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A Method for Estimating the Limiting Current Density in Electrodialysis

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Abstract

An equation for estimating the limiting current density (LCD) in the electrodialytic equipment with spacers is derived by using the equation of the flow distribution and by assuming that the eddy diffusivity caused by the spacer is proportional to Re. The empirical constants of the eddy diffusivity in the equation were determined by measurement of LCD using spacers, electrolytes, feed concentrations, operating temperatures, and flow velocities. The results obtained show that LCD in the electrodialysis can be estimated from the equation derived here.

INTRODUCTION

Electrodialysis with ion-exchange membranes has been employed as one of the methods of separation with membranes in industrial fields. In the operation of the equipment, however, the limiting electric current appears at a heigher electric current, owing to polarization of the concentration in solution.

As for the limiting current density (LCD) in the electrodialytic equipment with spacers, Belfort et al. (*1*) indicated that LCD is influenced by the characteristics of the spacer. Estimating equations for

LCD were introduced by Mandersloot et al. (*2*) experimentally, by Rosenberg et al. (*3*) and Cowan et al. (*4*) with turbulent theory, and Kitamoto et al. (*5*) with dimensional analysis. Sonin et al. (*6–8*) investigated LCD of equipment in which some packing material was inserted into the compartment instead of a spacer, and Winograd et al. (*9*) proposed an experimental estimating equation for LCD with the characteristics of a spacer. Further, other workers (*10–14*) have investigated LCD. However, an estimating equation of LCD derived by the use of the velocity distribution in the ion-exchange compartment with spacers has not been determined. Even if the analytical description of LCD is only an approximation, the number of experiments and calculations for design that must be performed would be deduced.

This paper provides a method for estimating LCD in electrodialysis with spacers. An estimating equation for LCD is derived by using the equation of the flow distribution in the ion-exchange compartment in the previous paper (*15*) and by assuming that the eddy diffusivity caused by a spacer is proportional to Re.

In order to determine the empirical constants of eddy diffusivity in the equation, the values of LCD are measured under various conditions; several kinds of spacers and electrolytes, different feed concentrations, various operating temperatures, and different flow velocities are employed. Then the equation to estimate LCD in electrodialysis is checked by the experimental results.

EQUATION FOR ESTIMATING LCD

1. Flux in the *y* Direction

The electrolyte transfer in the ion-exchange compartment with spacers as shown in Fig. 1. may be caused concurrently by the following driving forces:

(a) Electric field
(b) Molecular diffusivity by concentration gradient
(c) Eddy diffusivity due to spacer
(d) Convection

The electrolyte transfer by electric field (a) has been discussed previously (*16*), and the flux by convection in the y direction (d) is negligibly small.

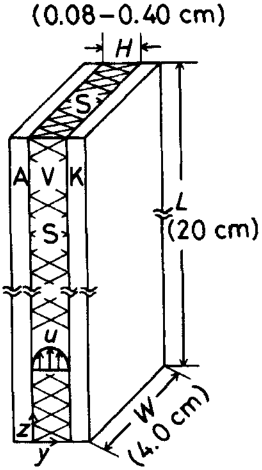

FIG. 1. View of the electrodialytic cell. A: Anion-exchange membrane. *H*: Channel thickness of the compartment. K: cation-exchange membrane. *L*: Height of the compartment. S: Spacer. *u*: Flow velocity. V: Ion-exchange compartment. *W*: Width of the compartment.

Since the fluid in the equipment with a spacer is well mixed by the presence of the spacer (*15*), it is presumed that the electrolyte transfer by molecular diffusivity (b) is negligible compared with that by the eddy diffusivity due to the spacer (c).

The results calculated from the data of Page et al. (*17*) and Sherwood et al. (*18*) indicate that the relation between the eddy conductivity of the turbulent flow (K_e) and the molecular conductivity (K) is represented as Eq. (1) in the region near the wall, i.e., $0 < y < H/4$.

$$\frac{K_e}{K} = a' \, \mathrm{Re}^{b'} \left(\frac{y}{H} \right) \tag{1}$$

where a' and b' are constants, Re is the Reynolds number, and y is the distance from one wall to the parallel wall of thickness is H.

