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Enrichment Factor, Height of Separation Unit, and Separation Efficiency by Ion Exchange with Chemical Reaction

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ABSTRACT

The enrichment factor (ϵ_u) in the separation unit of ion-exchange and chemical reactions is a function of the enrichment factor of the chemical reaction ($\epsilon_{r,s}$) and the "equilibrium coefficients" (ζ) which are determined only by the distribution of the ions in both the solution and ion-exchange phases. The factor can be simply expressed as $\epsilon_u = \zeta\epsilon_{r,s}$. The height of the separation unit is the sum of heights due to ion exchange, chemical reaction, and flow pattern. The height is also due to the "kinetics coefficient" (ζ), which is a function of the distribution as well as the "equilibrium coefficient." The separation efficiency is proportional to $\zeta/\sqrt{\upsilon}$, both of which depend on the concentration of ions in the unit. Several schemes for the separation units are also discussed.

Key Words. Ion exchange; Chemical reaction; Separation unit; Enrichment factor; Height of separation unit; Separation efficiency; Equilibrium coefficient; Kinetics coefficient

INTRODUCTION

In the 1940s and the following decades, various new ion exchangers were synthesized from such petrochemical products as styrene and divinylbenzene. These new ion exchangers show higher performances than do older ion exchangers such as phenol–formaldehyde resin. Ion exchangers having a poly(styrene–divinylbenzene) backbone and a sulfonated benzene ring or tetremethylammonium chloride as the functional group have been applied to so many ion-exchange studies that researchers in this field know their characteristics without carefully reading their description in articles. The same circumstances apply in the field of solvent extraction.

The kinetics and the equilibrium theories and principles of the ion-exchange method for separation processes have been studied, developed, and successfully applied by Helfferich (1), Giddings (2), Gluekauf (3), Senou (4), and many other researchers. Al-Bazi wrote an excellent reviews on the separation of platinum elements by ion exchange and solvent extraction in 1984 (5). He cited more than 200 articles and books as references for his research review and focused on ion exchanges with such chemical reactions as complex formation. However, the names of Helfferich, Janauer, and Alexantroposs could not be found in the review in spite of the fact that they are known to be famous scholars in this field (6). This implies that there are too many reports on the separation of platinum metals by ion exchange and solvent extraction to judge whether an article is important or not.

On the other hand, the literature on inorganic ligand complexes of metal ions has not been organized although many data on the stability constants of metal ions have been reported (7). The enrichment factors, the exchange rates of complexes, and the chemical structures of complexes in ion exchangers, which play an important role in ion exchange phenomena, are fragmentary knowledge. Although one can obtain numerous articles on this interesting field through the Internet, the data and theory by which one can develop his research step by step is difficult to find.

In this article the enrichment factor and the height of the separation unit of ion exchange with chemical reaction are described in order to contribute to the orderly arrangement of the available data. It is theoretical treatments which are mainly described in this report. However, experimental data are also discussed in order to aid in comprehension of the equations.

SEPARATION UNIT OF ION EXCHANGE WITH CHEMICAL REACTION

Ion-exchange research has usually been done under the assumption that ion exchangers have selectivity for the target ions. That is, different ions can be separated by the ion-exchange method when the ion exchanger has high selectivity. Contrarywise, separation cannot be done by using an ion exchanger which has little selectivity for the target ions. In the latter situation, the researchers can do nothing but give up using separation by the ion-exchange process or to synthesize new polymers in order to obtain a "good" ion exchanger. Generally speaking, the synthesis of a new type of ion exchanger is very difficult even when the researcher can refer to similar works in the field of solvent extraction and other separation processes.

Ion exchange accompanied by a chemical reaction can be applied to separation processes, and it is one of the best methods to use when an ion exchanger has no selectivity for specific ions. An appropriate chemical reaction for separation should be selected from among such chemical reactions as acid–base, redox, and complex formation.

