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Axial mixing and mass transfer characteristics of pulsed extraction column with discs and doughnuts

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Abstract: The axial mixing is a key factor for design and scaling up of the pulsed extraction column which has strong influence on the mass transfer performances of the pulsed extraction column with discs and doughnuts. A steady-state concentration profile measurement was used to evaluate the mass transfer and axial mixing coefficients for the nitric acid/water/30%TRPO (in kerosene) system in the pulsed extraction column with the diameter of 38 mm on the condition of the TRPO-kerosene solution as the continuous phase and the flow ratio of 1 : 1. Experimental results indicate that E_y evaluated by the experiments is in good agreement with that given by Burratti's correlation; the axial mixing is far smaller for the continuous phase than that for the dispersed phase. However the empirical correlation for H_{OX} is only given based on the present data.

Key words: pulsed extraction column; axial diffusion model; mass transfer; steady-state concentration profile

1 Introduction

Liquid-liquid extraction has been regarded as one of the most useful separation process. In recent years, a new type of extraction column, called the discs and doughnuts pulsed extraction column, has been developed and extensively applied to reprocess the spent nuclear fuel in France and Japan[1, 2]. The extraction column has shown the following advantages: high efficiency, perfect flooding throughputs, no rotational parts, easy clearance and so on.

The conclusion that the axial mixing of the two phases countercurrent flow leads to the decreasing of the mass transfer efficiency[3] has been summarized by LUO[4] for the pulsed sieve-plate extraction column. However, the knowledge concerning the hydrodynamics and mass transfer performance of the pulsed extraction with discs and doughnuts is rarely reported. Only the axial mixing characteristics for the extraction column have been investigated and summarized in Table 1.

In Table 1, the correlations of the axial dispersion coefficient of continuous phase in the column are given by ZAHARIEVA and MASBERRAT[5], NABLI and GUIRAUD [6], OH[7], BURATTI[8] respectively, by means of the numerical simulation, and the conclusions are drawn as follows:

1) $E_{\rm c}$ increases with the increase of the plate

spacing H.

2) The effect of D on E_c can be neglected, as described by BURRATTI.

3) Packing free area α is independent of $E_{\rm c}$.

It is shown that at the same column parameters and operational parameters, E_c predicted by BURRATTI's correlation is smaller than that by OH and NABLI's correlations. From Table 1, it is easily observed that E_c increases with the increase of the pulsed parameters, namely, A and f.

The dispersed phase axial mixing characteristics was only reported by BARDIN-MONNIR and GUIRAUD[9] for pulsed extraction column with discs and doughnuts. The dispersed phase axial mixing coefficient, E_d , was simulated with computational fluid dynamics and Lagrangian simulations, and the result showed the axial mixing was far less important (about one tenth) for the continuous phase than for the dispersed phase, but the continuous phase axial mixing was larger than the axial mixing of a single drop.

In order to provide the sufficient knowledge of the design and scaling up when the pulsed extraction column with discs and doughnuts is used in the nuclear the plate spacing on the axial mixing and mass industry of China, the axial mixing model combined with the steady-state concentration profiles method is used to

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Author	Column parameters and experimental system	Investigation method	Investigation results
A. ZAHARIEVA and O. MASBERRAT[5]	D=0.28 m, $\alpha = 23.5\%,$ H=0.045 m, h = 0.16, $H_c=2.6 \text{ m},$ unknown experimental system	The space-time flow structure in the column has been investigated using laser doppler velocimetry. Measurements of radial and axial velocity components have been performed. A numerical procedure was used to calculate the axial dispersion coefficient as a function of the geometrical and pulsation parameters.	$\frac{E_c}{AfD} = 1.1h^{1.64}A^{*-0.22}f^{*0.034}$ $A^* = A/D, f^* = FD^2/v$ $0.057 \le A^* \le 0.173,$ $55\ 406 \le f^* \le 124\ 582$
M. A. NABLI, and P. GUIRAUD[6]	$D=0.288 \text{ m},$ $17 \le \alpha \le 40\%,$ $0.045 \le H \le 0.125 \text{ m},$ $0.15 \le h \le 0.41,$ unknown experimental system	Hydrodynamic characteristics and axial mixing were calculated by the ESTET code.	$E_c = 1.67 H^{1.56} A^{0.69} f^{0.92}$
N.BARDIN- MONNIER, and P. GUIRAUD[9]	D=0.3 m, H=0.045 m, $\alpha = 22\%,$ continuous phase: water(1 000 kg/m ³) dispersed phase: toluene-CEROL SOUDANIV (867 kg/m ³)	This paper intends to evaluate in which way the computational fluid dynamics, and Lagrangian simulations, can be used as a tool for getting the information about the hydrodynamic and axial mixing.	$E_c \propto Af$ $E_d > E_c > E_{sd}$
M.BURATTI[8]	0.025 m $\leq D \leq$ 0.100 m, 0.012 5 m $\leq H \leq$ 0.050 m, 0.25 $\leq h \leq$ 1.11, 12.5 $\leq \alpha \leq$ 25		$E_c = 0.71 H^{0.74} A^{1.13} f^{0.98}$ $0.10 \leqslant A \leqslant 0.05$ $0.333 \leqslant f \leqslant 1.5$
W.Z.OH[7]	D=0.28 m, $0.15 \leq h \leq 0.23$		$E_c = \frac{H}{\alpha} (Af)$ $0.01 \leq Af \leq 0.35$

