Correlations of Ionic Mass Transfer Rate in Ion Exchange Membrane Electrodialysis

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The correlations of the Reynolds number, Re, the Schmidt number, Sc, and the shape factor of the electrodialysis cell, *de/L*, with the ionic mass transfer rate $(\mathrm{Nu})_{\mathrm{exp}}$ in ion exchange membrane electrodialysis are obtained from the measurement of the limiting current densities at various conditions. The limiting current densities of electrodialysis at constant temperature (25 °C) and various Re, Sc, and *de/L* are measured from the voltage–current curves. The experimental results within the laminar flow region show that the ionic mass transfer rate can be expressed by the empirical equation $(\mathrm{Nu})_{\mathrm{exp}} = 3.58\mathrm{Re}^{0.329}\mathrm{Sc}^{0.392}(de/L)^{0.354}$ in the range $25 < \mathrm{Re} < 2000$, $344 < \mathrm{Sc} < 2200$, and $0.06 < de/L < 0.36$ for a 0.03 N NaCl solution at 25 °C. It is found that the experimental results are coincident with the results predicted from the theoretical equation. The effect of Re on limiting current density in the turbulent flow region is measured at constant Sc and *de/L* in the range $2000 < \mathrm{Re} < 2800$. The ionic mass transfer rate can be expressed by $(\mathrm{Nu})_{\mathrm{exp}} = K\mathrm{Re}^{0.78}$. This empirical equation is similar to the equation obtained from fully developed turbulent mass transfer in an annulus electrodialysis cell.

The ionic transfer rate in electrodialysis is usually controlled by the diffusion process in the boundary layer or the electron transfer process on the surface of the electrodes, if the solution contains large quantities of supporting electrolyte (*1, 3, 12, 14*). However, the rate of ionic mass transfer in the boundary layer will be determined by the interaction of three driving forces, i.e., the forces caused by convection, diffusion by concentration gradient, and electromigation by potential gradient, if very little or no supporting electrolyte is contained in the solution (*15*).

Although the ion exchange membrane electrodialysis systems are better to operate at high current densities (desalination rates) for an optimum balance between capital and operating costs, the practicable current densities are limited by polarization due to a remarkable difference in transference number of ion between solution and membrane phase. As the ions are forced out of the dialysate cells under the action of the applied electric field, the salt concentration near the membranes decreases and the ohmic resistance of the cells increases. The concentration drop increases with applied potential (driving force) and a limiting current tends to be reached when the salt concentration at the membrane–solution interface decreases toward zero. When operating current density is larger than the limiting value, the H^+ and OH^- ions in the solution participate in the process, the electrodialysis is ineffective in desalination or concentration, and low concentration at the diffusion layer represents high electrical resistance. Practical systems are operated below the limiting current not only because the energy losses increase rapidly at higher currents but also because the transfer of H^+ and OH^- ions gives rise to pH changes which in turn may cause scale precipitation and other harmful side effects (*7*). The value of the limiting current is influenced by the concentration, temperature, viscosity of solution, and hydrodynamic and other operation conditions (*8–10*) and can be increased by increasing the flow rate or by introducing turbulence promoters into the cells.

The concentration polarization phenomenon is essentially important in the ion exchange membrane electrodialysis. Some fundamental features of concentration polarization have been revealed by Rosenberg and Tirrell (*16*), Yamabe (*23, 24*), Seno et al. (*17, 18*), Mandersloot and Hicks (*13*), Forgace et al. (*4*), Huang et al. (*8–10*), and Spiegler (*19*).

The distribution and diffusion of ion in the membrane phase for various electrolyte solutions have been studied intently by Helfferich (*6*) and Tuwiners (*20*). Because of the existence of concentration polarization in the diffusion layer, the ionic mass transfer rate in electrodialysis is partly or mostly controlled by the boundary layers in a desalting compartment. Therefore, the ionic mass transfer rate could be satisfactorily discussed in liquid phase only and described by a few dimensionless parameters. In this work, the effects of the Reynolds number Re, Schmidt number Sc, and shape factor *de/L* on the Nusselt number Nu have been studied and discussed under the condition of limiting current density. The experimental data obtained for a two-dimensional flow through a straight channel with flat parallel ion exchange membranes walls in the laminar flow region are compared with the theoretical equation.

