LIQUID-PHASE MIXING IN A MULTISTAGE VIBRATING-DISK COLUMN WITH CONCURRENT GAS-LIQUID FLOW^{*}

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The liquid-phase mixing characteristics of a multistage vibrating-disk column with a concurrent gas-liquid flow are studied by means of a step response method and a moment analysis. The mixing characteristics can be expressed by the back-flow model. The model parameter (the back-flow ratio) is well correlated by a dimensionless equation of the experimental variables. The correlation is definitely affected by the wave form of the disk vibration.

Introduction

A multistage vibrating-disk column is a gas-liquid and liquid-liquid contactor consisting of many stages separated by circular partition plates. The fluids in every stage are agitated by the vibrating disks which are attached to an agitating shaft. The partition plates have a hole at their center, and fluids flow through the holes concurrently or counter-currently. This operating unit has usually been used as a gas-liquid reactor with suspended solid catalysts³), and also used as gas absorption and liquid-liquid extraction columns^{5,7}). In all cases it has been proved to be useful equipment, but the effects of the design variables and the operating conditions on the fluid mixing characteristics of the column have rarely been reported⁴).

In this paper, the liquid-phase mixing characteristics of a multistage vibrating-disk column are discussed by means of a step response method and a moment analysis in the case of gas-liquid contacting operations with concurrent upward flows. The tracer material is generated by electrolysis of an electrolyte and is injected stepwise into the liquid flow without any change of flow rate⁸⁾.

Experimental

The experimental apparatus is shown schematically in **Fig. 1**. **Figure 2** shows the details of the multistage vibrating-disk column. The column consists of seven stages which are fabricated from methyl metaacrylate

* Received on May 24, 1973

Presented at The 36th Annual Meeting of The Soc. of Chem. Engrs., Japan, at Tokyo, April 1971 〒591 堺市百舌鳥梅町4-804 大阪府立大学工学部化学工学科 宮南 啓 resin (MMA) pipes, 50 mm inside diameter and 35 mm length. Each of the stages is partitioned by a MMA circular plate having a hole at its center. Agitation in the column is provided by vibrating disks driven by a single agitating shaft. The interval between the adjacent disks is the same as that of the partition plates. At the top of the agitating shaft, two pieces of a small iron block are attached so that the shaft can move up and down by activating a pair of solenoid coils alter-



ST; storage tank, P; pump, HT; constant head tank, V_1 , V_2 ; needle valves, RM; rotameter, EC; electrolytic cell, PM; premixer, IFC; inlet flow cuvette, CP; compressor, O; oriffice flow meter, CR; reactor, S; gas-liquid separator, CM; colorimeter, A; ammeter, PS; constant current source OFC; outlet flow cuvette

Fig. 1 Experimental apparatus



Fig. 2 Multistage vibrating-disk column

natively. At the bottom of the column, the gas distributor (a perforated plate having 16 holes of 1 mm diameter) is installed.

Air and 0.05N sodium bromide solution containing $6-10 \ \mu \text{mol}/l$ of methyl orange (MO) are used for the gas and liquid phases. The pH value of the solution is adjusted to 2 by sulfuric acid. The solution is pink-red in color and its physical properties are essentially those of water.

The flow rate of air is regulated by the needle valve V_2 and is measured by the orifice flow meter O. The solution from the constant-head tank HT flows through the needle valve V_1 , the flow meter (rotameter) RM, the electrolytic cell EC, the premixer PM and the inlet flow cuvette IFC, and then is fed to the column at the bottom.

The gas and liquid phases enter the column at the bottom, flow upward concurrently, and leave the column from the top. After separation of the gas and liquid phases in the separator S, the liquid phase enters the outlet flow cuvette OFC.

Bromine is generated stepwise at the anode of the electrolytic cell EC by passing an electrolytic current through the cell. The bromine reacts instantaneously with MO in the solution, thus decreasing its color intensity stepwise. The concentration of MO can be continuously followed by the appropriate colorimeter⁸⁾.

The gas holdup in the column is measured statically. The dimensions and the operating conditions of the column are listed in **Table 1**.

Results and Discussion

The mean residence time \overline{t} and the variance σ^2 of the residence time distribution of the liquid phase are calculated by Eqs.(1) and (2):

 $\sigma^2 = \overline{t^2} / (\overline{t})^2 - 1$

$$\overline{t} = \int_0^T \left\{ 1 - \mathbf{f}(t) \right\} dt \tag{1}$$

(2)

where

$$\overline{t^2} = 2 \int_0^T t \left\{ 1 - f(t) \right\} dt \tag{3}$$

f(t) is the tracer step response of the column and T the time at which the response can be regarded as reaching its steady state value.

Eqs.(1) and (3) have been numerically integrated by Simpson's formula. The integrals of Eqs.(1) and (3) are truncated at T=2.6 to 4.0 when the value of 1-f(t), becomes less than 0.01, which corresponds to the accuracy of the instrument used for the analysis.

