LIQUID-LIQUID EXTRACTION CHARACTERISTICS OF A SPRAY COLUMN WITH A D.C. ELECTRIC FIELD

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Key Words: Extraction, Axial Dispersion Coefficient, Electrostatic Coalescence Dispersion, Electric Field, Electrostatic Spray Column, Liquid-Liquid Extraction, Mass Transfer Coefficient, Simulation Column Efficiency

Axial mixing in the continuous phase and the mass transfer characteristics of a new spray column with four rod electrodes were examined in a range of applied voltages up to 23 kV using a system of water drops in cyclohexane. Iodine was used as a transfer solute for measuring the mass transfer rate and as a tracer for measuring liquid mixing in the continuous phase. The axial dispersion coefficients under applied voltages showed lower values than that in the absence of applied voltage. The extraction efficiencies were increased by about 50 percent for applied voltages of up to 23 kV. It is concluded that these significant enhancement of mass transfer were due to the increased frequency of coalescence and redispersion of the drops.

Introduction

Recently, a new liquid-liquid contactor with a d.c. electric field was developed.⁹⁾ It was shown qualitatively that the contactor had some favorable characteristics for processes such as liquid dispersion, mixing of liquid phases and phase separation, but the dispersed-phase holdup was low.⁹⁾ To improve the holdup, a new spray column equipped with four rod electrodes was designed. The electrohydrodynamics of the drops and holdup characteristic of the column were reported in a previous paper.⁸⁾ In the present study, the effects of applied voltage on mass transfer rate in the dispersed phase and axial mixing in the continuous phase for the column were experimentally examined. The characteristics of the present column and those of a rotating-disk column reported in the literature were compared. Simulation of the present column efficiency was carried out by using fundamental data obtained.

1. Experimental

Figure 1 shows a schematic diagram of the experimental apparatus, which is essentially the same as used in the previous study.⁸⁾ The physical properties of liquids used are listed in Table 1.

1.1 Axial mixing in the continuous phase

The axial mixing in the continuous phase was examined by the technique of pulse injection of a tracer²⁾ as in the following three steps. 1) A dispersed phase (distilled water) and a continuous phase (cyclo-

hexane) were fed into the column from the upper and lower distributors ①, ③, respectively, by roller pumps ⑩. The lower distributor has two feed inlets for the liquid phase: one (black spots in section C-C in Fig. 1) is used for feeding the continuous phase and the other (white spots in section C-C) for feeding a tracer phase. The dispersed and continuous phases were contacted in a two-phase flow countercurrently and a specified voltage ⑮ was applied. 2) After a steady-state flow in both phases was achieved under the applied voltage, the feeding of the continuous phase was cut off and simultaneously a tracer phase (cyclohexane including a tracer (iodine)) was fed at the same flow rate. After the tracer phase was fed for a specified period of time, the process was reversed: the feed time of the tracer phase was chosen as one-tenth of the residence time of the continuous phase in the column.²⁾ 3) After the tracer phase was fed, the continuous phase at the outlet ⑦, ⑧ of the column was sampled at a specified interval and the tracer concentration in the phase was analyzed by iodometry.

1.2 Mass transfer

The extraction efficiency of the iodine transfer from a population of charged drops of aqueous iodine solution into the continuous phase was examined for various applied voltages. Iodine as a transfer material was used to evaluate the mass transfer rate in the dispersed phase because the distribution coefficient of iodine between the dispersed and continuous phases was 0.015. The countercurrent contacting procedure of both phases was essentially the same as mentioned in step 1 of paragraph 2-1). The total amount of iodine transferred was determined as follows. In the

Received May 6, 1988. Correspondence concerning this article should be addressed to T. Katayama. H. Sugaya is now with Toray Ind., Inc., Ohtsu 520.