The influence of y/H was considered in the eddy viscosity of the flow distribution in the compartment with a spacer (*15*). The concentration distribution in the compartment can be regarded as a kind of turbulent flow, because the fluid in the compartment is well mixed by the fiber of the spacer. Therefore, the ratio of the eddy diffusivity (D_e) due to the spacer to the molecular diffusivity (D) can be written as in Eq. (2), by analogy with Eq. (1):

$$\frac{D_e}{D} = a \, \mathrm{Re}^b \tag{2}$$

where a and b are constants influenced by the characteristics of the spacer, $\mathrm{Re} = Hu_0/\nu$.

Thus, the flux in the y direction in Fig. 1 is represented in Eq. (3) by the first and third driving forces:

$$\begin{aligned} N_y &= \frac{\tau}{F} I + D_e \frac{\partial C}{\partial y} \\ &= \frac{\tau}{F} I + aD \, \mathrm{Re}^b \frac{\partial C}{\partial y} \end{aligned} \tag{3}$$

where C is the concentration of the solution, F is Faraday's constant, I is the electric current density, and τ is the transport number of the electrolyte in solution.

2. Flux in the *z* Direction

In a previous paper (*15*) we proposed Eq. (4) for the flow distribution in an ion-exchange compartment with a spacer:

$$u = u_0 \frac{m[(m+1)\ln(2my/H + 1) - 2my/H]}{(m+1)^2 \ln(m+1) - 1.5m^2 - m} \tag{4}$$

where m is a variable represented by using the characteristics (d, n, t, and ε) of a spacer as in

$$m = 2.1 \times 10^5 [n(t-d)]^{2.4} \left[\frac{(1-\varepsilon)^2}{\varepsilon^3}\right]^{0.8} \tag{5}$$

When the equation of mass flux in the z direction given by using Eq. (4) is applied to the fundamental one, it is very difficult to solve theoretically, but it is necessary to investigate the influence of each operating factor on LCD. Therefore, we use the approximate method that the thickness of the diffusion boundary layer is smaller than that of the hydrodynamic boundary layer when Sc ($= \nu/D$) is large (*19*), and the flow distribution can be represented as linear near the wall of the compartment as in

$$u = \frac{du}{dy}\bigg|_{y=0} y$$

$$= 2\frac{Mu_0}{H}y \tag{6}$$

where M is a variable expressed by using m as in

$$M = \frac{m^3}{(m+1)^2 \ln(m+1) - 1.5m^2 - m} \tag{7}$$

Therefore, the flux of the z direction in Fig. 1 is given as Eq. (8) from Eq. (6):

$$N_z = -uC$$

$$= -2\frac{Mu_0}{H}Cy \tag{8}$$

3. Distribution of Concentration

The ratios $\partial I/\partial y$ and $\partial u_0/\partial z$ are zero in the compartment. From Eqs. (3), (8), and the equation of continuity (*20*), the fundamental equation can be written

$$2\frac{Mu_0}{D_eH}y\frac{\partial C}{\partial z} = \frac{\partial^2 C}{\partial y^2} \tag{9}$$

This equation can be solved under the following boundary conditions (*21*):

$$\left.\begin{array}{l} C = C_w \text{ at } y = 0 \\ C = C_0 \text{ at } z = 0 \end{array}\right\} \tag{10}$$

and Eq. (11) is obtained:

$$\frac{C - C_w}{C_0 - C_w} = \frac{1}{\Gamma(4/3)}\int_0^{\xi} e^{-X^3}dX \tag{11}$$

in which

$$\xi = y\left(\frac{2Mu_0}{9D_eHz}\right)^{1/3} \tag{12}$$

where C_0 and C_w are the concentrations of the inlet solution and of the solution on wall of the compartment, respectively. Equation (11) represents the concentration distribution in a compartment with a spacer.

