2}{dx}\right|, \left|\frac{dc_3}{dx}\right| \ll \left|\frac{dc_1}{dx}\right| \text{ when } \left|\frac{dc_1}{dx}\right| \gg 0 \tag{5a}$$

and

$$\frac{dc_2}{dx} \approx \frac{dc_3}{dx} \approx 0 \text{ when } \frac{dc_1}{dx} \simeq 0 \tag{5b}$$

Using these assumptions, the transport equations for the cations take the forms

$$J_1 = -\nu_1 D_{11} \frac{dc_1}{dx} + t_1 \frac{I}{z_1 F} + c_1 v \tag{6}$$

$$J_2 = -\nu_2 D_{21} \frac{dc_1}{dx} - \nu_2 D_{22} \frac{dc_2}{dx} + t_2 \frac{F}{z_2 F} + c_2 v \tag{7}$$

$$J_3 = -\nu_3 D_{31} \frac{dc_1}{dx} - \nu_3 D_{33} \frac{dc_3}{dx} + t_3 \frac{I}{z_3 F} + c_3 v \tag{8}$$

where the transport quantities are

$$t_1 = \frac{\lambda_1}{\lambda_1 + \dfrac{z_1}{|z_4|} \lambda_4} \tag{9a}$$

$$t_2 = \frac{c_2}{c_1} \frac{\lambda_2}{\lambda_1 + \dfrac{z_1}{|z_4|} \lambda_4} \tag{9b}$$

$$t_3 = \frac{c_3}{c_1} \frac{\lambda_3}{\lambda_1 + \dfrac{z_1}{|z_4|} \lambda_4} \tag{9c}$$

$$D_{11} = \frac{\lambda_1 RT}{z_1^2 F^2} - \frac{RTt_1}{z_1^2 F^2}\left(\lambda_1 - \frac{z_1^2 \lambda_4}{z_4^2}\right) \tag{10a}$$

$$D_{22} = \frac{\lambda_2 RT}{z_2^2 F^2} \tag{10b}$$

$$D_{33} = \frac{\lambda_3 RT}{z_3^2 F^2} \tag{10c}$$

$$D_{21} = -\frac{RTt_2}{z_2^2 F^2}\left(\frac{z_2}{z_1}\lambda_1 - \frac{z_2 z_1}{z_4^2}\lambda_4\right) \tag{10d}$$

$$D_{31} = -\frac{RTt_3}{z_3^2 F^2}\left(\frac{z_3}{z_1}\lambda_1 - \frac{z_3 z_1}{z_4^2}\lambda_4\right) \tag{10e}$$

The following conclusions can be drawn from transport Eqs. (6)–(8) together with Eqs. (9) and (10): The main binary system has the form of a pure binary system and the trace ions do not have effect on it. The binary system couples strongly with the diffusion of the trace ions in the case of steep concentration profiles of the binary system. These coupling effects vanish when the binary system is homogeneous or when the diffusion potential approaches zero, i.e., D_{21} and/or $D_{31} \simeq 0$ since $(z_2\lambda_1/z_1) - (z_2 z_1\lambda_4/z_4^2) \simeq 0$ and/or $(z_3\lambda_1/z_1) - (z_3 z_1\lambda_4/z_4^2) \simeq 0$. The transport of a trace ion is independent of the other trace ions present in the system.

Equations (6)–(10) can now be solved if, e.g., the boundary concentrations, convection, and electric current density are given. In order to solve this system we first solve the binary case, i.e., Eq. (6), and then use this solution to solve Eqs. (7) and (8). All the equations are linear first-order differential equations that are easy to solve.

An important fact can be found by comparing Eqs. (6)–(10) with the Nernst-Planck equations. If we write the Nernst-Planck equation for every ion and first solve the binary case without taking the trace ions into account, the solution for the concentration profiles and for the electric potential distribution $\varphi = \varphi(x)$ is obtained. By substituting this potential distribution into the Nernst-Planck equations of the trace ions and solving the resulting equations, the same solutions obtained for the trace-ion case using Eqs. (6)–(10)* results. Thus it is concluded that the electric potential in the Nernst-Planck equation takes into account the coupling effects. Furthermore, this is an easy way to consider the behavior of the trace ions, since it is only necessary to solve the main system to obtain $\varphi = \varphi(x)$ and thus obtain a set of independent differential equations for the trace ions. The situation resembles the case of Goldman's constant field approximation (*6*) except that the above procedure is not an approximation, it is as exact as the Nernst-Planck equations.