- 1; Distributor of dispersed phase
- 2; Spray column with electrodes
- 3; Distributor of continuous (tracer) liquid phase
- 4, 5; Copper rod electrode
- 6; Inlet and outlet of dispersed phase
- 7; Inlet and outlet of tracer liquid
- 8; Inlet and outlet of continuous phase
- 9; Burette 10; Roller pump
- 11; Air damper 12, 13, 14; Liquid reservoir
- 15; High d.c. voltage generator
- 16; Electrometer 17; T.V. system

Fig. 1. Schematic diagram of experimental apparatus

Table 1. Physical properties of liquids used at 20°C

	Density	Inter- facial tension	Viscosity	Dielectric constant	Electric conductivity
	[kg/m ³]	[mN/m]	[mPa·s]	[—]	[S/m]
Water	998	—	1.00	80.1	10 ⁻⁶
Cyclohexane	770	48.0	0.98	2.02	10 ⁻¹⁶

dispersed phase the initial and final concentrations of the iodine were sampled at the inlet and outlet of the column, respectively. To prevent volatilization of iodine into the atmosphere the samples were taken directly into aqueous potassium iodine solutions. The iodine concentration was analyzed by iodometry.

2. Results and Discussion

2.1 Axial mixing in the continuous phase

Typical examples of axial mixing characteristics in the continuous phase are depicted in Fig. 2, in which the experimental tracer concentration flowing out from the top of the column is plotted against the time lapse after feeding the tracer into the bottom of the column. Examples are shown for applied voltages of 0 and 20 kV under a constant feed rate for the dispersed

Fig. 2. Concentration response of the liquid flowing out of the spray column in the absence and presence of an electric field ($U_d = 0.029$ cm/s, $U_c = 0.098$ cm/s)

and continuous phases, respectively. The maximum value of the tracer concentration in the absence of an electric field is reached earlier than that in the presence of an electric field. This may be due to the following reasons. In the latter case, contact between the dispersed and continuous phases resulted in a complicated flow; the charged drops fell with a violent zig-zag motion accompanying the coalescence-redispersion of the drops⁸⁾ and hence the forward flow of the tracer was depressed. In the former case, the contact resulted in a simpler flow; the uncharged drops fell straight down⁸⁾ and hence the forward flow of the tracer was unimpeded. The mixing behavior in the continuous phase was analyzed by the diffusion model.^{2,7)} The response curves (solid and dotted) of the tracer concentration in Fig. 2 were calculated by the model, in which Pe_c -values as a fitting parameter were determined from the measurement in each run in order to obtain the least-squares error between the function in the model and the experimental data. The diffusion model used can satisfactorily express the axial mixing characteristic of the column. The experimental axial dispersion coefficient, E_c , obtained from Pe_c -value is plotted against the applied voltage in Fig. 3. The coefficient decreases significantly with the voltage. It is considered that this occurs because the forward axial mixing in the continuous phase was depressed with the increase in the voltage, and the radial mixing in the continuous phase was promoted by the complicated motion of the drops.

2.2 Mass transfer

Figure 4 shows the experimental extraction efficiency, F , of iodine in the dispersed phase as a function of the applied voltage at a constant flow rate for both phases. As shown in the figure, up to 10 kV the effect of applied voltage on the efficiency is very small. This is evident from the results that the population of drops fell through the continuous phase with

Fig. 3. Effect of electric field on axial dispersion coefficient in the continuous phase ($U_d = 0.029$ cm/s, $U_c = 0.098$ cm/s)

Fig. 4. Effects of applied voltage on extraction efficiency and overall volumetric mass transfer coefficient based on dispersed phase

a mild motion and the frequency of coalescence and redispersion of the drops was low. When the voltage exceeds 10 kV, the effect becomes more conspicuous. The efficiency at 23 kV increases about by 50% compared with that at 0 kV. This enhancement is due to the increased frequency of coalescence and redispersion of the drops (interfacial renewal) and the increased residence time together with marked internal mixing inside the drops: it is considered that the end effects of mass transfer during the formation and coalescence of the drops are constants because both the flow rates in both phases are constant. In general, as there is a concentration profile in the continuous phase, the extraction efficiency of the dispersed phase depends not only on the residence time distribution of the drops but also on the path of motion of the drops. Furthermore, because of the complicated behavior of the drops, it is difficult to estimate the efficiency analytically. Therefore, the overall volumetric mass transfer coefficient, $K_d a$, has been analyzed by a unidimensional dispersion model.³⁾ In applying the model to the dispersed phase, it is assumed that the concentration of each drop is the same through a