An ion-exchange separation unit without chemical reactions is shown in Scheme 1. Suppose that A_1 and A_2 dissolve in the solution phase of the separation unit, and A_2 adsorbs onto the ion exchanger more strongly than does A_1 because the ion exchanger has a stronger affinity to A_2.

The unit where the chemical reaction coexists in the separation unit is shown in Scheme 2. The coexisting chemical reaction is a reverse reaction in most cases, so it is expressed as

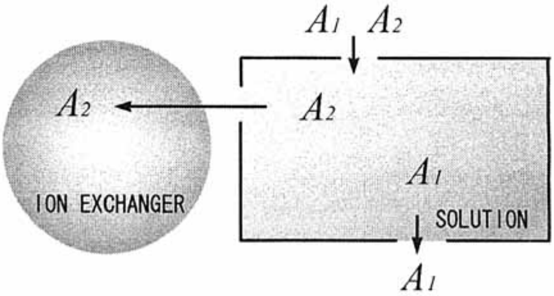
$$A + nX = D \tag{1}$$

where A is an acceptor, D is a donor, and X is an "addend" (8). For example, A is ammonia and X is a proton when the chemical reaction is an acid–base reaction for nitrogen isotope separation.

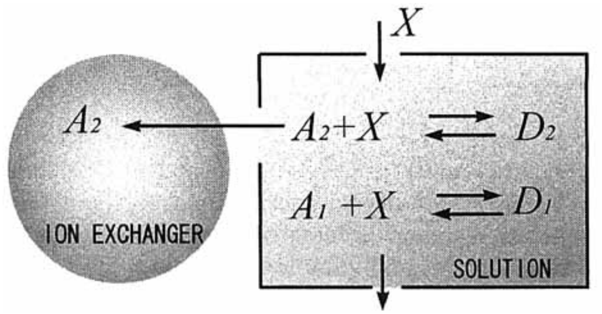
$$NH_3 + H^+ = NH_4^+ \tag{2}$$

A is lanthanide and X is inorganic ions or organic compounds when the reaction is complex formation for lanthanide elements separation:

$$Nd^{3+} + EDTA^{4-} = Nd(EDTA)^- \tag{3}$$



SCHEME 1 Separation unit of ion exchange.



SCHEME 2 Separation unit of ion exchange with chemical reaction.

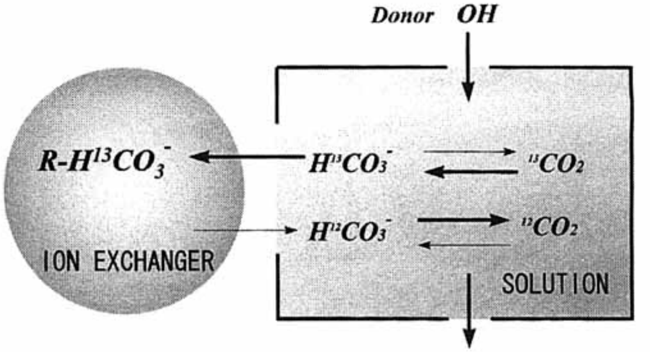
A is uranium and X is an electron when the reaction is a redox reaction for uranium-235 enrichment:

$$U^{6+} + 2e^{-} = U^{4+} \tag{4}$$

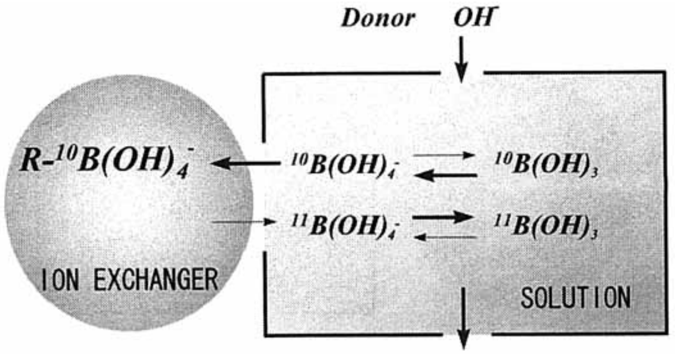
The addend (X) in Eq. (1) represents the general symbol of these reactions because many kinds of chemical reactions can be applied.