*Note that in Eqs. (6)–(10) we have used Nernst-Plack equations to get approximations for the transport quantities D_{ij} and t_i; they are not Nernst-Planck equations.

TRACE IONS IN COUNTERCURRENT ELECTROLYSIS

Let us consider a cell as schematically represented in Fig. 1. We set the problem as follows: Into Compartment β we put the mixture of three univalent salts in water, i.e., $z_1 = z_2 = z_3 = -z_4 = 1$. The concentration of one of these salts is approximately one thousand times greater than the concentrations of the two other salts. When the stationary state has been reached, the fluxes of each ion through the porous membrane are given by

$$J_i = -C_i^\alpha \dot{V}^\alpha / A \tag{11}$$

It is convenient to use the following dimensionless quantities:

$$y_i(X) = C_i(X)/C_0$$

$$I_r = Il/FC_0D_0$$

$$v^c = \dot{V}^c l/AD_0$$

$$v^\alpha = \dot{V}^\alpha l/AD_0$$

$$X = x/l$$

$$d_{ij} = D_{ij}/D_0$$

where C_0 is the scaling concentration, l is the thickness of the membrane, A is its surface area, A/l is the membrane constant, and D_0 is the scaling diffusion coefficient. Using these notations, the solution of Eq. (6) can be presented in the form

$$y_i(X) = \left[y_i^\alpha + \frac{t_1 I_r}{v^c} + \frac{y_1^\alpha v^\alpha}{v^c}\right] \exp\left(\frac{v^c X}{d_{11}}\right) - \left(\frac{t_1 I_r}{v^c} + \frac{y_1^\alpha v^\alpha}{v^c}\right) \tag{12}$$

By substituting Eq. (12) into the Eqs. (7) and (8), solutions for the trace ions are obtained:

$$\frac{y_2^\beta}{y_2^\alpha} = \left\{(y_1^\alpha)^{-B_1} + \frac{v^\alpha}{d_{22}} \int_0^1 \exp(T_2 X)[y_1^\alpha + B) \exp(aX) - B]^{-B_1} dx\right\} \Big/ \exp(T_2)[(y_1^\alpha + B) \exp(a) - B]^{-B_1} \tag{13}$$

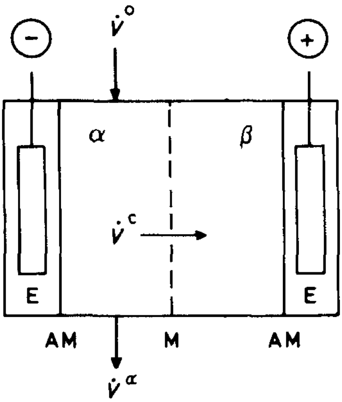

FIG. 1. Schematic drawing of the cell. A thin, porous membrane (M) divides the cell into two compartments α and β. The volume of Compartment β is so large compared to the volume of Compartment α that the concentrations in Compartment β remain constant during the measurements. The solutions in Compartments α and β are well mixed. ⊖ is the cathode and ⊕ is the anode, and they are separated from Compartments α and β by anion exchange membranes (AM). Water is pumped at a constant rate ($\dot{V}^0$) to Compartment α. Part of this water stream flows out of Compartment α as the outflow stream $\dot{V}^\alpha$ while the rest flows by convection through the porous membrane ($\dot{V}^c = \dot{V}^0 - \dot{V}^\alpha$).

$$\frac{y_3^\beta}{y_3^\alpha} = \left\{ (y_1^\alpha)^{-B_1} + \frac{v^\alpha}{d_{33}} \int_0^1 \exp(T_3 X)[(y_1^\alpha + B)\exp(aX) - B]^{-B_1} dx \right\} \Big/ \exp(T_3)[(y_1^\alpha + B)\exp(a) - B]^{-B_1} \quad (14)$$

where

$$a = \frac{v^c}{d_{11}}, \qquad B = \frac{t_1 I_r}{v^c} + \frac{y_1^\alpha v^\alpha}{v^c}$$

$$B_1 = \frac{\lambda_1 - \lambda_4}{\lambda_1 + \lambda_4} + \frac{I_r D_0}{aB(D_1 + D_4)} \qquad \left(D_1 = \frac{\lambda_1 RT}{F^2};\ D_4 = \frac{\lambda_4 RT}{F^2} \right)$$

$$T_2 = \frac{I_r D_0}{(D_1 + D_4)B} - \frac{v^c}{d_{22}}, \qquad T_3 = \frac{I_r D_0}{(D_1 + D_4)B} - \frac{v^c}{d_{33}}$$

As can be seen from Eqs. (13) and (14), the integrals cannot be presented using elementary functions. However, this is not a problem since these integrals are easy to evaluate by numerical integration routines.