4. Estimating Equation of LCD

The mean flux in solution on the wall of the compartment (N_s) is found from Eqs. (3) and (11):

$$\begin{aligned} N_s &= \frac{1}{L}\int_0^L\left(\frac{\tau}{F}I + D_e\frac{\partial C}{\partial y}\bigg|_{y=0}\right)dz \\ &= \frac{\tau}{F}I + \frac{3}{2}\,\frac{(C_0 - C_w)D_e}{\Gamma(4/3)}\left(\frac{2Mu_0}{9D_eHL}\right)^{1/3} \end{aligned} \tag{13}$$

On the other hand, the mean flux in the ion-exchange membrane (N_m) is represented by Eq. (14), as indicated in our earlier papers (*22–24*):

$$N_m = \frac{\bar{\tau}}{F}I \tag{14}$$

where $\bar{\tau}$ is the transport number of an ion in the ion-exchange membrane.

When C_w is 0, I (electric current density) is equal to I_s (LCD). Therefore, Sh_s which represents LCD (I_s) in the electrodialytic equipment with spacers, can be derived from the condition of $N_s = N_m$:

$$\mathrm{Sh}_s = \frac{Q}{\bar{\tau} - \tau}\left(\frac{H}{L}\right)^{1/3} M^{1/3}\mathrm{Re}^{(1+2b)/3}\mathrm{Sc}^{1/3} \tag{15}$$

in which $\mathrm{Sh}_s = HI_s/C_0DF$, and Q is a dimensionless variable represented by

$$Q = 1.017a^{2/3} \tag{16}$$

EXPERIMENTAL EQUIPMENT AND PROCEDURE

The electrodialytic equipment used in this study is of the filter press type as shown in Fig. 2.

Cation- and anion-exchange membranes were alternately piled up in an electrodialytic stack. The stack was composed of five compartments each for the condensed and desalted solutions. There were two additional compartments for plus and minus electrodes on both sides of the stack.

The net area for the electric current of each membrane was 20 cm × 4.0 cm ($L \times W$). Four kinds of spacers were tested for each experiment. The characteristics of the spacers are shown in Table 1.

The cation- and anion-exchange membranes used were Neosepta CL-25T and AV-4T, respectively, produced by Tokuyama Soda Co.

The conditions of the present experiments are shown in Table 2. The concentration (C_0) of the inlet solution was kept constant during each

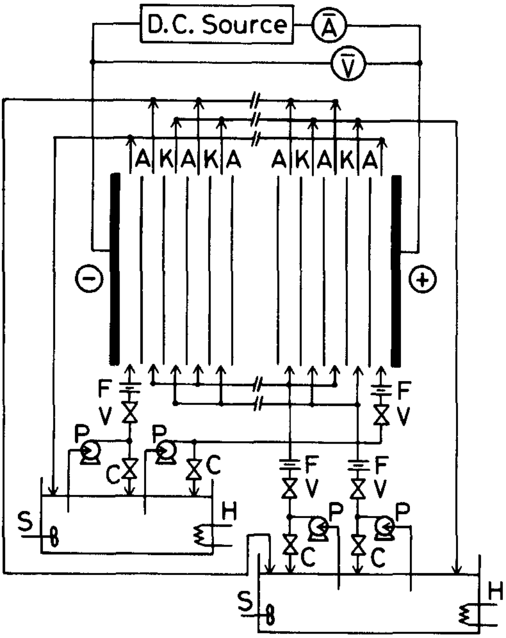

FIG. 2. Flow diagram of the electrodialytic equipment. A: Anion-exchange membrane. C: Control valve, F: Flowmeter. H: Heater and controller. K: Cation-exchange membrane. P: Pump. S: Stirrer. V: Valve.