Theoretical Consideration

For the ion exchange membrane electrodialysis, the equation of convective diffusion for binary electrolyte solution can be expressed by the following equation (*1*)

$$(\partial C/\partial t) + V\cdot\nabla C = \nabla(D\nabla C) \qquad (1)$$

where

$$D = (Z_+ - Z_-)D_+D_-/(Z_+D_+ - Z_-D_-) \qquad (2)$$

The physical meaning of D can be considered as the molecular diffusion coefficient of the binary electrolyte in solution.

In order to find the ionic concentration distribution and ionic mass transfer rate in ion exchange membrane electrodialysis, we consider the electrolyte solution flow through two flat parallel ion exchange membranes at steady state, i.e., $\partial C/\partial t = 0$. For this two-dimensional flow and constant molecular diffusivity, eq 1 can be written as

$$v_x(\partial C/\partial x) + v_y(\partial C/\partial y) = D(\partial^2 C/\partial y^2) \qquad (3)$$

with the boundary conditions

$$\begin{cases} y = 0 & C = C_s \\ x = 0 & C = C_\infty \\ y = \infty & C = C_\infty \end{cases} \qquad (4)$$

The solution of eq 3 with boundary conditions, eq 4, is given as (*14*)

$$\frac{C - C_s}{C_\infty - C_s} = \frac{1}{\Gamma(4/3)} \int_0^{\xi} e^{-\xi^3} d\xi \qquad (5)$$

where

$$\xi = y(\beta^{1/2}) \Big/ \left[9D \int_0^x (\beta)^{1/2}\, dx \right]^{1/3} \qquad (6)$$

and $\Gamma(4/3) = 0.89298$.

At the condition of the limiting current density, the concentration at the membrane surface is equal to zero. For a two-dimensional flow channel, $\beta(x)$ can be expressed as (*14*)

$$\beta = 6\langle V\rangle/h \qquad (7)$$

The concentration gradient at the membrane surface can be obtained from eq 5, 6, and 7, and the result is given by

$$\nabla C\big|_{y=0} = \frac{C_\infty}{\Gamma(4/3)} \frac{(6\langle V\rangle/h)^{1/2}}{[9D(6\langle V\rangle/h)^{1/2}x]^{1/3}} \tag{8}$$

The dimensionless ionic mass transfer rate called the Nusselt number, Nu, in present ion exchange membrane electrodialysis can be defined as

$$\mathrm{Nu}(x) = -N + de/C_\infty D_+ \tag{9}$$

where *de* is the equivalent diameter of the channel, and for a two-dimensional flow channel *de* is equal to 2*h*.

For a binary electrolyte solution under limiting current density, if the diffusivity of the cation is smaller than that of the anion in solution, the concentration polarization is developed first in the cation exchange membrane surface, and the following equations are satisfied.

$$N_+ = I_{\mathrm{lim}}/Z_+F \tag{10}$$

and

$$N_+/\nu_+ = -(1 - Z_+/Z_-)D_+\nabla C \tag{11}$$

From eq 8, 9, and 11 and $h = de/2$, the Nusselt number is

$$\mathrm{Nu}(x) = \nu_+ \left(1 - \frac{Z_+}{Z_-}\right) \frac{1}{\Gamma(4/3)} \left(\frac{4\langle V\rangle de^2}{3Dx}\right)^{1/3} \tag{12}$$

For sodium chloride solution, $\nu_+ = 1$, $Z_+ = 1$, and $Z_- = -1$, the above equation becomes

$$\mathrm{Nu}(x) = 2.465(\mathrm{ReSc}(de/x))^{1/3} \tag{13}$$

where Re represents the Reynolds number, $\langle V\rangle de/\nu$, Sc represents the Schmidt number, ν/D, and ν is the kinematic viscosity, μ/ρ.

