

STAGE EFFICIENCY OF MIXER-SETTLER EXTRACTION COLUMN

KATSUROKU TAKAHASHI*, SUSUMU NII,
KEIJI NAKANISHI AND HIROSHI TAKEUCHI
*Department of Chemical Engineering, Nagoya University,
Nagoya 464-01*

Key Words: Extraction, Extraction Column, Multistage Column, Stage Efficiency, HTU, Iodine, Coalescence

Stage efficiency of a mixer-settler extraction column which could realize a larger throughput at a higher agitation speed was measured with a three-stage column for the extraction of iodine from aqueous phase into heptane. The efficiency increased monotonously with agitation speed; *i.e.*, high-stage efficiency could be obtained as well as a large throughput at a high agitation speed with the extraction column. The stage efficiency based on the dispersed-phase concentration varied with the ratio of slopes between the equilibrium line and the operating line. The overall height of a transfer unit transformed from the stage efficiency was divided into the heights of a transfer unit of the dispersed and the continuous phases, which were correlated with the agitation speed.

Introduction

Multistage countercurrent extraction is useful for the separation of liquid mixtures, and an extraction column of high performance which achieves large throughput and a high stage efficiency is desirable. Mechanical agitation can promote stage efficiency due to the large interfacial area with small dispersed drops. As the drop size decreases with agitation speed, however, the relative velocity between the dispersed drops and the continuous phase decreases, which makes the throughput small⁵⁾. Moreover, axial mixing, which lowers the stage efficiency, becomes significant at high agitation speed. To increase the relative velocity between phases, it is desirable that the small drops be coalesced in the section between stages. Scheibel⁶⁾ set up the mixing part and the packing part alternately to promote drop coalescence in the packing part. For the same purpose, Steiner *et al.*⁷⁾ used a three-dimensional lattice as a partition of the mixing stages. In the case of aqueous dispersion, Bails-Stitt¹⁾ used an electrostatic coalescence to obtain stable operation under vigorous agitation. The division of flow channels into a dispersed phase channel and a continuous phase channel is also useful to obtain large throughput as an effective utilization of buoyancy. Gaubinger *et al.*²⁾ achieved a large throughput and small axial mixing by dividing the flow channels with concentric cones. The section in which the drops are coalesced is a sort of settling part, and acts as a suppresser of axial mixing. Horvath-Hartland⁴⁾ realized a high stage efficiency with a mixer-settler extraction column in which the interstage mixing was extremely small, while the throughput of the column was also small.

In the previous paper⁹⁾, the maximum throughput was measured for a mixer-settler column (MS column), which had a drop coalescer between each mixer and set-

tlar, divided flow channels and a lifter-turbine impeller in the mixer. It was indicated that the drop coalescer was useful for stable operation, especially at a large flow rate of dispersed phase, and that a large throughput was realized by the suction pressure induced with the lifter-turbine impeller as well as by the division of flow channels. In the present study, the stage efficiency of the MS column is measured with iodine extraction from the aqueous phase into heptane, and the effects of the agitation speed, the flow ratio of the dispersed phase to the continuous phase and the distribution ratio of iodine between phases on the stage efficiency or the height of a transfer unit is studied.

1. Experimental

An experimental flow sheet with a MS column is shown in Fig. 1. Details of the column were given in the previous paper⁹⁾. The extraction column consists of three stages and a bottom-drop coalescer of nonwoven material, and the inside diameter of the column is 60 mm. Each stage is partitioned into an upper settler of 40 mm height and a lower mixer of 50 mm height. A 6-blade lifter-turbine of 30 mm in diameter is used for agitation in the mixer and a three-dimensional lattice-drop coalescer is set on the stator ring of 30 mm in opening diameter between the mixer and the settler. The lattice coalescer, which is made of glass fiber mesh coated with PTFE, is 10 mm in height and 2.5 × 2.3 mm rectangular pitch.

An aqueous solution (continuous phase) fed to the mixer of the top stage rises through the coalescer into the settler with the dispersed phase, goes down through two downspouts located at opposite sides of the column into the lower-stage mixer after settling into two phases and finally is led to the leveller from the bottom of the

* Received June 3, 1993. Correspondence concerning this article should be addressed to K. Takahashi.