Fig. 5. Modified dispersion model

given cross section of the column. In the absence of applied voltage, the size distribution of the drops is close to monodispersity⁸⁾ and so the drop motion is similar at a given position in the column. On the other hand, in the presence of applied voltage an increase of the voltage widens the drop size distribution, but the concentration of each drop will be the same because of their intensive coalescence and redispersion. The model equations for both phases were solved numerically by the Runge-Kutta-Gill method: the $K_d a$ value in the equations was determined by fitting calculated inlet and outlet iodine concentrations of the column for both phases to experimental ones. The values obtained are plotted against the applied voltage in Fig. 4. The enhancement of $K_d a$ -value with increased voltage shows a trend similar to that of the experimental extraction efficiency.

2.3 Simulation of column efficiency

To simulate the column efficiency, the dispersion model³⁾ was modified under the following assumptions. 1) Coalescence and redispersion of the drops do not occur, 2) concentrations of iodine in both phases are low, hence the physical properties of both phases are constant during the experiment, 3) the drops are spherical, 4) the mass transfer resistance is controlled in the dispersed phase, and 5) size distribution and residence time distribution of the drops, respectively, are estimated by the Mugele-Evens distribution function⁴⁾ and the velocity equation for falling charged drops.⁸⁾ The axial dispersion coefficient and the holdup fraction used are those obtained in the present experiments. The model diagram is shown in Fig. 5. The model is expressed by the following integral differential equations:

for dispersed phase

$$U_d \frac{dC_d}{dz} + 6\phi \int_{d_{e,min}}^{d_{e,max}} \frac{K_{d,j} f(d_{e,j})}{d_{e,j}} (C_{d,j} - C_d^*) d(d_{e,j}) = 0 \quad (1)$$

for continuous phase

$$(1-\phi)E_c \frac{d^2 C_c}{dz^2} + U_c \frac{dC_c}{dz} + 6\phi \int_{d_{e,\min}}^{d_{e,\max}} \frac{K_{d,j} f(d_{e,j})}{d_{e,j}} (C_{d,j} - C_d^*) d(d_{e,j}) = 0 \quad (2)$$

for j -group of drop

$$\frac{dC_{d,j}}{dz} = -\frac{1}{U_{z,j}} K_{d,j} \frac{6}{d_{e,j}} (C_{d,j} - C_d^*) \quad (3)$$

The $k_{d,j}$ -value in the equations is estimated by an empirical equation⁵⁾ for single uncharged drops.

$$k_{d,j} = 31.4 \left(\frac{4D_d t_{r,j}}{d_{e,j}^2} \right)^{-0.338} \left(\frac{\mu_d}{\rho_d D_d} \right)^{-0.125} \left(\frac{d_{e,j} U_{z,j}^2}{\sigma} \right)^{0.371} \frac{D_d}{d_{e,j}} \quad (4)$$

When the drop size distribution obtained in the previous work⁸⁾ was divided into 20 intervals, the drop velocity, holdup fraction and mass transfer coefficient corresponding to the drop diameter can be estimated. Consequently, 23-element simultaneous ordinary differential equations of the first order were obtained from Eqs. (1), (2) and (3). These equations were solved numerically by the Runge-Kutta-Gill method. The calculated concentration profiles of the dispersed and continuous phases are shown for $E_p = 0$ and 23 kV in **Figs. 6a** and **6b**; the dotted lines show simulated profiles and the solid lines show the values calculated by using the experimental extraction efficiency. For $E_p = 0$ kV, the differences between experimental and simulated values are small and so it is considered that column simulation is good. On the other hand, for $E_p = 23$ kV the solid lines show higher efficiency than the dotted lines. The present model takes into account size distribution and residence time distribution of the drops, but does not take into account the effect of the coalescence and redispersion of the drops. Therefore, the difference between the values may be caused by interfacial renewal and mass transfer promotion due to repetition of the coalescence and redispersion of the drops falling through the continuous phase.