The average value of the Nusselt number, corresponding to the average mass transfer rate over the length *L* of the ion exchange membrane, is

$$\mathrm{Nu_{av}} = (1/L)\int_0^L \mathrm{Nu}(x)\,\mathrm{d}x = 3.7(\mathrm{ReSc}(de/L))^{1/3} \tag{14}$$

The use of the dimensionless groups such as Re, Sc, and *de*/*L* instead of ordinary variables such as concentration, velocity, viscosity, density, and the thickness of channel in this study is to simplify the experimental conditions and to compare easily with the different systems.

Experimental Section

Membrane. The ion-exchange membranes used in this work were Selemion CMV and AMV. Both cation exchange membrane Selemion CMV and anion exchange membrane Selemion AMV are homogeneous membranes reinforced with vinyl chloride fiber cloth. Their chemical structures and physical properties are listed in Table I. These membranes were preconditioned with 0.5 N NaCl solution before used.

Chemical Reagents. The chemical reagents used in this work were E.P. grade sodium chloride, potassium chloride, potassium cyanide, potassium nitrate, and glycerine. They were all of E.P. reagents and were used without any further purification. The water used for the preparation of the solution was pretreated by ion-exchange resin.

Cells and Procedures. There are many ways for the measurement of the limiting current density in the ion exchange membrane electrodialysis (*2*). In the present work, the limiting current densities were measured from the inflection point of the current density vs. voltage curves obtained in the two-dimensional flow electrodialysis channel at various experimental conditions.

As shown in Figure 1, the two-dimensional flow electrodialysis channel is composed of two flat PMMA plates with a square exposure area of 20 cm^2. The ion exchange membranes Selemion CMV and AMV are adhered to the inner faces of these two PMMA plates. The Ag–AgCl electrodes are equipped at the outer sides of the membranes at the square exposure area for the voltage measurement. The flow diagram of the regulation of flow rate and the measurement of current density and voltage is shown in Figure 2. The sodium chloride solutions were fed into the electrodialysis channel by circulating pump at a given flow rate from a constant-temperature reservoir. The constant voltages were applied by a regulated dc power supply, and the currents were automatically recorded by an electronic recorder with a 1-Ω standard resistor.

Table I. Specifications and Working Characteristics of Selemion CMV and AMV Membranes

	Selemion CMV	Selemion AMV
	High acidic ion exchange membrane	High basic ion exchange membrane
Base material	Tevilon cloth (PVC)	Tevilon cloth (PVC)
Thickness,[a] mm	0.12–0.15	0.11–0.14
Effective electrical resistance,[b] Ω/cm^2	2.5–3.5	3.0–4.5
Specific electrical resistance, Ω cm	190–230	280–320
Transport no.[c]		
t_{Na^+}	0.91–0.93	
t_{Cl^-}		0.94–0.96
Bursting strength, kg/cm^2	6–8	4–7

[a] Measured by micrometer in wet state. [b] Measured in 0.5 N NaCl solution at 25 °C. [c] Obtained from membrane potential resulting from membrane separation of 0.5 N NaCl and 1 N NaCl solutions at 25 °C.