Table 1 Re	ported results of	faxial mixing	characteristics in	pulsed extraction	columns with	discs and doughnuts
		<i>U</i>				

evaluate the effect of the pulsation intension and transfer characteristics for the pulsed extraction column with discs and doughnuts at the phase ratio R=1.

2 Experimental

2.1 Experimental set-up and system

The experimental apparatus, as shown in Fig.1, consisted of four parts: a discs and doughnuts column (1), the pneumatically pulsed system (20-22), the feeding and discharging system of the two phases (9-19), and the measurement system (5-8). The column, which was made of a vertical cylindrical glass pipe, contained a series of discs and doughnuts alternately arranged and

maintained by the braces of constant length, as shown in Fig.2, and Fig.3. The parameters of the column and the plate used in the experiments are shown in Table 2.

Table 2 Structure parameters of column

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Parameter	Value	
D/m	0.038	
$H_{\rm c}/{ m m}$	1.670	
<i>H</i> /m	0.015, 0.012, 0.	.010
$D_{\rm o}/{ m m}$	0.018	
$D_{\rm i}/{ m m}$	0.034	
α /%	23.0	

There were six pairs of sampler arranged along the wall of column, as shown in Fig.1(23). The sampling tubes in each pair, which withdrawn the samples for the continuous phase and the dispersed phase respectively at the same time, were filled with the hydrophilic fibre and the lipophilic fibre respectively to avoid the mixing of two phases.

The experimental materials used in the experiments were nitric acid solution for the aqueous phase and



Fig.1 Schematic diagram of pulsed extraction column with discs and doughnuts: 1 Column; 2 Top settler; 3 Bottom settler; 4 Pulsation leg; 5 Pressure transducer; 6 Computer; 7 Top purge probe; 8 Bottom purge probe; 9 Aqua outlet valve; 10 Aqua outlet rotameter; 11 Aqua feed tank; 12 Aqua inlet valve; 13 Aqua inlet rotameter; 14 Solenoid valve; 15 Aqua pump; 16 Organic inlet valve; 17 Organic inlet otameter; 18 Organic valve; 19 Organic feed tank; 20 Pressurized air; 21 Rotary valve; 22 Free air; 23 Sampler



Fig.2 Schematic diagram of column



Fig.3 Schematic diagram of discs and doughnuts

30%TRPO-kerosene solution for the organic phase. The physical properties of the two solutions are listed in Table 3. During the experimental process, the feed concentration of HNO₃ solution was about 0.6 mol/L, and that of HNO₃ solution in the organic phase was less than 0.1 mol/L. The equilibrium relationship of HNO₃ in the two phases within the experimental range at room temperature is expressed as

 $x^* = 9.860y^3 - 5.163y^2 + 1.378y - 0.001$ (1) (0.02 mol/L \le v \le 0.6 mol/L)

System	$\rho/$	μ /	σ/
	$(\text{kg} \cdot \text{m}^{-3})$	$(10^{-3}$ Pa • s)	$(10^{-3}N \cdot m^{-1})$
30%TRPO-	201	2.6	16.07
Kerosene	801	2.0	10.87
1.0 mol/L HNO ₃	1015	1.0	
solution	1015	1.0	

2.2 Procedure

When the organic phase was as a continuous phase, the column was firstly full of the TRPO-kerosene solution, then the pulse amplitude and frequency were set at the desired values, and the flow rates of the two phases were regulated to the desired values at R=1: 1, finally, the interface of the two phase in the lower expanded section was observed at the required level. After 30 min, the state of the flow of two-phase in the column was considered as the steady. The dispersed phase hold-up was on-line measured by the air-purge method[10]. The experimental conditions are shown in Table 4.

Table 4 Operation parameters used in experiment

Parameter	Range
$u_{\rm c}/({\rm m} \cdot {\rm s}^{-1})$	0.00121-0.00607
A/m	0.01-0.03
<i>f</i> /Hz	1.00—1.67

When the volume of the dispersed phase withdrawn from the aqueous outlet was about five times larger than that of the column and the HNO_3 concentration of the aqueous phase from the outlet was sampled and remained constant by titration, therefore, the steady-state concentration profiles was developed in the column. The two phases were sampled and then the HNO_3 concentrations in the two phases were determined by titration, respectively.