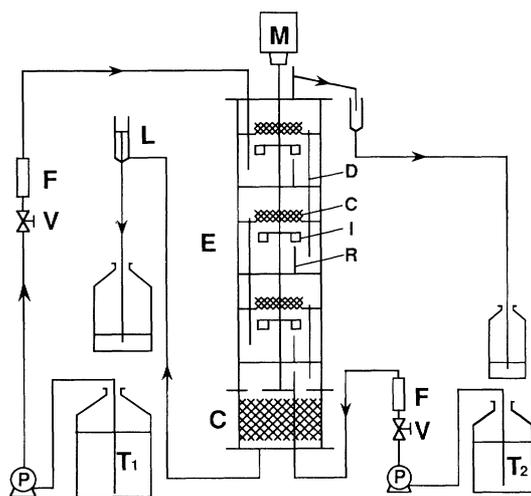


Fig. 1 Experimental apparatus. *E*: extraction column, *C*: drop coalescer, *T*₁: tank for aqueous phase, *T*₂: tank for organic phase, *I*: 6-blade lifter-turbine impeller, *D*: downspout for aqueous phase, *R*: riser for organic phase, *F*: flow meter, *P*: pump, *V*: valve, *L*: leveller

column. An organic solution (dispersed phase) fed at the bottom of the column rises from the mixer into the settler with the continuous phase, from the settler into the upper-stage mixer through a riser and overflows from the top of the column. The downspout is a glass tube of 5.5 mm inside diameter and the riser is a PTFE tube of 4 mm inside diameter. The continuous phase and the dispersed phase flow countercurrently as a whole.

Stage efficiencies were measured by experiments of iodine extraction from an aqueous solution of I₂-KI into the dispersed phase of *n*-heptane. After the column was filled with the aqueous solution, iodine-free heptane was fed and the outlet aqueous solution and the outlet organic solution were taken at intervals of 5 minutes. The iodine concentration of these samples were measured by a spectrophotometer. Steady state was achieved after flowing solutions of over three times the column volume when the flow ratio of aqueous phase to organic phase was 4. Since the distribution ratio of iodine for the present system varies with concentration of iodic ion in the aqueous solution⁸⁾, aqueous solutions of various KI concentrations were used in the experiment to change the distribution ratio. The distribution ratio for each experimental run was determined by measuring the iodine concentrations of the dispersed and the continuous phases after equilibrating the two phases.

Stage efficiencies were measured with a MIXCO column for the comparison with the MS column. The same column shell as with the MS column was used, and the column was divided into five stages of 60 mm in height by stator rings of 31 mm in opening diameter. The top and the bottom stages were used as settlers, and a 6-blade turbine impeller of 30 mm diameter was set at the center of each stage for three stages. The time to reach steady state was about twice that for the MS column.

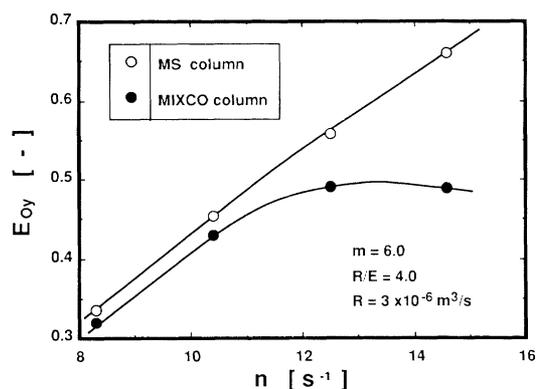


Fig. 2 Comparison of E_{Oy} between MS column and MIXCO column

2. Results and Discussion

2.1 Stage efficiency

Liquid-liquid equilibrium for the extraction of iodine from aqueous phase into heptane is given by