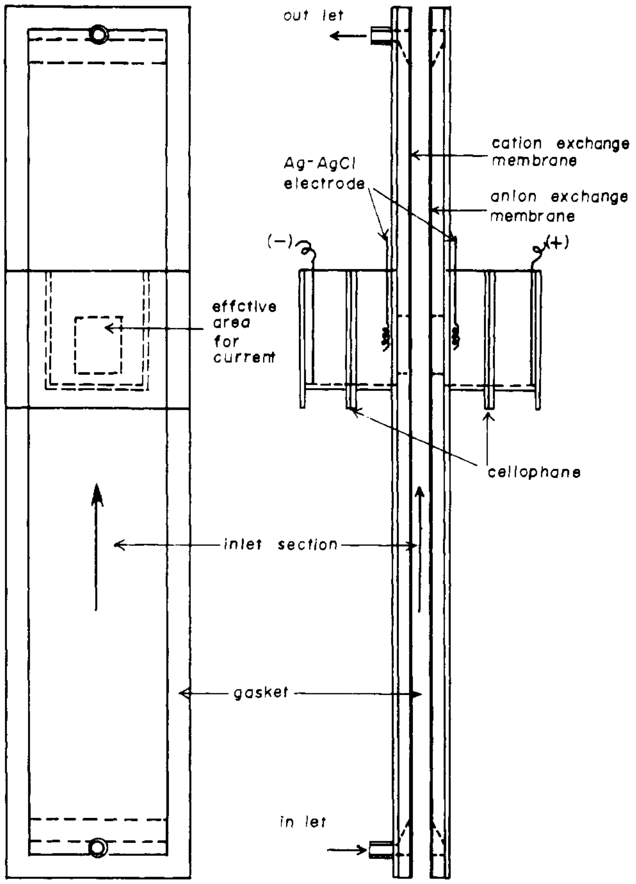

Figure 1. Front and side view of the two-dimensional flow channel.

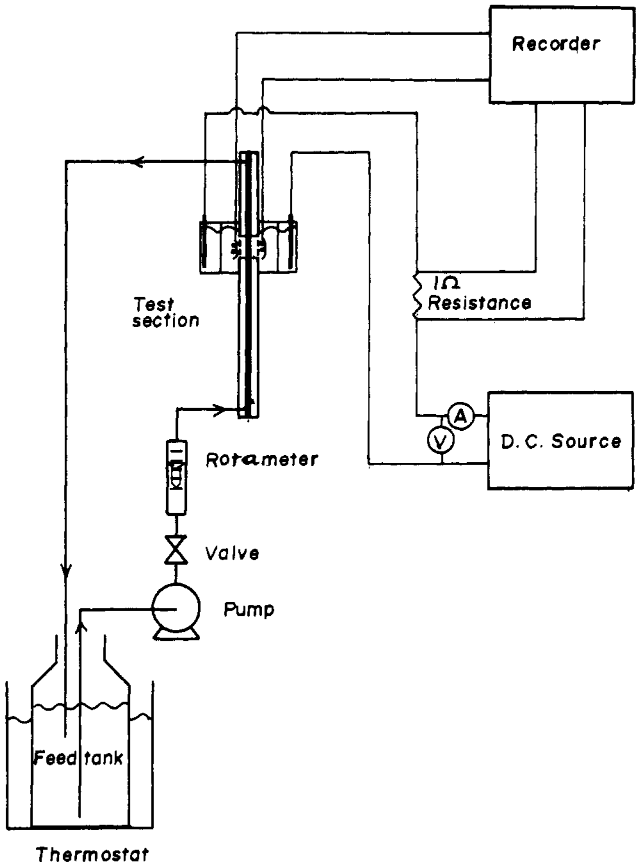

Figure 2. Flow diagram for measuring the limiting current.

In all experiments the working fluid in the dialysate cell was a solution of sodium chloride in distilled water, with a feed concentration of 0.03 N in most tests. The solution used in the electrode cells was also NaCl solution. The concentration of the NaCl solution was measured with a conductivity meter.

The effective applied voltage, $\Delta\phi$, to a single desalting cell can be expressed by

$$\Delta\phi = \Delta\phi_{\text{applied}} - \Delta\phi_M - (r_{AM} + r_{CM})I$$
$$= \Delta\phi_{\text{applied}} - \frac{2RT}{F}\ln\frac{a_c}{a_d} - (r_{AM} + r_{CM})I \quad (15)$$

where $\Delta\phi_{\text{applied}}$ is the applied voltage, $\Delta\phi_M$ is the membrane potential, a_c and a_d are the activities of the concentrated and diluted solution in the electrodialyzer, respectively, and $(r_{AM} + r_{CM})I$ is the ohmic drop in both the anion- and cation-exchange membranes. This term is usually negligibly small as compared with $\Delta\phi_{\text{applied}}$ and $\Delta\phi_M$ if the concentration of the desalting solution is lower than 0.2 N (*11*).