TABLE 1
Characteristics of Spacers

Spacers	*d* (cm)	*t* (cm)	ε^a	Textile	Material
A	0.0238	0.30	0.940	Honeycomb	Saran
B	0.0217	0.20	0.938	Honeycomb	Polypropylene
C	0.0568	0.15	0.727	Pointed twill	Polypropylene
D	0.0294	0.08	0.600	Pointed twill	Polypropylene

[a] ε was calculated from both the apparent volume of a spacer and the increasing volume of water when its spacer was placed into the measuring cylinder with water.

experiment. The values of C_0 for each electrolyte were measured with an atomic absorption spectrophotometer (Perkin-Elmer 403). The values of D, ν, τ and $\bar{\tau}$ in the operating conditions were taken from references *24–27.*

From the values of I and V (electric potential) measured in each condition, the relations between V/I and $1/I$ were plotted. The sharp

TABLE 2
Experimental Conditions

Run	Electrolyte	Spacer	*n*	*H* (cm)	$C_0 \times 10^3$ (g-eq/cm^3)	*T* (°C)	Re	Sc
1	NaCl	A	1	0.30	0.05	25	9.30–465	557
2	NaCl	B	1	0.20	0.05	25	9.30–465	557
3	NaCl	B	2	0.40	0.05	25	9.30–465	557
4	NaCl	C	1	0.15	0.05	25	9.30–465	557
5	NaCl	C	2	0.30	0.05	25	9.30–465	557
6	NaCl	D	1	0.08	0.05	25	9.30–205	557
7	NaCl	D	5	0.40	0.05	25	9.30–465	557
8	NaCl	A	1	0.30	0.005	25	23.2–465	557
9	NaCl	A	1	0.30	0.01	25	23.2–465	557
10	NaCl	A	1	0.30	0.10	25	23.2–465	557
11	KCl	A	1	0.30	0.05	25	23.2–465	450
12	$MgCl_2$	A	1	0.30	0.05	25	23.2–465	718
13	$CaCl_2$	A	1	0.30	0.05	25	23.2–465	671
14	$NaHCO_3$	A	1	0.30	0.05	25	23.2–465	715
15	Na_2SO_4	A	1	0.30	0.05	25	23.2–465	729
16	NaCl	A	1	0.30	0.05	10	15.9–319	1222
17	NaCl	A	1	0.30	0.05	20	20.7–414	709
18	NaCl	A	1	0.30	0.05	30	25.9–518	444
19	NaCl	A	1	0.30	0.05	40	31.5–630	292

changing point of the resulting curve was regarded as the value of LCD, i.e., I_s (*2*, *4*, *5*, *16*). The constant electric current sources used were changeable types from 0.001 to 10.00 A (Takasago Seisakusho Ltd., Gp 0110-5 and Gp 0250-10).

RESULTS AND DISCUSSION

1. Relation between Re and Sh_s

The values of LCD were obtained for Runs 1, 2, 4, and 6 in Table 2. The relations between Re and Sh_s for several kinds of spacers are shown in Fig. 3.

This figure indicates that Sh_s increases in proportion to the ½ power of Re in each spacer. From the full lines, Eq. (17) is obtained for all spacers:

$$Sh_s \propto Re^{1/2} \tag{17}$$

Therefore, the value of b in Eq. (15) is ¼.

2. Effect of Kind of Spacer on LCD

Figure 3 shows that Sh_s is influenced by the kind of spacer. Spacer A is best, because the Sh_s of this spacer is the largest among the spacers employed.

The values of Sh_s calculated from the data (*2*, *3*, *9*) and the lines of Sh_s calculated from the equations (*5*, *8*, *9*) of other workers are also shown in Fig. 3. The values of LCD measured by Rosenberg et al. (*3*) and Winograd et al. (*9*) are nearly equal to those obtained for Spacer A. The values from Mandersloot et al. (*2*) are approximately the same as those for Spacer D.

$Sh_0(= HI_0/C_0DF)$, which represents LCD (I_0) in the equipment without a spacer (*16*), is expressed by the broken line in Fig. 3. This result demonstrates that the values of Sh_s are about 2–5 times those of Sh_0, and that some spacer should be inserted into the compartment to obtain a larger value of LCD.