2.3 Modeling and algorithm[4]

Based upon the axial diffusion model and mass balance in the column, over the differential elements of the column with a cross-section area and total effective height H_c , the equation set for the steady-state process was established as follows, under the constant superficial velocities u_x and u_y at any given pulse amplitude and frequency:

$$\begin{cases} \frac{\partial}{\partial Z} \left(x - \frac{1}{Pe_x} \frac{\partial x}{\partial Z} \right) + N_{OX} \left(x - x^* \right) = 0 \\ \frac{\partial}{\partial Z} \left(y + \frac{1}{Pe_y} \frac{\partial y}{\partial Z} \right) - N_{OX} \frac{u_x}{u_y} \left(x - x^* \right) = 0 \end{cases}$$
(2)

where $Pe_x = H_c u_x / E_x$, $Pe_y = H_c u_y / E_y$, $Z = h' / H_c$. $K_{acc} \alpha H$

Noting that in Eqn.(2)
$$N_{OX} = \frac{N_{OC} \omega n_c}{u_c}$$
 represents

$$N_{\rm OX} = \frac{H_{\rm c}}{H_{\rm OX}}.$$

The steady-state boundary conditions are

As Z=0,

$$\begin{cases} -\frac{\mathrm{d}x}{\mathrm{d}Z} = Pe_{x} \left(x_{\mathrm{F}} - x_{Z=0^{+}} \right) \\ \frac{\mathrm{d}y}{\mathrm{d}Z} = 0 \end{cases}$$
(3)

$$\begin{cases} -\frac{\mathrm{d}x}{\mathrm{d}Z} = 0\\ -\frac{\mathrm{d}y}{\mathrm{d}Z} = Pe_{y} \left(y_{Z=1^{-}} - y_{s} \right) \end{cases}$$
(4)

For solution of this equation set, a differential equation in mean implicit form was applied with the suitable distance step. The three parameters of Pe_x , Pe_y and N_{OX} in the equation set were evaluated by combining Marquadt optimization with the measured steady-state concentration profile[10].

3 Results and discussion

Fig.4 shows the steady-state concentration profiles at H=0.015 m, $A_{\rm f}$ =0.02 m/s and Φ =10%. In Fig.4, the solid points represent the measured concentrations profile and the line does that evaluated by the model. The results show that the steady-state concentration profile in the column is in good agreement with that predicted by the axial diffusion model at the evaluation of three parameters, such as $E_{\rm x}$, $E_{\rm y}$ and $N_{\rm OX}$.

3.1 Continuous phase E_y

The influence of the dispersed phase hold-up Φ and the plate spacing H on E_y at the $A_f=0.02$ m/s is shown in Fig.5. Within the experimental range, E_y increases with the increase of *H*.



Fig.4 Steady-state concentration profiles



Fig.5 Influence of Φ and H on E_{y}

Fig.6 shows the influence of the pulse amplitude A and the frequency f on E_y . From Fig.6, it is shown that E_y increases with the increase of A and f respectively.



Fig.6 Influence of A and f on E_y

The comparison of the values of E_y with those predicted by NABLI's, OH's and BURRATTI's correlation is shown in Fig.7. It is shown that E_y obtained by experiment fits well to that of BURRATTI's correlation obtained by experiment, but is smaller than those of NABLI and OH's correlation by numerical simulation method. Therefore, the values of E_y in the pulsed extraction column with discs and doughnuts can be described by BURRATTI's correlation.



Fig.7 Comparison of E_y correlated with E_y measured

3.2 Dispersed phase E_x

The influence of H and Φ on E_x is shown in Fig.8. The result shows that within the experimental range, E_x increases with the decrease of Φ , however at the same Φ , E_x increases with the decrease of H. The resistance of axial mixing increases for the increase of dispersed phase volume ratio in the vortex with the increase of Φ , and the axial mixing decreases for the decrease of the vortex intensity while increasing H.



Fig.8 Influence of *H* and Φ on E_x

Fig.9 shows the effect of Φ and H on E_x/E_y . From Fig.9, it is shown the E_x/E_y increases with the decrease of Φ , but at the same Φ , E_x/E_y increases with the

decrease of *H*. E_x/E_y is larger than 1 is the same as the BARDIN-MONNIER's conclusion[8], which the axial mixing is far less important (about one tenth) for the continuous phase than for the dispersed phase. Based on the correlation of E_y , the following correlation for E_x has been established by linear regression of the experimental data:

$$E_{\rm x} = 0.026\Phi^{-0.92}H^{-0.45}A^{1.13}f^{0.98}$$
⁽⁵⁾

The comparison of the calculated E_x with the experimental values is shown in Fig.10 with a maximum deviation of 10%.