In this experiment two graphite plates were used as working electrodes. A pair of Ag-AgCl reversible electrodes were placed at the outer sides of the desalting compartment in order to measure the actual applied voltage, $\Delta\phi_{\text{applied}}$. The concentration of the concentrated compartment was kept at the same as that of the desalting compartment, $a_c = a_d$, then the membrane potential was diminished. So the applied voltage measured by Ag-AgCl electrodes was the effective applied voltage. The limiting current and the limiting voltage were determined by the current-voltage curves.

Equation 13, indicates that the ionic mass transfer rate is affected by the dimensionless groups Re, Sc, and *de*/*L*. Therefore an empirical formula for the ionic mass transfer rate can be expressed in the same form as eq 13 for the comparison, i.e.

$$(\text{Nu})_{\text{exp}} = a\text{Re}^b\text{Sc}^c(de/L)^d \quad (16)$$

Table II. Limiting Current Densities at Various Reynolds Numbers (25 °C) (System: Selemion CMV and AMV in 0.03 N NaCl Solution, Sc = 558.1, *de*/*L* = 0.06)

Re	I_{lim}, mA/cm^2	Re	I_{lim}, mA/cm^2
25.78	3.795	644	14.22
51.5	5.29	772.5	16.58
77.3	5.45	901	18.09
154.5	6.84	1030	18.72
206.0	8.29	1277	20.07
257.8	8.83	1545	21.40
309	10.00	2060	32.38
412	10.91	2578	37.45
515	13.38	2835	40.70

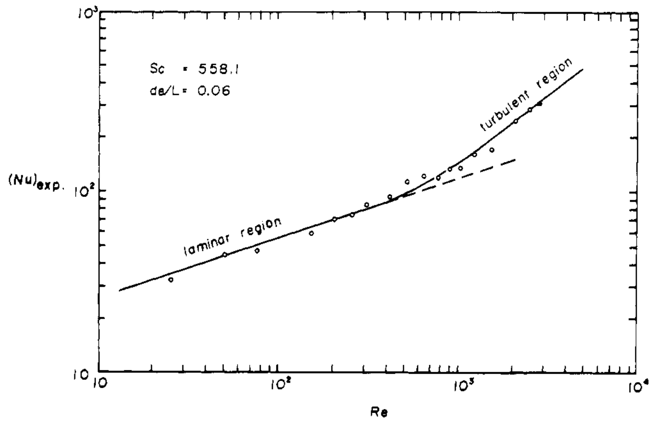

Figure 3. Experimental relation between Re and $(\text{Nu})_{\text{exp}}$.

The constant *a*, *b*, *c*, and *d* can be determined from the experiments.

Results and Discussions

The constant *b* in eq 16 can be obtained from the experimental relation between $(\text{Nu})_{\text{exp}}$ and Reynolds number at constant Schmidt number and shape factor of electrodialysis cell. The limiting current densities of the ion exchange membrane Selemion CMV in 0.03 N NaCl solution have been measured from the inflection point of the current density-voltage curves under 25 °C for various flow rates of NaCl solution into the two-dimensional flow electrodialysis channel from 5 to 550 L/h. In this experiment, the length of the exchange membrane and the thickness of the channel are kept constant at 5 and 0.15 cm, respectively. The Reynolds number of these experimental conditions are correspondent to 25.78–2835. The kinematic viscosity of NaCl solution at 25 °C is equal to 8.986×10^{-3} cm^2/s, and the ionic mobility and the diffusivity of sodium ion at 25 °C are 52.0×10^{-5} cm^2/(V s) and 1.61×10^{-5} cm^2/s, respectively. The obtained limiting current densities at various Reynolds numbers are given in Table II.

The values of $(\text{Nu})_{\text{exp}}$ at various Reynolds numbers can be calculated from the experimental limiting current densities by using eq 9 and 10. The relation between Re and $(\text{Nu})_{\text{exp}}$ is shown in Figure 3. The result shows that the slope of the curve has an obvious change when the Reynolds number is larger than 2000. It indicates that there is a different relationship between Re and $(\text{Nu})_{\text{exp}}$, for the laminar region and turbulent region.