TABLE 1. Experimental Results in the System

v^c (expt)	$-I_r$ (expt)	$-I_r$ (theoret)	$Y^\alpha_{Na^+}$ (expt)	$Y^\alpha_{K^+} \times 10^3$ (expt)	$Y^\alpha_{K^+} \times 10^3$ (theoret)	$Y^\alpha_{Li^+} \times 10^3$ (expt)
1.76	18.65	18.78	1.22	0.62	0.52	0.91
3.39	24.87	27.87	1.31	0.76	0.65	0.87
4.84	33.16	31.42	1.28	0.84	0.71	0.70
6.64	36.27	35.27	1.21	0.92	0.76	0.51
9.90	39.38	35.90	0.96	0.93	0.75	0.22
1.76	37.31	35.07	2.16	0.83	0.87	1.74
3.39	49.74	47.25	2.26	1.11	1.02	1.53
4.84	66.32	49.74	2.62	1.16	1.07	1.21
6.64	72.54	65.08	2.23	1.51	1.22	1.10
9.90	78.76	69.22	1.85	1.65	1.23	0.60
1.76	78.76	68.39	4.11	1.79	1.55	3.39
3.39	99.48	89.12	4.26	1.98	1.75	3.10
4.84	132.6	165.7	4.30	2.20	1.92	2.75
6.64	145.1	120.2	4.12	2.70	2.00	2.27
9.90	157.5	131.8	3.43	2.98	2.07	1.37

[a] The measured concentrations and fluxes (expt) of sodium, potassium, and lithium ions are presented as a function of electric current and convection. The theoretical values (theoret) for electric current and concentrations of potassium and lithium are calculated by using the theoretical model described in the present paper. In the theoretical calculations, convection and the concentration of

Consider a few special cases:

1) When the outflow stream $v^\alpha = 0$, the selectivity ratio (3) for trace ions takes a simple form

$$S = \frac{y_2^\beta \, y_3^\beta}{y_2^\alpha \, y_3^\alpha} = \exp\left[\left(\frac{1}{d_{22}} - \frac{1}{d_{33}}\right)v^c\right] \tag{15}$$

Equation (15) is the same as previously obtained in the ternary case when the cation fluxes were zero (*1*). Note that the exponential dependence of the selectivity ratio on convection is also valid for trace ions.

2) The diffusion potential is equal to zero, i.e., $\lambda_1 = \lambda_4$. In this case the coupling effects of diffusion vanish since $D_{21} = D_{31} = 0$. The binary system influences only the transport numbers of the trace ions (see Eqs. 9b and 9c) through the conductance, which is proportional to $\lambda_1 + \lambda_4$. The solution of the problem does not become easier in practice.

$NaCl-H_2O$ with Trace Ions K^+ and Li^+ [a]

$Y^{\alpha}_{Li^+} \times 10^3$ (theoret)	$-K_{Na^+}$ (expt)	$-K_{K^+} \times 10^3$ (expt)	$-K_{Li^+} \times 10^3$ (expt)	S_{KNa} (expt)	S_{NaLi} (expt)	S_{KLi} (expt)
0.83	5.99	3.04	4.42	2.91	1.54	3.40
0.75	6.43	3.73	4.28	2.52	1.73	4.36
0.61	6.29	4.12	3.43	2.85	2.10	5.99
0.47	5.94	4.52	2.31	3.31	2.73	9.04
0.23	4.71	4.57	1.08	4.21	5.02	21.1
1.59	10.6	4.08	8.54	1.67	1.43	2.39
1.44	11.1	5.46	7.51	2.14	1.70	3.64
1.08	9.92	5.70	5.94	2.50	1.92	4.80
1.02	11.0	7.41	5.40	2.95	2.33	6.87
0.54	9.09	8.10	2.94	3.88	3.54	13.7
3.25	20.2	8.79	16.7	1.89	1.39	2.63
3.61	20.9	9.72	15.2	2.02	1.58	3.19
2.70	21.1	10.8	13.6	2.23	1.80	4.01
2.18	20.2	13.2	11.0	2.85	2.11	6.01
1.25	17.3	14.7	6.73	3.67	2.96	10.9