$$y = mx \quad (1)$$

where y and x are the iodine concentrations in heptane and aqueous phase, respectively, and m is the distribution ratio of iodine, which is expressed by $m = 36.6/(1 + 748 [I^-]^8)$ where $[I^-]$ is iodic ion concentration in kmol/m³. Since mutual solubilities between heptane and water are very small, the operating line for the multistage countercurrent extraction is expressed by

$$y_n - y_{in} = (R/E)(x_{n+1} - x_{out}) \quad (2)$$

where R is the raffinate phase flow rate, E the extract phase flow rate, y_{in} the iodine concentration in organic phase fed to the bottom of the column, and x_{out} the iodine concentration in aqueous phase from the bottom of the column. The stage number is counted from the top of the column. The stage efficiency E_{Oy} based on the concentration of organic phase is defined as follows.

$$E_{Oy} = (y_n - y_{n-1}) / (y_n^* - y_{n-1}) \quad (3)$$

where y_n^* ($= mx_n$) is the organic phase concentration in equilibrium with the aqueous phase of the n -th stage. Under the assumption that stage efficiency does not vary with the stage number, the following relation is derived for N -stage column from Eqs. (1) to (3) as shown in **Appendix**.

$$E_{Oy}(mx_{out} - y_{in})(1 + r_y + r_y^2 + \dots + r_y^{N-1}) - (y_{out} - y_{in}) = 0 \quad (4)$$

where

$$r_y = E_{Oy} \{ m / (R/E) - 1 \} + 1 \quad (5)$$

The flow ratio is given by $R/E = (y_{out} - y_{in}) / (x_{in} - x_{out})$. By using the measured values of x_{in} , x_{out} , y_{in} , y_{out} and m , E_{Oy} can be determined from Eq. (4).

Stage efficiencies E_{Oy} of the MS column and the MIXCO column are shown in **Fig. 2** against the agitation

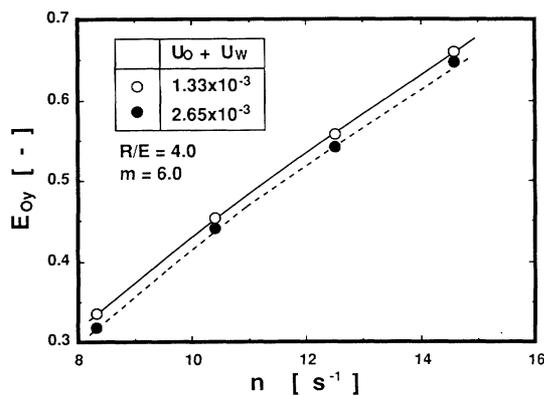


Fig. 3 Effect of throughput on stage efficiency

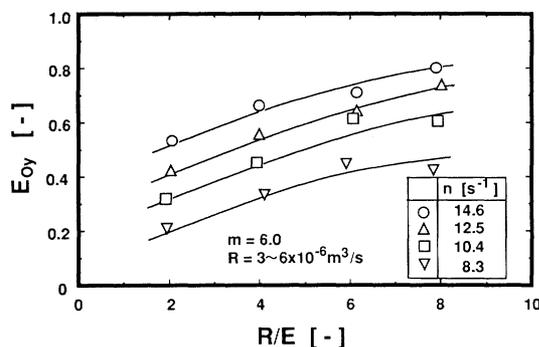


Fig. 4 Effect of flow ratio on stage efficiency

speed n for the distribution ratio $m = 6.0$ and flow ratio $R/E = 4$. E_{Oy} of MS column increased monotonously with increase in n , and a high stage efficiency could be obtained with strong agitation under the present experimental conditions. E_{Oy} of the MIXCO column stopped increasing at large values of n , and the values were smaller than those of the MS column in spite of the fact that the holdup of dispersed phase (*i.e.*, the interfacial area) of the MIXCO column was larger than those of MS column. The stage efficiency of the MIXCO column might be affected by the axial mixing between stages. Since the axial mixing increased with agitation speed, the difference in E_{Oy} between the two columns became large with n . The MS column may be a high-performance extraction column, which can achieve a high stage efficiency as well as a high throughput and stable operation at vigorous agitation. However, if the agitation speed continues to increase, a large part of the dispersed drops will pass through the coalescer without coalescing and be accompanied by the continuous phase, and a decrease in stage efficiency follows. Though smaller drops can be coalesced with a coalescer of smaller mesh pitch, the pressure drop in the coalescer increases with the decrease in mesh pitch, which decreases the throughput⁹.