Fig.9 Effect of Φ and H on E_x/E_y



Fig.10 Comparison of $E_{x, exp}$ with $E_{x, cal}$

3.3 'True' height of transfer unit H_{OX}

The definition of 'true' height of mass transfer unit, H_{OX} , is given as follows:

$$H_{\rm OX} = \frac{u_{\rm c}}{K_{\rm oc}\alpha} \tag{4}$$

Fig.11 shows the influence of H on H_{OX} under the same Φ . The results indicate that within the experimental range, H_{OX} is independent of H in this experimental system.



Fig.11 Influence of H on H_{OX} under the same Φ

For the pulsed sieve plate extraction column, LUO[4] derived the following equation for the 'true' height of transfer unit, which is independent of column diameter, by using the two-film model, single drop mass transfer and two phase equation:

$$H_{\rm OX} = K \frac{u_{\rm c}}{\Phi(1-\Phi)} \tag{5}$$

From Eqn.(5), H_{OX} is the function of the parameters, namely, K, u_c and Φ , where the constant Kvaries with the physical properties of the experimental system and the configuration of the column. Compared the pulsed sieve plate extraction column to that with discs and doughnuts, it is the similarity that a turbulent circulating pattern dominates the mass transfer performance. Therefore, we can induce that the influencing factors of the H_{OX} for the latter column is the same as those for the former, i.e. H_{OX} is also the function of K, u_c and Φ . In our experimental process, the values of Φ are measured on-line by air-purge method and the configuration of column remains unchanged, therefore, H_{OX} is only correlated with Φ and u_c by linear regression of the experimental data:

$$H_{\rm OX} = 0.29\Phi^{-0.24}u_{\rm c}^{-0.21} \tag{6}$$

The calculated values of H_{OX} from Eqn.(6) are compared with the experimental data as shown in Fig.12 with a maximum deviation of 5%. H_{OX} decreases with the increase of Φ and the increase of u_c , however the influence of H on H_{OX} can be neglected.

4 Conclusions

The steady-state concentration profile method for the evaluation of mass transfer and axial mixing parameters worked successfully in a pulsed extraction column with discs and doughnuts. The results are as follows:



Fig.12 Comparison of $H_{OX, exp}$ with $H_{OX, cal}$

1) E_y increases with the increase of A_f and H, and is satisfied to the BURRATTI's correlation.

2) E_x increases with the decrease of Φ , and at the same Φ , E_x increases with the decrease of *H*, and is predicted by

$$E_{\rm x} = 0.026 \Phi^{-0.92} H^{-0.45} A^{1.13} f^{0.98}$$

3) H_{OX} decreases with the increase of Φ and u_{c} , however the influence of H on H_{OX} can be neglected. The relation of H_{OX} to Φ and u_{c} is

$$H_{\rm OX} = 0.29 \Phi^{-0.24} u_{\rm c}^{-0.21}$$

Further experimental studying on the mass transfer should be focused on forming a simple equation of H_{OX} so as to predict the performance for the design and scaling up of a pulsed extraction column with the discs and doughnuts.

Nomenclature

- *a* Specific surface area, m^2/m^3 ;
- *A* Pulse amplitude, m;
- C Concentration of HNO₃ solution, mol/L;
- *D* Column diameter. m:
- D_i Disc diameter, m;
- *D*_o Doughnut aperture, m;
- *E* Axial mixing coefficient of solute, m^2/s ;
- f Pulse frequency, Hz;
- *h'* Distance step;
- $H_{\rm c}$ Effective height of the column, m;
- *H* Plate spacing, m;
- *H*_{OX} 'True' height of transfer unit, m;
- *K* Constant;
- K_{oc} Overall mass transfer coefficient based on continuous phase, m/s;
- *N*_{OX} Number of 'true' transfer unit;
- *Pe* Peclet number;
- *R* Phase ratio;

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- *u* Superficial velocity, m/s;
- *x* Aqueous phase concentration, mol/L;
- x^* Equilibrium concentration of the aqueous phase, mol/L;
- *y* Organic phase concentration, mol/L;

Greek letters

- α Packing free area, %;
- μ Viscosity, Pa s;
- ρ Density, kg/m³;
- Φ Holdup of dispersed phase, %;
- σ Interfacial tension, N/m;

Subscripts

- c Continuous phase;
- d Dispersed phase;
- F Feed flow;
- s Solution;
- sd Single drop;
- x Aqueous phase;
- y Organic phase;

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