sodium in Compartments α and β are taken to be exact. Dimensionless values for the parameters are calculated from the experimental and theoretical ones by taking $A/l = 12.5$ cm, $c_0 = 0.01$ mol/dm^3, $D_0 = 2 \times 10^{-5}$ cm^2/s, $v^c = \dot{V}^c l/AD_0$, $I_r = Il/c_0FD_0$, $K_i = J_i l/c_0D_0$, and $y_i = c_i/c_0$. Concentrations in Compartment β are $y^{\beta}_{Na^+} = 4.35$, $y^{\beta}_{K^+} = 1 \times 10^{-3}$, and $y^{\beta}_{Li^+} = 5 \times 10^{-3}$.

3) The binary system is homogeneous, i.e., $dc_1/dx = 0$. This resembles the case where the diffusion potential is equal to zero. The solution of the problem is greatly simplified, and the concentration ratio for trace-ion 2 is

$$\frac{y_2^{\beta}}{y_2^{\alpha}} = \exp(-T_2)\left[1 + \frac{v^{\alpha}}{d_{22}T_2}(\exp(T_2) - 1)\right] \tag{16}$$

The concentration ratio for trace-ion 3 can be obtained by changing the subscripts 2 to 3 in Eq. (16). Furthermore, if there is no outflow from Compartment α, i.e., $v^{\alpha} = 0$, the separation ratio for the trace ions assumes the form given by Eq. (15).

4) When the diffusion potential is zero ($\lambda_1 = \lambda_4$) and the electric current density $I_r = 0$, equations identical to (15) and (16) are obtained. However, remember that the concentration levels differ greatly for the case where $I_r \neq 0$.

EXPERIMENTAL

The system studied had $NaCl-H_2O$ as the binary system and Li^+ and K^+ as the trace ions of the corresponding chlorides. The apparatus used is schematically described in Fig. 1. A detailed description of the experimental set-up has been presented previously (*3*). For every measurement the total concentration in Compartment β was kept constant ($\sim 4.35 \times 10^{-2}$ mol/dm^3) and the concentrations of the different ions in this compartment were kept constant throughout the experiment: $C^{\beta}_{Na^+} = 4.35 \times 10^{-2}$ mol/dm^3, $C^{\beta}_{K^+} = 1 \times 10^{-5}$ mol/dm^3, and $C^{\beta}_{Li^+} = 5 \times 10^{-5}$ mol/dm^3. The concentrations in Compartment α were determined by analyzing the concentrations in the outflow stream $\dot{V}^{\alpha}$. The system was considered to have reached the stationary state when the concentrations in the outflow stream $\dot{V}^{\alpha}$ remained unchanged.

The measurements were carried out by changing both the convection and the electric current density. In practice, we chose five different convective regimes. With the aid of the mathematical model presented here, suitable electric current densities were calculated to obtain boundary concentrations $C^{\alpha}_{Na^+} = 1 \times 10^{-2}$, 2×10^{-2}, and 4.35×10^{-2} mol/dm^3 for sodium and for each of five convection values. The concentrations of Li^+, K^+, and Na^+ ions were determined by AAS.

RESULTS

The results of the measurements in the binary system $NaCl-H_2O$ with trace ions Li^+ and K^+ with varying electric current density and convection are reported in Table 1. The theoretical results are reported in the same table, and there is satisfactory agreement between theory and experiment. For higher current densities the predictions of the model deviate from the results obtained. Evidently this is due to the leakage of ions through the ion-exchange membrane.

Figure 2 shows the logarithm of the selectivity ratio as a function of convection for different cation pairs. Again, an exponential dependence of the selectivity ratio on convection is clearly evident. It is also clearly seen that the stronger the coupling of the binary system (i.e., the greater the concentration gradient of the binary system), the better is the separation.