Assuming that the ion exchanger has no selectivity to any A_i, a similar amount of A_1 and A_2 ions might be found in the ion exchanger. On the contrary, only A_2 is adsorbed onto the ion exchanger when 1) the affinity of the addend X to A_1 is larger than the affinity to A_2, and 2) the ion exchanger adsorbs any A_i more strongly than any D_i. A_2 and D_1 are major components on the ion exchanger and in the solution phase, respectively, after equilibrium.

Several examples of the separation unit of ion exchange with chemical reactions are shown in the six parts of Scheme 3. The addend (X) is an



SCHEME 3a Separation unit of carbon isotopes with acid–base reaction.

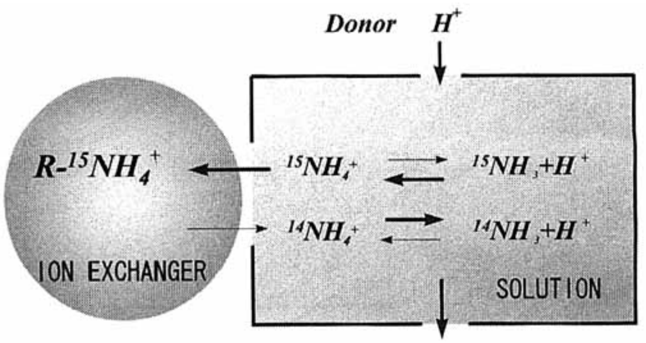


SCHEME 3b Separation unit of carbon isotopes with acid–base reaction.

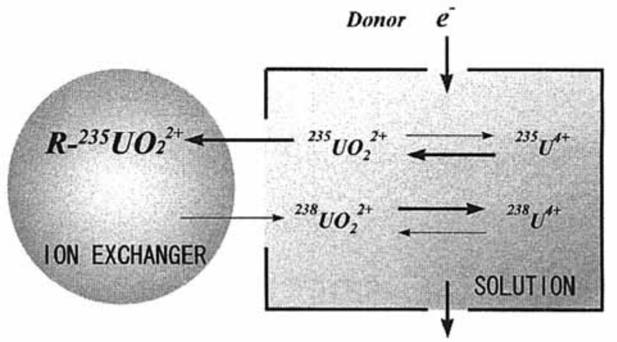
electron in this case. Carbon isotopes separation is shown in Scheme 3a. As the pK_b of carbon-13 is higher than that of carbon-12, the concentration of $H^{13}CO_3^-$ is higher than that of $H^{12}CO_3^-$ in the solution. The ion exchanger has no selectivity to carbon isotopes (9). Schemes 3b and 3c are the separation units of boron isotopes and nitrogen isotopes. The addends are protons in both cases.

Uranium isotopes separation by the ion-exchange method is shown in Scheme 3d, where uranium-235 tends to be oxidized more easily than uranium-238, and U(VI), the oxidized species, adsorbs more strongly on the ion exchanger than does U(IV), the reduced species. Uranium can be separated by ion exchange although the ion exchanger has no selectivity to uranium isotopes (10).

One of the famous cases of ion exchange with chemical reaction is shown in Scheme 3-e. Pr and Nd form complex ions with EDTA (ethylene-



SCHEME 3c Separation unit of carbon isotopes with acid–base reaction.

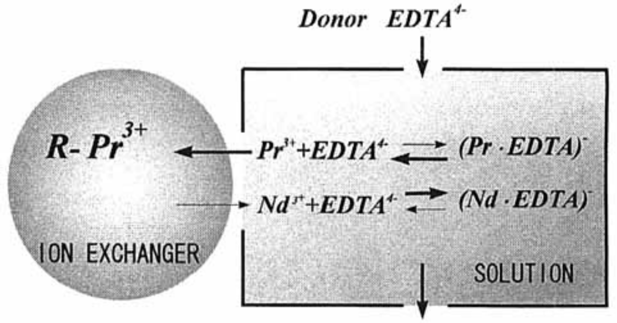


SCHEME 3d Separation unit of uranium isotopes with redox reaction.