2.4 Comparison between present column and RDC performances

To evaluate the column efficiency, the axial dispersion coefficient and the overall volumetric mass transfer coefficient in the present column were compared with those of a RDC (rotating-disk column) in the literature.¹⁾ The axial dispersion coefficient in the RDC was estimated by an empirical equation of Strand *et al.*⁶⁾ by using experimental data.¹⁾ The values are plotted against electrical energy consumed

Fig. 6. Column efficiency based on modified dispersion model

Fig. 7. Overall volumetric mass transfer coefficient ($K_d a$) and axial dispersion coefficient (E_c) vs. energy consumption (P). $\triangle$, $\circ$: this work, $U_d = 0.0287$ cm/s, $U_c = 0.0981$ cm/s. $\blacktriangle$, $\bullet$: Al-Aswad *et al.*,¹⁾ Clairsol350 (d)-acetone (s)-water (c), $U_d = 0.028$ cm/s, $U_c = 0.66$ cm/s

for each flow rate of dispersed phase in **Fig. 7**. Both the E_c - and $K_d a$ -values in the present column are somewhat better than those in the literature.

Conclusion

The axial mixing and mass transfer characteristics were studied for the liquid-liquid spray column developed in the previous work. The characteristics obtained were as follows.

- 1) The coalescence and redispersion behavior of charged drops in an electric field significantly influence extractor performance. This behavior can easily be controlled by means of an applied voltage.
- 2) The axial mixing coefficient decreases with the applied voltage.
- 3) The extraction efficiency and overall volumetric mass transfer coefficient increase effectively with the applied voltage.
- 4) Accurate simulation of the present column's efficiency requires much more fundamental information about coalescence and redispersion of the drops.

Acknowledgment

This work was partially supported by a Grant-in-Aid for Energy Research (2) (Project No. 61040052) from the Ministry of Education, Science and Culture of Japan, and by a Financial Aid to Research Grant from the Asahi Glass Foundation for Industrial

Technology (1987). The authors are grateful to Mr. S. Yoshiyasu for his assistance with the calculation of the column simulation.

Nomenclature

C	= concentration of iodine	[mol/m ³]
D_d	= diffusivity of iodine in water	[m ² /s]
d_e	= equivalent spherical drop diameter	[m]
E_c	= axial dispersion coefficient in continuous phase	[m ² /s]
E_p	= applied voltage	[V]
F	= total extraction efficiency	[—]
$f(d_e)$	= volumetric distribution density of drop	[—]
K_d	= overall mass transfer coefficient based on dispersed phase	[m/s]
K_{da}	= overall volumetric mass transfer coefficient based on dispersed phase	[1/s]
k_d	= mass transfer coefficient in dispersed phase	[m/s]
L	= column height	[m]
P	= energy consumption/dispersed phase flow rate	[kJ/m ³]
Pe_c	= Peclet number ($= U_c L / \{2(1 - \phi)E_c\}$)	[—]
t	= time	[s]
t_c	= residence time of continuous phase	[s]
t_r	= residence time of drop	[s]
U	= superficial velocity of liquid phase	[m/s]
U_z	= falling velocity of drop	[m/s]
U_t	= moving velocity of drop	[m/s]
z	= distance from top of column	[m]
μ	= liquid viscosity	[Pa · s]
ρ	= liquid density	[kg/m ³]

σ	= interfacial tension	[N/m]
ϕ	= holdup fraction of dispersed phase	[—]

<Subscripts>

c	= continuous phase
d	= dispersed phase
i	= inlet
j	= j -th group of drops
$\max$	= maximum
$\min$	= minimum
o	= outlet

<Superscript>

$*$	= equilibrium
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