From the results we can conclude that trace-ion assumptions (4) and (5) are justified when the concentrations of the trace ions are about one thousand times smaller than the concentration of the salt of the binary system. Of course, the validity of the trace-ion assumptions can be

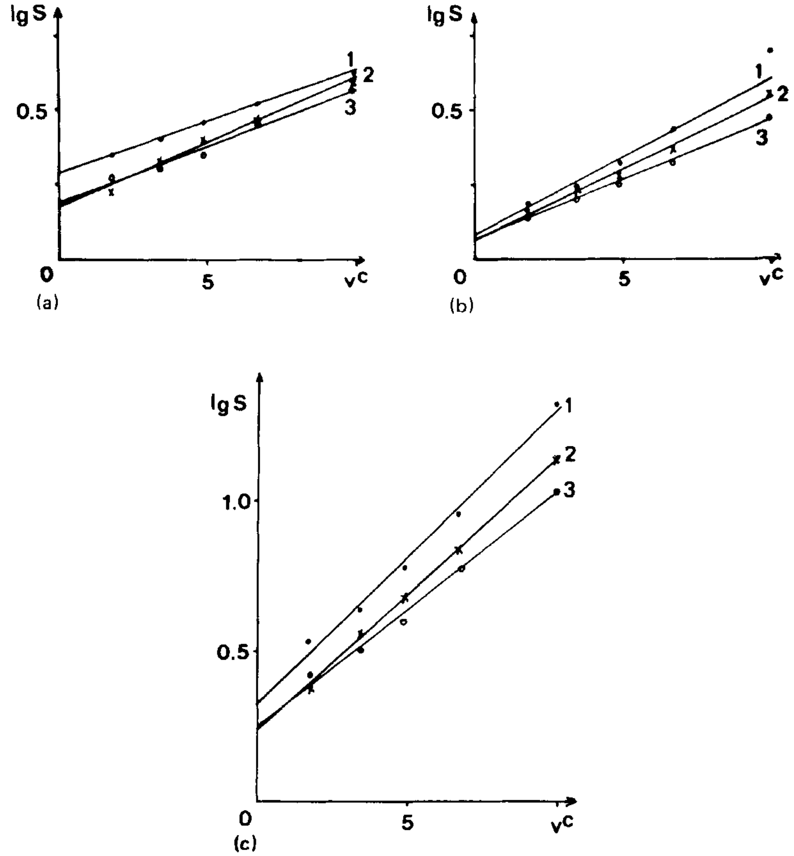

FIG. 2. The experimentally obtained relationships between the logarithm of the selectivity ratio (S) and the convection (v^c) in the system NaCl–H_2O with the trace ions Li^+ and K^+. In Figs. (a), (b), and (c) the Curves 1, 2, and 3 denote $y^{\alpha}_{Na^+} \simeq 1, 2$, and 4.35, respectively.

$$\text{(a)}\ \lg S_{KNa} = f(V^c);\ S_{KNa} = \frac{y^{\alpha}_{K^+}}{y^{\beta}_{K^+}} \cdot \frac{y^{\beta}_{Na^+}}{y^{\alpha}_{Na^+}}$$

$$\text{(b)}\ \lg S_{NaLi} = f(v^c);\ S_{NaLi} = \frac{y^{\alpha}_{Na^+}}{y^{\beta}_{Na^+}} \cdot \frac{y^{\beta}_{Li^+}}{y^{\alpha}_{Li^+}}$$

$$\text{(c)}\ \lg S_{KLi} = f(v^c);\ S_{KLi} = \frac{y^{\alpha}_{K^+}}{y^{\beta}_{K^+}} \cdot \frac{y^{\beta}_{Li}}{y^{\alpha}_{Li}}$$

estimated with the aid of theoretical models for ternary and quaternary systems. However, the validity of the trace-ion approach depends greatly on the particular case studied. From this work it is clear that the trace-ion assumptions appear to be valid when the concentrations of the trace ions are $\leqslant 5 \times 10^{-3}\, y_{BINARY}$ (y_{BINARY} is the concentration of the salt of the binary system).

CONCLUSIONS

In countercurrent electrolysis in a thin porous membrane, the behavior of trace ions added to the binary system can be modeled to obtain closed form solutions. This model predicts the transport behavior of trace ions fairly well. The predictions of the theoretical model and the experimental results verify that the binary system strongly couples the diffusion of trace ions when the binary system is nonhomogeneous. An increase of those coupling effects increases the separation efficiency.

The main merit of the trace-ion concept is its suitability to model the separation process based on countercurrent electrolysis when the impurity ions are present only in small amounts. The reason for this is that mathematical modeling of the separation process becomes significantly easier than for real ternary or quaternary systems.

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