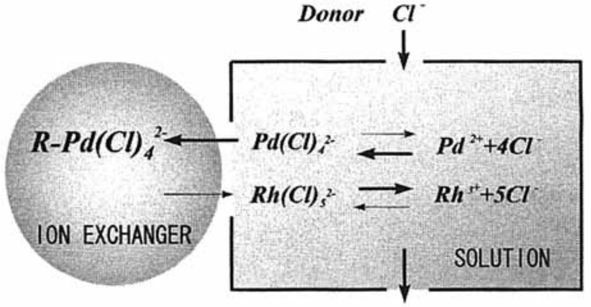
diamine-tetraacetic acid) and convert to anions: (Pr(EDTA^{4-})$^-$ and Nd(EDTA^{4-})$^-$. Nd^{3+} is rich in the solution. The concentration of Nd(EDTA^{4-})$^-$ is higher than that of Pr(EDTA^{4-})$^-$ because Nd^{3+} has stronger affinity to EDTA^{4-} than does Pr^{3+}. The last scheme (Scheme 3f) shows the separation of platinum metals (which the authors have studied). In every case above, the ion exchanger cannot separate the isotopes, isomers, and elements, but they can be separated by the aid of chemical reactions.

Strictly speaking, the chemical reaction which makes it possible to separate the target ions works both in the solution and the ion exchanger phase. The separation unit can be explicitly depicted as in Scheme 4.

The equilibrium cubic has six faces, and one face corresponds to one equilibrium reaction. Scheme 4 can be written like Scheme 2, if the reac-



SCHEME 3e Separation unit of rare earth elements with complexation.



SCHEME 3f Separation unit of platinum group elements with complexation.

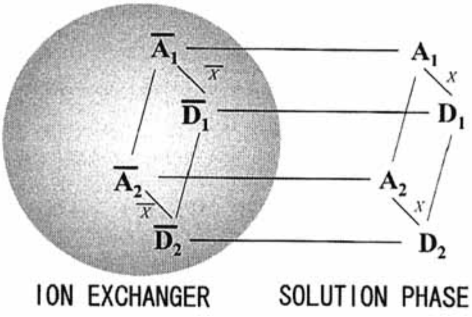
tion rate of the chemical reaction in the solution phase is faster or the concentration of A_2 and D_2 are much higher than A_1 and D_1 in the ion exchanger. The separation equilibria in the separation unit can be expressed as follows,

$$^2D_s + {}^2A_a \leftrightarrow {}^2D_a + {}^2A_s$$

$$K_{a,2} = \frac{(^2D_a)/(^2D_s)}{(^2A_a)/(^2A_s)} \tag{5}$$

$$^1D_s + {}^1A_a \leftrightarrow {}^1D_a + {}^1A_s$$

$$K_{a,1} = \frac{(^1D_a)/(^1D_s)}{(^1A_a)/(^1A_s)} \tag{6}$$



SCHEME 4 Explicit model of separation unit.

