

Preferential transport behaviors of ternary system cupric–nickel–zinc ions through cation-exchange membrane with a complexing agent by dialysis

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Abstract

Preferential transport behaviors of ternary metal ion system were studied in a stirred batch cation-exchange membrane dialyzer. Cu^{2+} – Ni^{2+} – Zn^{2+} system was chosen for investigation. Malonic acid, oxalic acid and citric acid were employed, respectively, as a complexing agent in order to enhance the discrepancy in transport fluxes of metal ions. The results show that the effective order of complexing agents is malonic acid > oxalic acid > citric acid. The stoichiometric ratio of complexing agent to metal ions and the pH value of the feed solution are the primary factors on the preferential transport behaviors of metal ions. The optimal dimensionless permeation flux ratio of $P_{\text{Zn-Na}}/P_{\text{Ni-Na}}/P_{\text{Cu-Na}}$ is obtained about 5.3/3.6/0.7. On the basis of the Nernst–Planck equation and interface equilibrium, a theoretical model of the system is established. The predicted results obtained by theoretical model are in good agreement with the experimental results.

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1. Introduction

The clean technology using ion-exchange membranes has been taken into great account in chemical industry [1]. Ion-exchange membrane dialysis is a useful process for ion separation, in which ions move across membrane based on the Donnan membrane equilibrium principle [2]. Among various techniques that have been used for ion separation and concentration, Donnan dialysis is an efficient method in pretreatment stage in industrial applications because of its permselective transport within low ionic concentration [3]. Many investigations of ion transfer across the ion-exchange membrane have been reported [4,5]. One of the prominent features in the development of modern technology is the cross-fertilization of ideas among different disciplines. Therefore, a separation process combined cation-exchange membrane with complexing agents for the recovery and enrichment of heavy metal ions has received much attention in recent years [6,7].

Preferential transport behaviors of binary metal ion system using membrane dialysis in the presence of complexing

agents had been studied in our previous researches [8–11]. The experimental results show that the uptake of metal ions in the cation-exchange membrane can be changed by adding a complexing agent in the solution phase, and both the anion ligands and the kinds of metal ions can differentiate the equilibrium uptake of metal ions so as to increase the membrane selectivity of the metal ions. These results are in accordance with the reports of other investigators [12–14]. Based on these results, the combined use of a cation-exchange membrane and a complexing agent seem to be a feasible method for the simultaneous separation and concentration of metal ions [15,16]. However, it is seemed to need more experimental work on multi-ion membrane dialysis in order to develop an ion fractionation process that can be applied to the enrichment and separation of metal ions in wastewater treatment.

The aim of this paper is to study the preferential transport behaviors of ternary ions system across the cation-exchange membrane in a batch dialyzer with a complexing agent. Cu^{+2} – Ni^{+2} – Zn^{+2} ions system was chosen for the experimental investigation because they are often associated in electroplating waste solutions of electronic industry. The complexing agents used in this study include malonic acid, oxalic acid and citric acid. The influences of the stoichiometric ratios of the complexing agent to the metal ions and the pH values of the feed solution on the dimensionless permeation fluxes of metal ions have been

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Nomenclature

a_i	activity of species i in solution
C_i	concentration of ion i (kmol m^{-3})
D_i	diffusion coefficient ion i ($\text{m}^2 \text{s}^{-1}$)
E	electric potential (V)
f	molar activity coefficient
F	Faraday's constant ($9.648 \times 10^4 \text{ C mol}^{-1}$)
J_i	permeation flux of ion i ($\text{mol m}^{-2} \text{s}$)
K_B^A	apparent selectivity coefficient
L	thickness of ion-exchange membrane (m)
R	universal gas constant ($8.314 \text{ kJ kmol}^{-1} \text{ K}^{-1}$)
X	normal direction to the ion-exchange membrane (m)
z_i	valence of ion i

Greek symbols

Φ	stoichiometric ratio of complexing agent to metal ions
Ψ	electrical potential (V)