In the laminar region ($\text{Re} < 600$), the slope of the curve, log $(\text{Nu})_{\text{exp}}$ vs. log Re, keeps constant at 0.329 for $\text{Re} < 600$, and it increases gradually from 0.329 to 0.78 as the Reynolds number increases from 600 to 2000. Because of the ripple surface of the exchange membrane at the test section due to the swelling of the ion-exchange membrane, the fluid tends to become a turbulent flow as $\text{Re} > 600$. The experimental relation between $(\text{Nu})_{\text{exp}}$ and Re in the laminar region can be written as

$$(\mathrm{Nu})_{\mathrm{exp}} = K_1\mathrm{Re}^{0.329} \tag{17}$$

Comparing eq 17 with eq 14, it shows that the constant 0.329 obtained from the experiment is very agreeable with the theoretical value 0.333.

In the turbulent region (Re > 2000), the slope of the curve is kept at 0.78, and the relation between $(\mathrm{Nu})_{\mathrm{exp}}$ and Re is expressed by

$$(\mathrm{Nu})_{\mathrm{exp}} = K_2\mathrm{Re}^{0.78} \tag{18}$$

The diffusion layer of the electrolyte solution at membrane surface is very thin for the turbulent flow, and the concentration gradient across the layer is very large, so the accurate concentration gradient at the membrane surface can hardly be obtained by numerical methods. Therefore the theoretical relation between $(\mathrm{Nu})_{\mathrm{exp}}$ and Re for the present system cannot be derived precisely. However, the mass transfer in an annulus electrodialysis cell can be expressed by the following empirical relation obtained from the correlation for the fully developed turbulent flow (*1*)

$$(\mathrm{Nu})_{\mathrm{exp}} = 0.023\mathrm{Re}^{0.8}\mathrm{Sc}^{1/3} \tag{19}$$

Comparing eq 18 with eq 19, it shows that the constant 0.78 is fairly agreeable with the value 0.8 obtained by others.

In order to see the effect of the Schmidt number on the ionic mass transfer in ion exchange membrane electrodialysis, the limiting current densities of the ion exchange membrane Selemion CMV in 0.03 N NaCl solution have been measured for various Schmidt numbers at constant Reynolds number and constant shape factor of the electrodialysis cell, $de/L = 0.06$. The values of the Schmidt number of 0.03 N NaCl solution can be changed by changing the temperature of electrolyte solution or adjusting the viscosity of solution. In this experiment, glycerine was added to the electrolyte solution in order to change the viscosity of solution. The viscosity of solution was measured by an Ostwald viscometer. When the Schmidt number is increasing, the flow rate of solution must also be increasing in order to keep the Reynolds number constant.

The relations between $(\mathrm{Nu})_{\mathrm{exp}}$ and the Schmidt number for constant Reynolds numbers 250 and 1200 in 0.03 N NaCl with the constant shape factor, $de/L = 0.06$, are shown in Figure 4. It indicates that $(\mathrm{Nu})_{\mathrm{exp}}$ increases as the Schmidt number increases at constant Reynolds number and shape factor of electrodialysis cell, and the slope of these straight lines in Figure 4 is equal to 0.392 for Re = 250 and Re = 1200 in the laminar flow region. Therefore, the experimental relation between $(\mathrm{Nu})_{\mathrm{exp}}$ and Schmidt number at constant Reynolds number and shape factor of the electrodialysis cell can be expressed by the following equation

$$(\mathrm{Nu})_{\mathrm{exp}} = K_3\mathrm{Sc}^{0.392} \tag{20}$$

These results are fairly reasonable in comparison with the theoretical equation, eq 14.