$^2A_s + {}^1A_a \leftrightarrow {}^2A_a + {}^1A_s$

$$K_{a,A} = \frac{(^2A_a)/(^1A_a)}{(^1A_s)/(^2A_s)} \tag{7}$$

$$K_{a,D} = \frac{(^2D_a)/(^1D_a)}{(^1D_s)/(^2D_s)} \tag{8}$$

$^1D_s + {}^2A_s \leftrightarrow {}^2D_s + {}^1A_s$

$$K_{r,s} = \frac{(^2D_s)/(^1D_s)}{(^1A_s)/(^2A_s)} \tag{9}$$

$^1D_a + {}^2A_a \leftrightarrow {}^2D_a + {}^1A_a$

$$K_{r,a} = \frac{(^2D_a)/(^1D_a)}{(^1A_a)/(^2A_a)} \tag{10}$$

In the above equations the chemical species in the solution are indicated by the subscript "s" and those on the ion exchanger are indicated by the subscript "a." Subscript "a" of equilibrium constants (K) means ion exchange and "r" means chemical reactions in the solution phase. The enrichment factor ($\epsilon = K - 1$) of the separation unit (ϵ_u) cannot be directly solved by the six equations. However, when aid by the chemical reaction is required, the selectivity to the target ions the ion exchanger is so small that $K_{a,A}$ and $K_{a,D}$ can be regarded as 1.0. $K_{a,2}$ and $K_{a,1}$ are much larger than $K_{a,A}$ and $K_{a,D}$. $K_{r,a}$ has the same value as $K_{r,s}$ if the mechanism of the chemical reaction in ion exchange can be regarded as the same as in the solution. Under these assumptions the six equilibrium constants can be arranged in the form of enrichment factors as follows:

$$\epsilon_u = \frac{\left(g(1 + \epsilon_{r,s})\dfrac{^TA_a}{^TD_a} + 1\right)\left(g\dfrac{^TA_s}{^TD_s} + 1\right){}^2D_a{}^1D_s}{\left(g(1 + \epsilon_{r,s})\dfrac{^TA_s}{^TD_s} + 1\right)\left(g\dfrac{^TA_a}{^TD_a} + 1\right){}^2D_s{}^1D_a} - 1 \tag{11}$$

$$= \frac{g(^TA_a{}^TD_s - {}^TA_s{}^TD_a)}{(g{}^TA_a + {}^TD_a)(g(1 + \epsilon_{r,s}){}^TA_s + {}^TD_s)}\,\epsilon_{r,s} \tag{12}$$

where the superscript "T" means the total amount of target ions. g is defined by (11, 12)

$$g = [1 - (1 + \epsilon_{r,s})(^2D_s/^1D_s)]/[1 - (^2D_s/^1D_s)]$$

The enrichment factor of the separation unit can be simplified to obtain Eq. (13):

$$\epsilon_u = \zeta\epsilon_{r,s} \tag{13}$$

where

$$\zeta = \frac{{}^{T}A_a{}^{T}D_s - {}^{T}A_s{}^{T}D_a}{({}^{T}A_a + {}^{T}D_a)({}^{T}A_s + {}^{T}D_s)} \tag{14}$$

Equation (13) is one of the most interesting equations in this article. The equation is very simple, and it shows that the enrichment factor of the chemical reaction multiplied by factor ζ is the enrichment factor of the separation unit. The coefficient ζ, called the "equilibrium coefficient," is the function of only the concentrations of the ions in the separation unit. It shows that the enrichment factor can be decided only by the distribution of the target ions and the enrichment factor of the chemical reaction. The equilibrium coefficient decreases when the fraction of A_1 which exists in the solution phase and the fraction of D_2 in the ion exchanger are high. The magnitude of ϵ_u, compared with that of ϵ_r, indicates how efficiently the fundamental separation equilibrium is transferred to the separation equilibrium between the solid and liquid phases.

The experimental data are listed in Fig. 1 for the case of uranium isotopes separation by using an ion exchanger with a redox reaction. There are three chemical reactions for uranium isotopes separation. The addends were electrons, protons, and chloride. The coefficient ζ could be easily controlled by changing the factors which gives the coefficient influence,

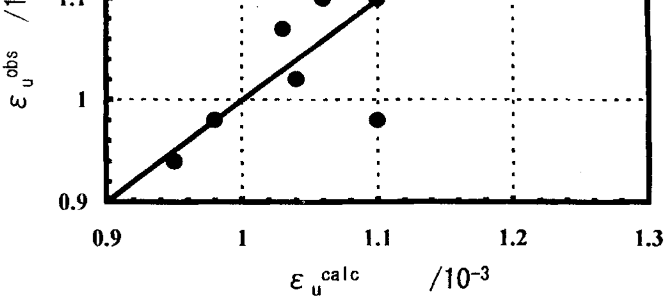


FIG. 1 Comparison of ϵ_u calculated from ϵ_u observed.