The effect of throughput on stage efficiency is given in Fig. 3 for a given R/E and m . U_O and U_W are superficial velocities of the dispersed and continuous phases respectively, and $U_O + U_W = 1.33 \times 10^{-3}$ m/s corresponds to $R + E = 3.75 \times 10^{-6}$ m³/s for the present

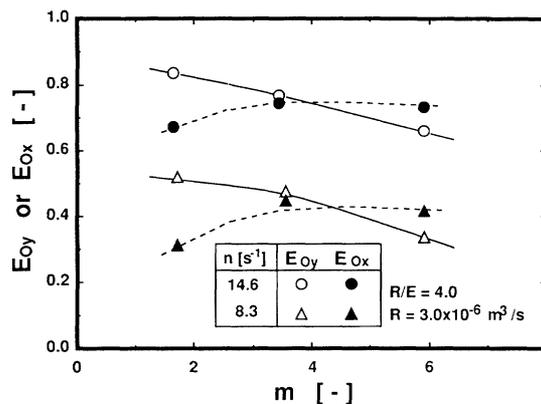


Fig. 5 Effect of distribution ratio on stage efficiency

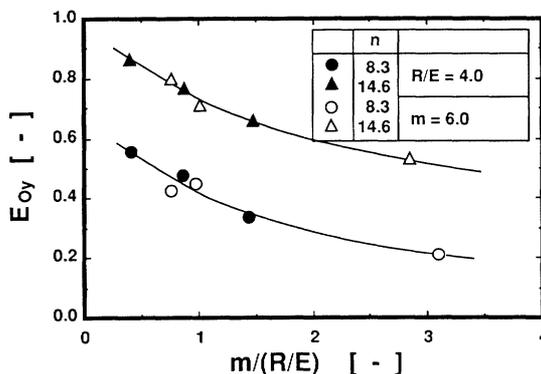


Fig. 6 Effects of distribution ratio and flow ratio on stage efficiency

column. E_{Oy} varied little with total throughput $U_O + U_W$. The holdup of dispersed phase increases with increase in U_O and decreases with increase in U_W , and the effect of U_O is larger than that of U_W ¹⁰, *i.e.*, the holdup may increase by doubling $U_O + U_W$ at constant R/E . The residence time of dispersed phase decreases with increase in U_W and the size of dispersed drop increases with decrease in residence time¹¹. As effects of the flow rate on stage efficiency, the contribution of the holdup is positive and that of the residence time is negative, for the interfacial area is proportional to the holdup and inversely proportional to the drop size. The former may be compensated by the latter in the case of Fig. 3.

On the other hand, E_{Oy} for a given m increased with the flow ratio R/E , and that for a given R/E decreased with the distribution ratio m as shown in Fig. 4 and 5. These data are replotted against $m/(R/E)$ in Fig. 6. Points for a given agitation speed are on one curve for both variations in R/E and n , *i.e.*, the stage efficiency, depends on the ratio of the slopes between the operating line and the equilibrium line.

When the stage efficiency E_{Ox} is defined by the following equation, using the concentration of continuous phase, the dependency of the stage efficiency on m and R/E may be different from the above results.