3. Decision of Estimating Equation of LCD

Equation (15) is converted to Eq. (18) by substituting the value of b:

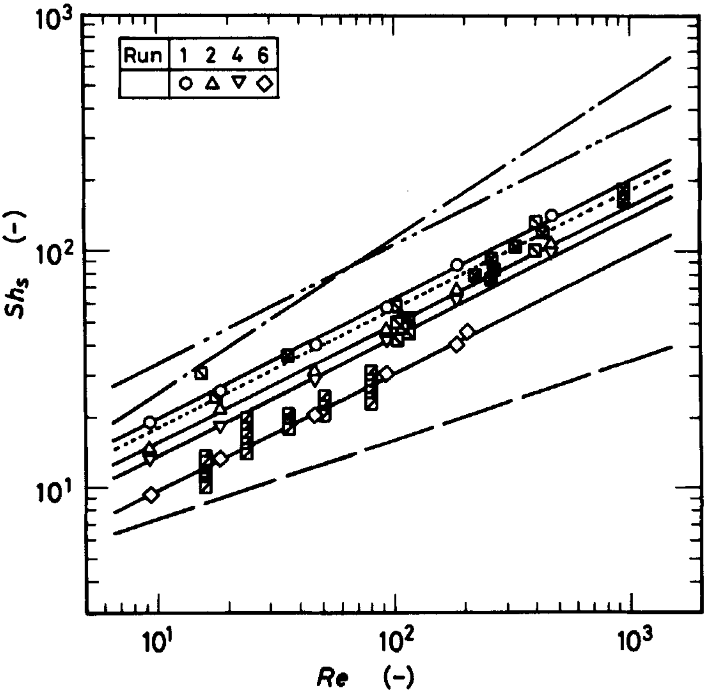

FIG. 3. Influence of Re on Sh_s for various kinds of spacers. Run numbers correspond to those indicated in Table 2. ⧄: Values calculated from the data of Mandersloot et al. (*2*). ⧅: Values calculated from the data of Rosenberg et al. (*3*). --⊠--: Values and line calculated from the results of Winograd et al. (*9*). —-—: Line calculated from the equation of Kitamoto et al. (*5*). —--—: Line calculated from the equation of Sonin et al. (*8*). ——: Line calculated from the equation of Miyoshi et al. for the equipment without spacer (*16*).

$$Q = \frac{\mathrm{Sh}_s(\bar{\tau} - \tau)}{(H/L)^{1/3}M^{1/3}\mathrm{Re}^{1/2}\mathrm{Sc}^{1/3}} \tag{18}$$

In order to calculate Sh_s from Eq. (15), the value of Q as defined by Eq. (16) has to be found. Because Q contains constant a, it will be influenced by the characteristics of the spacer, which are represented by d, t, and ε.

When the thickness of a spacer (t) is less than that of a frame of the compartment (H), a number of spacers (n) should be inserted to fill each compartment. The relationships between $n(t - d)$ and Q as calculated from Eq. (18) are shown in Fig. 4.

From this figure, the following relationship is given:

$$Q \propto [n(t - d)]^{-1/2} \tag{19}$$

The ion-exchange compartment is regarded as a kind of the packed bed, as mentioned before. Then, Q may also be influenced by the void fraction (ε), which is another characteristic of a spacer and which is employed for the study of packed beds. Hence, the relationship between

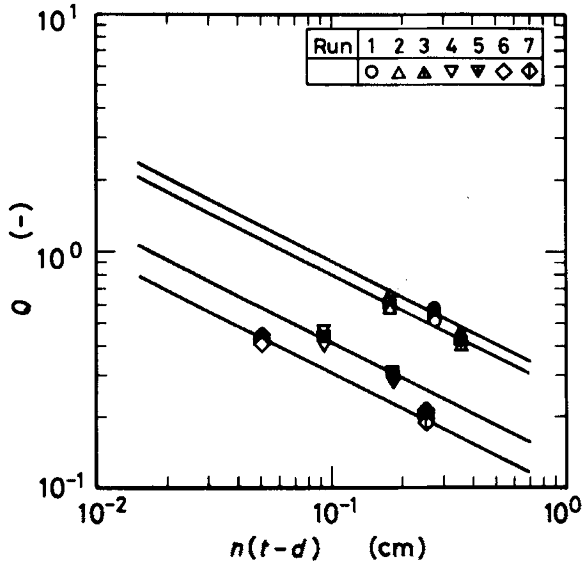

FIG. 4. Variation of Q with $n(t - d)$ for various spacers. Run numbers correspond to those indicated in Table 2. The full lines represent Eq. (19).