TABLE 1
Representative Data of Enrichment Factors in the Case of Uranium Enrichment by Ion Exchange with Redox Reaction

Expt	H^+ (mol/L)	Cl^- (mol/L)	A_a (mol/L)	A_s (mol/L)	D_a (mol/L)	D_s (mol/L)	ζ	$\epsilon_{cal} \times 10^{-3}$	$\epsilon_{obs} \times 10^{-3}$
436	0.8	0.9	248	47	40	310	0.73	0.73	0.72
380	0.7	1.5	125	18	10	124	0.8	0.8	0.78
432	0.7	1.3	199	16	3	148	0.89	0.89	0.94

for example, the concentrations of redox agents and ligands, the temperature, and the ion exchanger. Typical examples obtained from the experiments for uranium enrichment are listed in Table 1.

The equilibrium coefficients and ϵ_u were calculated by using the concentration observed, and the calculated ϵ_us were in good agreement with the observed ϵ_us.

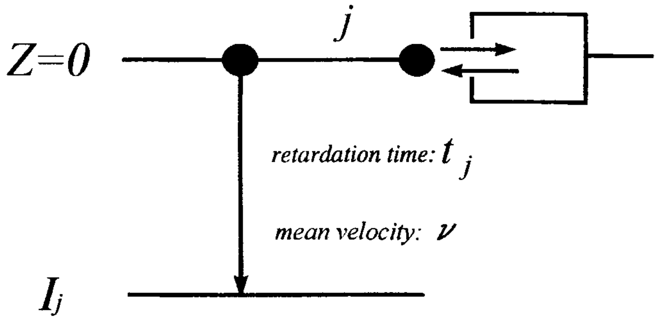
HEIGHT OF SEPARATION UNIT

The influence of the height of the separation unit in an adsorption process has been studied by many scholars and engineers. Major methods include direct analysis of the results of separation such as the plate height in the distillation process. The same method was applied to an ion-exchange process where the height (HETP or other similar expressions) can be easily calculated. However, this procedure does not give information about the separation mechanism. An excellent book by Giddings made it clear that the height could be divided into several terms derived from molecular movements. Helfferich also studied the two rate-determining process in ion exchange rate. This article models the height after Giddings (2).

The height of ion exchange with chemical reactions depends on such kinetics constants as the diffusion constants in the solution and ion exchanger, particle diameter in the ion exchanger, kinetic constants of chemical reactions, and nonuniform flow of solutions. A kinetic model of the separation unit is shown in Scheme 5a.

An expression simpler than the conventional definition will be proposed in order to understand the separation unit of ion exchange with chemical reaction more inherently.

The height can be defined as the length of the path between two molecules. Suppose that molecule 1 flows straightforwardly without any reac-



SCHEME 5a General kinetics model of separation unit.

tions and collisions to ion exchanger particles and other materials. Molecule 2 reacts chemically, including reaction with addends in solution or the ion exchanger. Molecule 2 is delayed by the reaction compared molecule 1. If the reaction takes time t_j, the length (M_j) is

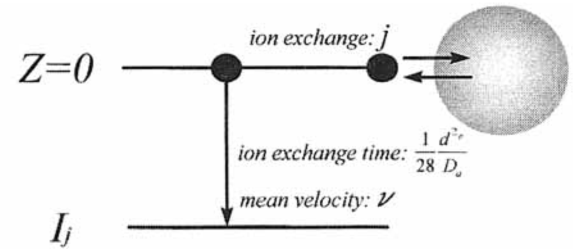
$$M_j = vt_j, \tag{15}$$

where v is the migration velocity of the adsorption band. The mole fraction of molecule 2 is

$$\xi_j \equiv N_j/N_t \tag{16}$$

where ξ_j is called the "kinetics localization coefficient."

In the case of an ion-exchange reaction, for example, molecule 2 is delayed due to the ion-exchange reaction as shown in Scheme 5b. The definition is essentially the same as the definition $H = \sigma^2/L$, where L means the length of the development.