The empirical constant d in eq 16 can be obtained from the experimental relation between $(\mathrm{Nu})_{\mathrm{exp}}$ and the shape factor of the electrodialysis cell, de/L, at constant Reynolds number and Schmidt number. In this investigation, the width and the length of the electrodialysis channel are fixed at 12 and 5 cm, respectively, and the thickness of the channel is changed from 0.2 to 0.9 cm in order to change the value of de/L from 0.08 to 0.36. Reynolds numbers and Schmidt numbers are not varied by changing the equivalent diameter of the electrodialysis channel when the volumetric flow rate, temperature, and concentration of the electrolyte solution are kept constant. In this experiment, the volumetric flow rate of sodium chloride solution (0.03 N) is kept at 20, 50, 125, and 400 L/h at constant temperature 25 °C; the Reynolds numbers of these experimental conditions correspond to 103, 257.5, 644, and 2060.

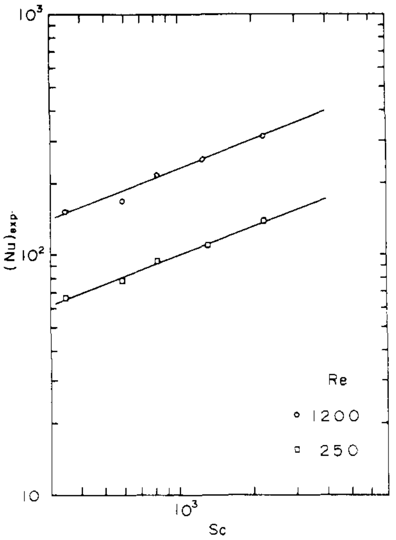

Figure 4. Experimental relation between Sc and $(\mathrm{Nu})_{\mathrm{exp}}$ for $de/L = 0.06$.

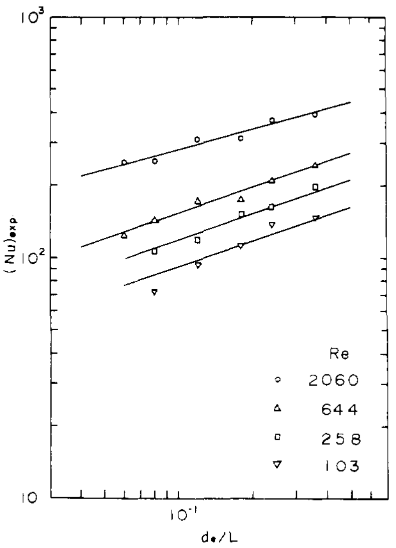

Figure 5. Experimental relation between de/L and $(\mathrm{Nu})_{\mathrm{exp}}$ for Sc = 558.

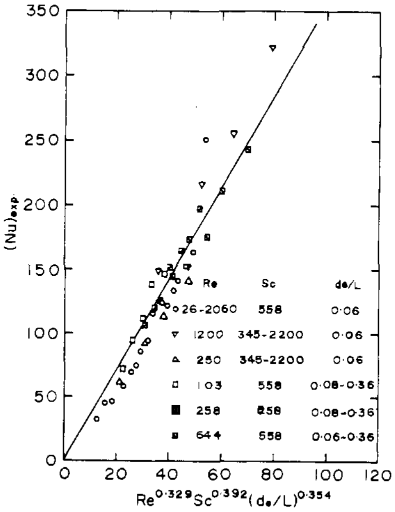

Figure 6. Experimental relation in the laminar region.

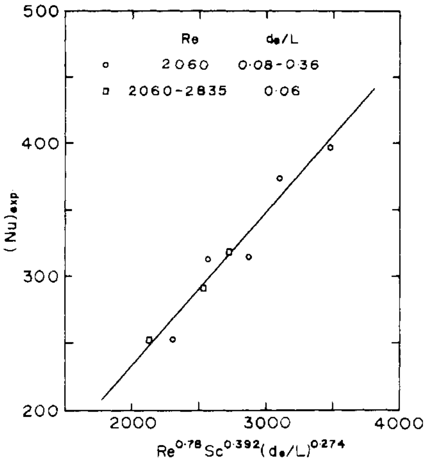

Figure 7. Experimental relation in the turbulent region.