Superscript

-	in the membrane phase
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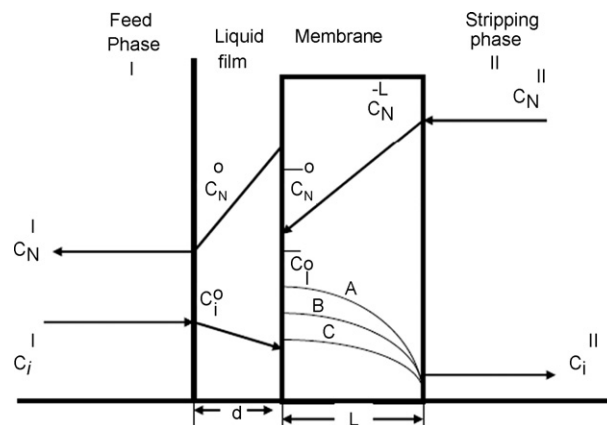


Fig. 1. The schematic mechanism of preferential transport behavior in ion-exchange membrane dialysis for a multi-ionic system.

For the metal ions and counter ion i , the transport flux is

$$J_i = -D_i \left(\frac{dC_i}{dx} + \frac{C_i z_i F}{RT} \frac{dE}{dx} \right), \quad i = A, B, C, N \quad (1)$$

For the common anion Y, the transport flux is

$$J_Y = -D_Y \left(\frac{dC_Y}{dx} + \frac{C_Y z_Y F}{RT} \frac{dE}{dx} \right) = 0 \quad (2)$$

where A, B and C denote the coded metal ions, N denotes the counter ion. E , D_i and C_i denote the diffusion potential, the diffusion coefficient and the concentration of ion i in the liquid phase, respectively. F , x and z_i denote the Faraday's constant, the distance normal to the membrane and the valence of ion i , respectively.

On the basis of the mass balance in the steady state, the following equation can be obtained:

$$z_A J_A + z_B J_B + z_C J_C + z_N J_N = 0 \quad (3)$$

The electroneutrality condition must be satisfied at any location of the electrolyte solution, i.e.,

$$z_A C_A + z_B C_B + z_C C_C + z_N C_N + z_Y C_Y = 0 \quad (4)$$

The potential gradient in Eqs. (1) and (2) can be eliminated by using Eqs. (3) and (4). Then, the concentration distribution of ion i in liquid film can be obtained by integrating these equations with the boundary condition, $C = C_i^0$ at $x = 0$, as follows:

$$C_i = \frac{C_i^0 [Q^0]^{z_i} - [J_i / D_i Q^{z_i}]}{Q^{z_i}}, \quad i = A, B, C, N \quad (5)$$

where

$$Q^0 = z_A C_A^0 + z_B C_B^0 + z_C C_C^0 + z_N C_N^0 \quad (6.1)$$

$$Q = Q^0 - \left(\frac{J_A}{D_A} + \frac{J_B}{D_B} + \frac{J_C}{D_C} + \frac{J_N}{D_N} \right) x \quad (6.2)$$

Next, let us consider the membrane-phase mass transport. If common ion, do not exists in the membrane and metal ions transport in one dimension, then the mass flux of each ion in the

investigated. A theoretical model is formulated on the bases of the Nernst–Planck equation and membrane–solution interface equilibrium. The theoretical approach can be obtained by using the numerical calculation of model equations.

2. Theoretical consideration

Consider an ion-exchange system in which a cation-exchange membrane separates two electrolyte solutions, i.e., one is the feed phase consisting of three bivalent cations species i with the common anion Y^- , and the other is the stripping phase composed of the N–Y electrolyte solution. The mechanism of the transport of counter ion across the cation-exchange membrane is postulated to consist of three rate processes [13,14]: (1) diffusion through the two liquid films adjacent to the membrane surface, (2) the ion-exchange reaction occurring at two membrane–solution interfaces, and (3) diffusion through the membrane.

It is assumed that the concentration of N–Y electrolyte solution and the agitation speed in the stripping side are sufficiently high to ignore the mass-transfer resistance of the stripping phase as compared to the other ones [17]. Furthermore, the film–membrane system has been allowed to adjust to a steady state during the dialysis operation, so that the ionic flux of the membrane is equal to that of the liquid phase [18]. The schematic transport mechanism is illustrated in Fig. 1.

Firstly, consider the mass transport of the counter ion in the liquid film of the feed phase. Based on to the Nernst–Planck diffusion equation, the transport flux of ion i can be expressed as the following [9–11].

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