$$E_{Ox} = (x_n - x_{n+1}) / (x_n^* - x_{n+1}) \quad (6)$$

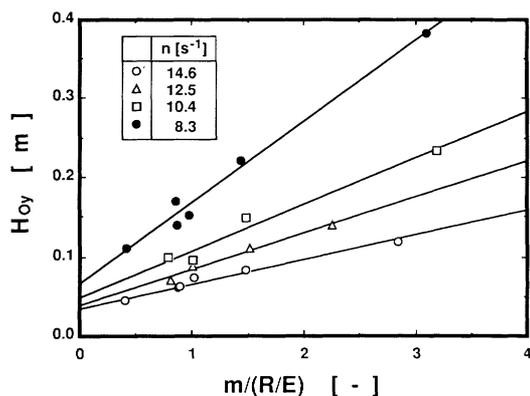


Fig. 7 Height of a transfer unit against $m/(R/E)$

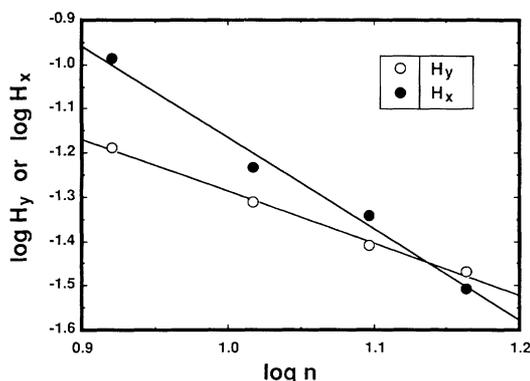


Fig. 8 Correlation of height of a transfer unit against agitation speed

where $x_n^* = y_n/m$. In the same way as for E_{Oy} , the following equation can be derived.

$$(E_{Ox} r_x / m)(mx_{out} - y_{in})(1 + r_x + r_x^2 \dots + r_x^{N-1}) - (x_{in} - x_{out}) = 0 \quad (7)$$

where $r_x = 1/\{E_{Ox}(R/E)/m + 1 - E_{Ox}\}$. In Fig. 5, E_{Ox} for the same data used to calculate E_{Oy} are also plotted against m . E_{Ox} increased with increase in m , in contrast to the change of E_{Oy} . The value of E_{Ox} was larger than E_{Oy} for $m > R/E$ and the reverse was also true. The change of E_{Ox} was large for $m < R/E$, while that of E_{Oy} was large for $m > R/E$.

2.2 Height of a transfer unit

The relation between the overall transfer unit N_{Oy} and E_{Oy} is given as follows in the same way as for the relation between the overall transfer unit and the point efficiency of the distillation column³⁾.

$$N_{Oy} = -\ln(1 - E_{Oy}) \quad (8)$$

With E_{Oy} shown in Figs. 4 and 5, N_{Oy} values were calculated by Eq. (8), and the values were converted to the overall height of a transfer unit by the relation $H_{Oy} = Z/N_{Oy}$, where Z was the height of one stage (9 cm for the present column). H_{Oy} is plotted against $m/(R/E)$ in Fig. 7. Points for a given agitation speed are on a straight line. Bailes-Stitt¹⁾ devised an extraction column which could be operated at vigorous agitation by use of electrostatic coalescence, and gave the height of a transfer unit of 13.4

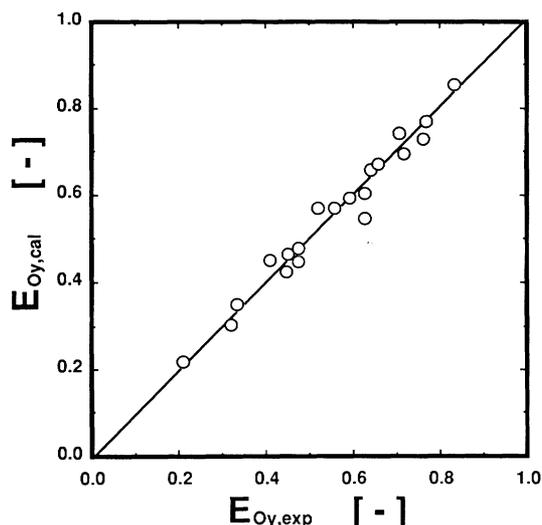


Fig. 9 Comparison of calculated stage efficiencies with experimental ones

cm at an agitation speed of 23 s^{-1} . The values of H_{Oy} of the MS column in Fig. 7 were in the range of 4.5–12 cm at an agitation speed of 14.6 s^{-1} . This indicates that the MS column is excellent in mass transfer characteristics as well as stable in operation.