The experimental relations between $(Nu)_{exp}$ and de/L at various Reynolds numbers for the ion exchange membrane Selemion CMV in 0.03 N NaCl solution are shown in Figure 5. The constants d found from the slope of these straight lines in Figure 5 are all equal to 0.354 for the runs of Reynolds numbers 103, 257.5, and 644, and equal to 0.274 for the run of Reynolds number 2060. Then the experimental relation between $(Nu)_{exp}$ and de/L at constant Schmidt number and Reynolds number in the laminar flow region can be written as

$$(Nu)_{exp} = K_4(de/L)^{0.354} \tag{21}$$

while in the turbulent flow region can be expressed by

$$(Nu)_{exp} = K_5(de/L)^{0.274} \tag{22}$$

It shows that the experimental results are in fair coincidence with the theoretical equation in the laminar flow region.

Combining the empirical relations of eq 17, 20, and 21 we obtain the experimental formula of the ionic mass transfer rate of ion exchange membrane electrodialysis in the laminar flow region

$$(Nu)_{exp} = aRe^{0.329}Sc^{0.392}(de/L)^{0.354} \tag{23}$$

and in the turbulent flow region from eq 18, 20, and 22

$$(Nu)_{exp} = a'Re^{0.78}Sc^{0.329}(de/L)^{0.274} \tag{24}$$

The correlations of the experimental data of $(Nu)_{exp}$ with $Re^{0.329}Sc^{0.392}(de/L)^{0.354}$ in the laminar flow region and $(Nu)_{exp}$ with $Re^{0.78}Sc^{0.392}(de/L)^{0.274}$ in the turbulent flow region are shown in Figures 6 and 7, respectively. It can see that there are linear relationships between $(Nu)_{exp}$ and $Re^{0.329}Sc^{0.392}(de/L)^{0.354}$ for the laminar flow region and $(Nu)_{exp}$ and $Re^{0.78}Sc^{0.392}(de/L)^{0.274}$ for the turbulent flow region. The constants $a = 3.58$ in eq 23 and $a' = 0.116$ in eq 24 are obtained from the slope of these relations by the least-squares method. Therefore, the ionic mass transfer rate in the ion exchange membrane electrodialysis with the functions of Reynolds number, Schmidt number, and the shape factor of electrodialysis cell in the laminar flow region can be expressed by

$$(Nu)_{exp} = 3.58Re^{0.329}Sc^{0.392}(de/L)^{0.354} \tag{25}$$

and in the turbulent flow region

$$(Nu)_{exp} = 0.116Re^{0.78}Sc^{0.392}(de/L)^{0.274} \tag{26}$$

It indicates that eq 25 is similar to the theoretical equation, eq 14, within the laminar flow region.

Glossary

C	concentration of binary electrolyte, mol/cm^3
C_s	concentration of ion at the membrane surface, mol/cm^3
C_∞	concentration of ion outside the boundary layer, mol/cm^3
D	molecular diffusion coefficient, cm^2/s
D_+	diffusion coefficient of cation, cm^2/s
D_-	diffusion coefficient of anion, cm^2/s
de	equivalent diameter of the channel, cm
F	Faraday's constant, C/equiv
h	thickness of the channel, cm
I_{lim}	limiting current density, A/cm^2
L	length of the ion exchange membrane, cm
Nu	Nusselt number, dimensionless
N_+	flux of cation, mol/(cm^2 s)
Re	Reynolds number, dimensionless
Sc	Schmidt number, dimensionless
t	time of electrodialysis, s
V	fluid velocity, cm/s
$\langle V \rangle$	average velocity of the electrolyte solution, cm/s
v_x	velocity in x direction, cm/s
v_y	velocity in y direction, cm/s
x	coordinate along the membrane surface from its upstream, cm
y	coordinate normal to the membrane surface, cm
Z_+	valence or charge number of cation
Z_-	valence or charge number of anion

Greek Letters

β	$6\langle v \rangle / h$, s^{-1}
ν_+	number of cations produced by dissociation of one molecule of electrolyte in solution

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