$Q[n(t-d)]^{1/2}$ and $(1-\varepsilon)^2/\varepsilon^3$, which was an important parameter in the Kozeny-Carman equation (*28*), was examined.

From the result shown in Fig. 5, the experimental equation to estimate Q is

$$Q = \frac{0.095}{[n(t-d)]^{1/2}[(1-\varepsilon)^2/\varepsilon^3]^{1/5}} \tag{20}$$

in which the dimension 0.095 is in $\mathrm{cm}^{1/2}$.

By substituting the values of a, i.e., Q and b, in Eq. (15), an equation for the estimation of LCD is derived:

$$\mathrm{Sh}_s = \frac{0.095(H/L)^{1/3}M^{1/3}}{(\bar{\tau}-\tau)[n(t-d)]^{1/2}[(1-\varepsilon)^2/\varepsilon^3]^{1/5}}\mathrm{Re}^{1/2}\mathrm{Sc}^{1/3} \tag{21}$$

Further, Eq. (2), which denotes the eddy diffusivity, is

$$D_e = \frac{0.0285D}{[n(t-d)]^{3/4}[(1-\varepsilon)^2/\varepsilon^3]^{3/10}}\mathrm{Re}^{1/4} \tag{22}$$

in which the dimension of 0.0285 is in $\mathrm{cm}^{3/4}$.

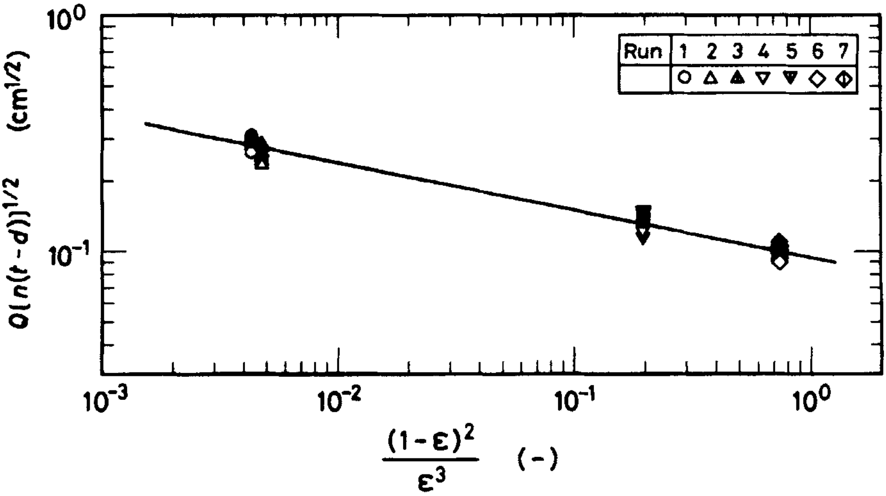

FIG. 5. Effect of $(1-\varepsilon)^2/\varepsilon^3$ on $Q[n(t-d)]^{1/2}$ for various spacers. Run numbers correspond to those indicated in Table 2. The full line represents Eq. (20).

4. Discussion on Eq. (21) for Estimating LCD

The relationship between Re, Sc, and Sh_s obtained was examined by employing Eq. (21) for Runs 1-19 in Table 2, where several kinds of spacers and electrolytes, and various values of C_0, T, and u_0 were used. The results are shown in Fig. 6. The full line in the figure represents Eq. (21).

It may be necessary to obtain the relationship between Re, Sc, and Sh_s over a wider range of Re and Sc, respectively, for greater accuracy. However, apparent agreement between experiment and theory has been obtained in the range of the present experimental conditions. Therefore, the estimating equation of LCD represented by Eq. (21) is valid here.