The relation among the overall H_{Oy} , H_y of the dispersed phase and H_x of the continuous phase is expressed as follows.

$$H_{Oy} = H_y + \{m/(R/E)\} H_x \quad (9)$$

Under an assumption that the straight lines in Fig. 7 correspond to Eq. (9), H_x can be determined from the slope of the line and H_y from the value on the ordinate. These values are plotted against the agitation speed in Fig. 8, and are correlated for the range of $n = 8.3 - 14.6 \text{ s}^{-1}$ as follows.

$$H_y = 0.76 n^{-1.16}, \quad H_x = 7.9 n^{-2.06} \quad (10)$$

Both H_y and H_x decreased with agitation speed, for the specific interfacial area increased due to the increase in dispersed-phase holdup and the decrease in drop diameter with n . The mass transfer coefficient of continuous phase might vary widely with n in comparison with dispersed phase, because the dependency of H_x on n was larger than that of H_y . Stage efficiencies E_{Oy} calculated by using Eq. (10) with the experimental conditions n , m and R/E are compared with the experimental values in Fig. 9, and the average deviation was 4.3 %.

Conclusion

The stage efficiency of the mixer-settler extraction column increased monotonously with increase in agitation speed. Since stable operation was possible at a high agitation speed with this extraction column, both a large stage efficiency and a large throughput, which is desirable for the countercurrent extraction column, could be achieved. The effect of flow ratio R/E and distribution

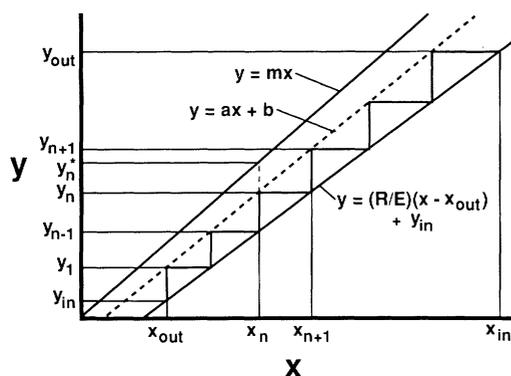


Fig. A-1 x-y diagram for countercurrent extraction column

ratio m on the stage efficiency E_{Oy} were put together as the effect of $m/(R/E)$; i.e., E_{Oy} decreased with the ratio of the equilibrium line slope to the operating line slope. The stage efficiencies E_{Oy} were converted to the overall height of a transfer unit H_{Oy} . The values of H_{Oy} at high agitation speed indicated that the mixer-settler extraction column had excellent mass transfer characteristics. H_{Oy} for each agitation speed was correlated with $m/(R/E)$ by a straight line, and H_x of the continuous phase and H_y of the dispersed phase were determined from the slope of the line and the value on the ordinate, respectively. These heights of a transfer unit of the two phases were correlated with the agitation speed, and the stage efficiency calculated with these correlation reproduced the experimental value with an average error of 4.3 %.

Appendix Determination of stage efficiency

In the present case, the equilibrium line and the operating line are given by Eqs. (1) and (2) in the text, which are straight lines on the x-y diagram as shown in Fig. A-1. Under the assumption that the stage efficiencies for every stage are given by a same value, points (x_{out}, y_1) , (x_2, y_2) , ..., (x_n, y_n) , ..., (x_N, y_{out}) , which represent the concentrations of the dispersed and the continuous phases leaving the stage, are on a straight line as shown by $y = ax + b$ in Fig. A-1, and the following relation can be obtained.

$$y_n - y_{n-1} = (R/E)(x_{n+1} - x_n) \quad (A-1)$$

$$y_{n+1} - y_n = a(x_{n+1} - x_n) \quad (A-2)$$

From these equations,

$$y_{n+1} - y_n = r_y(y_n - y_{n-1}) \quad (A-3)$$

where $r_y = a/(R/E)$. The series $\{y_n - y_{n-1}\}$ is a geometric progression, and the summation of the series is

$$\text{SUM}\{y_n - y_{n-1}\} = y_{out} - y_{in} \\ = (y_1 - y_{in})(1 + r_y + r_y^2 + \dots + r_y^{N-1}) \quad (A-4)$$