SCHEME 5b Model kinitics scheme of ion exchange separation unit.

The height due to ion exchange can be written by using the diffusion constant of ions in the ion exchanger (D_a), the particle diameter (d_p), the mole fraction of ions adsorbed (X_a), and the migration velocity (v) as listed in Table 1. The height can be easily obtained from the average exchange by using integral. The height which originates in the chemical reaction is also listed in Table 2, and it depends on the reaction rate constant (k) and the fraction of the molecule adsorbed in the entire molecule (X_s) when the chemical reaction is regarded as a first-order reaction.

The height is a function of such kinetic constants as the diffusion constant and the rate constant. When the interaction of ion exchange and the chemical reaction is so small that it can be disregarded, the height of the separation unit can be written as

$$H_T = H_a + H_r \tag{17}$$

Therefore, the height is

$$H_T = \left[\frac{1}{28}({}^{T}A_a + {}^{T}D_a)\frac{d_p^2}{D_a} + ({}^{T}A_s + {}^{T}D_s)\frac{1}{k}\right] v \tag{18}$$

When the two reactions proceed in parallel, the height is expressed as

$$\frac{1}{H_T} = \frac{1}{H_a} + \frac{1}{H_r} \tag{19}$$

Actually, the total height of the separation unit is composed of a nonuniform flow term as well as ion exchange and chemical reaction terms. Two different heights have to be considered. One is due to the micro-nonuniform flow (H_{mic}) and the other due to macro-nonuniform flow (H_{mac}). The detailed descriptions appear previous works, including our reports (13, 14). When the four terms are in series, the total height is

$$H_T = H_a + H_r + H_{mic} + H_{mac} \tag{20}$$

SEPARATION EFFICIENCY

The fundamental equation which dominates the separation system is

$$\left(\frac{\partial C}{\partial t}\right)_x = -v\left(\frac{\partial C}{\partial x}\right)_t + D\left(\frac{\partial^2 C}{\partial x^2}\right)_t \tag{21}$$

where v is the mean velocity of target ions and D is the overall diffusion constant. Several answers to the equation have been given. For example, the answer in the case of displacement chromatography using ion exchange is (15, 16)

$$\frac{C_2/C_1}{C_{2,0}/C_{1,0}} = \frac{2\epsilon_q N}{1 - \exp(-2\epsilon_q N)} \exp(-2\epsilon_q i) - \frac{4\epsilon_q}{N}$$

$$\cdot\exp(-\epsilon_q i) \sum_{m=1}^{\infty} \frac{n\{1 - (-1)^n \exp(\epsilon_q N)\}}{(\epsilon_q^2 + n^2)^2} \tag{22}$$

$$\cdot\{n \cos(ni) - \epsilon_q \sin(ni)\}\cdot\exp\left(-\frac{\epsilon_q^2 + n^2}{2(1 + \epsilon_q)} j\right)$$

The equation is so complicated that one can hardly understand the physical meaning of the answer.

We can understand the meaning of the following approximate equation for separation by ion exchange. The separation efficiency is approximately determined by using α (the separation factor during a certain time), which is a function of the separation factor and the height of the separation unit (17, 18).

$$\alpha = \frac{C_2/C_1}{C_2^0/C_1^0} = 160\sqrt{L}\frac{\epsilon}{\sqrt{H}} = \frac{\zeta}{\sqrt{\xi}} \tag{23}$$

where C is the concentration of ions, superscript "0" means C at $t = 0$, and L is the migration length of the adsorption band. If such kinetic constants as the diffusion constant in the ion exchanger and the reaction rate of the chemical reaction in solution are constant, the separation efficiency is greatly affected by the distribution of ions in the exchanger and the solution.