CONCLUSIONS

An equation useful for estimating LCD in electrodialytic equipment with spacers was derived as Eq. (15) by taking into account the eddy diffusivity caused by the spacers.

In order to determine the values of the empirical constants in Eq. (15), the values of LCD in the equipment were measured under the various conditions shown in Table 2.

The results demonstrate that the eddy diffusivity caused by spacers can be represented by Eq. (22). It is evident that LCD in the electrodialysis can be estimated from Eq. (21).

SYMBOLS

a, b	constants influenced by characteristics of spacer (−)
C	concentration of solution in ion-exchange compartment (g-eq/cm^3)
C_0	concentration of inlet solution (g-eq/cm^3)
C_w	concentration of solution on wall of the compartment (g-eq/cm^3)
D	molecular diffusivity (cm^2/s)
D_e	eddy diffusivity due to spacer (cm^2/s)
d	diameter of a fiber in spacer (cm)
F	Faraday's constant (C/g-eq)
H	channel thickness of the compartment (cm)
I	electric current density (A/cm^2)

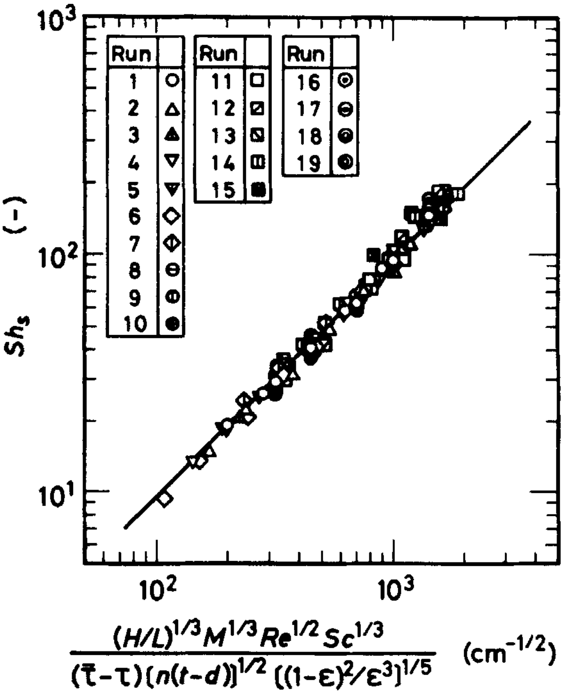

FIG. 6. Relation between Re, Sc, and Sh_s. Run numbers correspond to those indicated in Table 2. The full line represents Eq. (21).

I_s	limiting current density (LCD) in the electrodialytic equipment with spacers (A/cm^2)
I_0	LCD in the equipment without spacer (A/cm^2)
L	height of the compartment (cm)
M	variable influenced by m as defined by Eq. (7) ($-$)
m	coefficient of eddy viscosity due to spacer ($-$)
N_m	mean flux in ion-exchange membrane (g-eq/cm$^2 \cdot$ s)
N_s	mean flux in the solution (g-eq/cm$^2 \cdot$ s)
N_y	flux in the y direction (g-eq/cm$^2 \cdot$ s)
N_z	flux in the z direction (g-eq/cm$^2 \cdot$ s)
n	number of spacers in the compartment ($-$)
Q	variable ($-$)
Re	Reynolds number ($= Hu_0/\nu$) ($-$)
Sc	Schmidt number ($= \nu/D$) ($-$)
Sh$_s$	Sherwood number in the equipment with spacers ($= HI_s/C_0DF$) ($-$)
Sh$_0$	Sherwood number in the equipment without spacer ($= HI_0/C_0DF$) ($-$)
T	temperature (°C)
t	apparent thickness of a spacer (cm)
u	flow velocity (cm/s)
u_0	superficial flow velocity (cm/s)
V	electric potential (V)
y	horizontal axis in the compartment (cm)
z	vertical axis in the compartment (cm)

Greek Symbols

ε	void fraction of spacer ($-$)
ν	kinematic viscosity (cm^2/s)
τ	transport number of electrolyte in solution ($-$)
$\bar{\tau}$	transport number of ion in ion-exchange membrane ($-$)

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