From the definition of stage efficiency,

$$E_{Oy} = \frac{(ax_n + b) - \{(R/E)(x_n - x_{out}) + y_{in}\}}{mx_n - \{(R/E)(x_n - x_{out}) + y_{in}\}} \quad (A-5)$$

This equation is rearranged as follows.

$$\{a - mE_{Oy} - (1 - E_{Oy})(R/E)\}x_n + b \\ - (1 - E_{Oy})\{y_{in} - (R/E)x_{out}\} = 0 \quad (A-6)$$

To satisfy Eq. (A-6) for any value of x_n ,

$$a = mE_{Oy} + (1 - E_{Oy})(R/E) \quad (A-7)$$

$$b = (1 - E_{Oy})\{y_{in} - (R/E)x_{out}\} \quad (A-8)$$

And the following equations are derived.

$$r_y = a/(R/E) = E_{Oy}\{m/(R/E) - 1\} + 1 \quad (A-9)$$

$$y_1 - y_{in} = E_{Oy}(y_1^* - y_{in}) = E_{Oy}(mx_{out} - y_{in}) \quad (A-10)$$

By substituting Eq. (A-10) to Eq. (A-4),

$$E_{Oy}(mx_{out} - y_{in})(1 + r_y + r_y^2 + \dots + r_y^{N-1}) \\ - (y_{out} - y_{in}) = 0 \quad (A-11)$$

Eqs. (A-11) and (A-9) are Eqs. (4) and (5) in the text, respectively.

Nomenclatures

E	= flow rate of extract phase	[m ³ /s]
E_{Ox}	= stage efficiency based on aqueous phase concentration	[-]
E_{Oy}	= stage efficiency based on organic phase concentration	[-]
H_{Oy}	= overall height of a transfer unit	[m]
H_x	= height of a transfer unit for aqueous phase	[m]
H_y	= height of a transfer unit for organic phase	[m]
N_{Oy}	= overall transfer unit	[-]
n	= agitation speed	[s ⁻¹]
m	= distribution ratio of iodine	[-]
R	= flow rate of raffinate phase	[m ³ /s]
U_o	= superficial velocity of organic phase	[m/s]
U_w	= superficial velocity of aqueous phase	[m/s]
x	= iodine concentration in aqueous phase	[kmol/m ³]
y	= iodine concentration in organic phase	[kmol/m ³]
Z	= height of one stage	[m]

<Subscripts>

in	= inlet
n	= n-th stage
out	= outlet

<Superscripts>

*	= equilibrium
---	---------------

Literature cited

- Bailes, P.J. and E.H. Stitt: *Chem. Eng. Res. Des.*, **65**, 514-523 (1987)
- Gaubinger, W., G. Husung and R. Marr: *Ger. Chem. Eng.*, **6**, 74-79 (1983)
- Gerster, J.A., A.P. Colburn, W.E. Bonnet and T.W. Carmody: *Chem. Eng. Progr.*, **45**, 716-724 (1949)
- Horvath, M. and S. Hartland: *IEC Process Des. Dev.*, **24**, 1220-1225 (1985)
- Kirou, V.J. and L.L. Tavlarides: *AIChE J.*, **34**, 283-292 (1988)
- Scheibel, E.G.: *Chem. Eng. Progr.*, **44**, 681-690 (1948)
- Steiner, L., E. von Fisher and S. Hartland: *AIChE Symp. Ser.*, **80**, No.238, 230-238 (1984)
- Takahashi, K., M. Nakano and H. Takeuchi: *Kagaku Kogaku Ronbunshu*, **13**, 256-259 (1987)
- Takahashi, K., H. Nakashima, S. Nii and H. Takeuchi: *Kagaku Kogaku Ronbunshu*, **19**, 440-445 (1993)
- Takahashi, K. and H. Takeuchi: *J. Chem. Eng. Japan*, **23**, 12-17 (1990)
- Takahashi, K. and H. Takeuchi, "Solvent Extraction 1990", Elsevier, p. 1357-1362 (1992)