As shown in Eq. (14), the enrichment factor of the separation unit depends on the "equilibrium coefficient" and the height of the unit depend on "kinetics coefficients" as shown in Table 2. The natural result is that the separation efficiency strongly depends on the distribution of the ion

TABLE 2
Height of Separation Unit Due to Ion Exchange and Chemical Reaction

Term	General model	Ion exchange	Chemical reaction
t_i	t_i	$\frac{d_p^2}{28D_a}$	$\frac{1}{k}$
ξ_i	N_i/N_t	a	$A_s + D_s$
H_i	$\xi_i M_i$	$\frac{d_p^2 v (A_a + D_a)}{28D_a}$	$\frac{v (A_s + D_s)}{k}$

concentration in the separation unit because the efficiency is proportional to $\epsilon/\sqrt{H}$.

The rate-determining step is the chemical reaction in solution, $\epsilon/\sqrt{H}$, can be written as

$$\frac{\epsilon}{\sqrt{H_a}} \approx \frac{\zeta}{\sqrt{\xi}} = \frac{(A_aD_s - A_sD_a)}{\sqrt{(A_a + D_a)^3(A_s + D_s)^2}} \tag{24}$$

and when the ion-exchange reaction is the rate-determining step,

$$\frac{\epsilon}{\sqrt{H_r}} \approx \frac{\zeta}{\sqrt{\xi}} = \frac{(A_aD_s - A_sD_a)}{\sqrt{(A_a + D_a)^2(A_s + D_s)^3}} . \tag{25}$$

Experimental data which can be compared to values calculated by the above equations were difficult to obtain. The diffusion constant in the ion exchanger and the rate constant of the chemical reaction depend on the concentration of ions as well as the dependence of the localization coefficients on the concentration. The effect of Eqs. (24) and (25) can be demonstrated only when we can find the conditions where the physical constants do not change while the localization coefficients vary. The data are shown for the case of uranium isotopes separation by ion exchange with a redox reaction in Fig. 2.

As seen in Scheme 4, there are two exchange routes between A_2 in the resin and D_1 in the solution. If the concentration of A_2 in the solution is high, the main exchange route is A_2 (in the ion exchanger)–A_2 (in the solution)–D_1 (in the solution), and the kinetics coefficient is large enough to exchange A_2 and D_1. However, the enrichment factor is reduced because the higher concentration of A_2 is a major factor in reduction. That

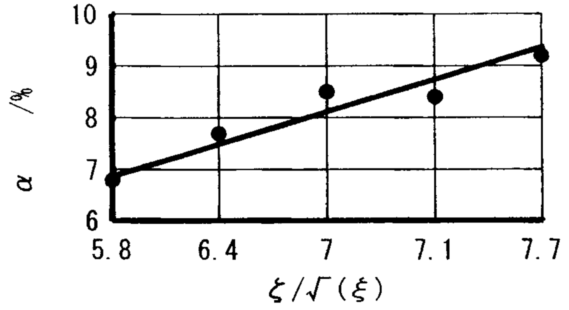


FIG. 2 Separation efficiency as a function of localization coefficients.

is, A_2 in the solution uses the required route from A_2 in the ion exchanger to D_1 in the solution, but it reduces the equilibrium coefficient. For the second route from A_2 in the ion exchanger to D_1 in the solution, D_1 in the ion exchanger plays the same role as does A_2 in the solution in the first route.

The larger the equilibrium coefficient, the smaller the kinetics coefficient, and vice versa. This is unavoidable as long as the above two routes are considered for the case of ion-exchange separation with chemical reaction. If the distributions of ions in the ion-exchange column can be measured and are reported, the enrichment factor and the kinetics effect of distribution on the separation efficiency and the phenomena of ion-exchange separation with chemical reactions can be organized more easily, and thus reduce the number of trial and error tests in the field of ion-exchange separation.

CONCLUSION

The distribution of ions in the separation unit plays a important role as well as do the kinetics characteristics, such as the diffusion constant and rate constants, in the ion-exchange separation process with a chemical reaction (19). A higher separation efficiency can be achieved if the best distribution of the concentrations of ions as well as the ion exchanger, the chemical reaction in the solution phase, and some other condition is selected.

ACKNOWLEDGMENT

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