The reciprocal of σ^2 is equivalent to the number of equi-volume perfect mixers in series, N_{eq} :

$$1/\sigma^2 = N_{eq} \tag{4}$$

 N_{eq} has been measured at various experimental conditions of u_g , u_l , ν , A, d_h and d_d . The results show that N_{eq} varies between 3.5 (half the number of actual



Fig. 3 Effect of disk diameter and hole diameter on N_{eq}

Table 1Experimental conditions

number of stages, hole diameter vib. disk diameter vib. frequency vib. amplitude liq. velocity or velocity	$N/2$ d_h d_d ν $2A$ u_l	= 7 $= 1.2, 2.0, 3.0, 4.0$ $= 3.0, 4.0$ $= 0, 50, 100, 150, 200$ $= 0, 3.0$ $= 0.16-0.77$ $= 0.4.2$	[—] [cm] [cpm] [cm] [cm/sec]
gas velocity	u_g	= 0-4.2	[cm/sec]

stages) and 14 (twice the number of actual stages). A typical case is shown in **Fig. 3**, where the effects on N_{eq} of the vibrating-disk diameter d_d and the hole diameter d_h of the partition plate are given. The abscissa of the graph represents the ratio of the disk area S_d to that of the partition plate $S_i - S_h$, i.e.,

$$R_s = S_d / (S_i - S_h) = (d_d / d_i)^2 / \{1 - (d_h / d_i)^2\}$$
(5)

where d_i is the inner diameter of the column. The gas velocity u_g has little effect on N_{eq} except at u_g closer to zero, similarly to the findings of Kusunoki *et al.*²⁾.

In the vibrating speed range of this experiment, the gas-liquid mixture in each stage has been observed to be divided into two parts by the vibrating disk. This result implied that the vibrating disk operates not only as an agitating device but also as a partition plate.

From these results, a back-flow model consisting of N equi-volume perfect mixers in series can be expected to express the liquid-phase mixing characteristics of the column. N is equal to twice the number of actual stages.

The rate of the back flow through the annulus between the disk edge and the column wall may be considered to be different from that of the back flow through the hole in the partition plate in general. In this study, however, the sectional area of the annulus is of the same order as that of the hole (0.56 to 6.2 in the ratio), and the same back flow rate throughout the column is assumed for both the annulus part and the partition plate hole. In this model, a decrease of N_{eq} can be attributed to an increase of the back flow rate F' and the model parameter, the back flow ratio α , is usually defined by Eq.(6):

$$\alpha = F'/F \tag{6}$$

where F is the main liquid flow rate. The relation between α and the variance σ^2 has been theoretically given by Eq.(7)^{1,6}:

$$\sigma^{2} = \frac{2}{\theta^{2}} \left[\theta - \left(1 - \frac{\theta^{2}}{4N^{2}} \right) \left\{ 1 - \left(\frac{2N - \theta}{2N + \theta} \right)^{N} \right\} \right]$$
(7)



where

$$\theta = 2N/(1+2\alpha)$$

The back flow ratios for the various experimental conditions have been calculated by Eq.(7) with N=14and the observed variances substituted. The step responses of this model have been evaluated by the analytical method presented by Roemer and Durbin⁶⁾. In **Fig. 4**, the observed responses are compared with those of the model, and good agreement between them is obtained as expected.

The back flow is caused by both the disk vibration and the gas flow, and its flow rate F' may be expressed by Eq.(8):

$$F' = F'_v + F'_g \tag{8}$$

where F'_v is the rate of the back flow produced by the disk vibration and F'_g by the gas flow. F'_v can be estimated from the back-flow ratio $\alpha_v (=F'_v/F)$ for the liquid system $(u_g=0)$. In **Fig. 5**, the back flow ratios $(\alpha = F'/F)$ for the gas-liquid systems (the cases of $u_g \neq 0$) are plotted against the corresponding ratios α_v for the liquid systems with the other experimental conditions unchanged. It is clear from this figure that α_v is approximately equal to α_g in the range of the present experimental conditions unless the back flow ratio is less than 0.1:

$$\alpha_v = \alpha_g = \alpha/2 \tag{9}$$

The wave forms of the disk vibration are observed



2.0

Fig. 7 Assumed wave form of the vibrating disk

by means of a motion picture (64 frames per second of a quarter size film—16 mm—) to estimate the amount of F'_v . The results are shown in **Fig. 6**. As is easily seen from the figure, a trapezoidal wave may be a good approximation to the wave form observed. In the following analysis, therefore, the wave form is assumed as shown in **Fig. 7**. This wave form becomes square at $\beta = 1/4\nu$ and triangular at $\beta = 0$. Then the velocity v of the vibrating disk (shown by the dotted lines in Fig. 7) is given by Eq.(10):

$$v = \frac{dx}{dt} = \begin{cases} \frac{A}{l} ; \begin{pmatrix} 0 \le t \le l \\ 1/\nu - l \le t \le 1/\nu \\ 0 ; \begin{pmatrix} l < t < 2\beta + l \\ 1/\nu - (l + 2\beta) < t < 1/\nu - l \\ -\frac{A}{l}; \begin{pmatrix} 1/2\nu - l \le t \le 1/2\nu \\ 1/2\nu \le t \le 1/2\nu + l \\ (l = 1/4\nu - \beta) \end{cases}$$
(10)

where x is the displacement of the disk from its neutral position (shown by the solid lines in Fig. 7). The back flow rate F'_{v} can be expressed by Eq.(11):

$$F_{v} = -\eta \nu \int_{-1/4\nu}^{1/4\nu} (u_{l} - v) S_{d} \varepsilon dt$$
 (11)

where u'_i is the linear velocity of the liquid phase through the hole in the partition plate and η is the effectiveness factor which is given by the fraction of the back flow reaching the adjoining stage to the back flow caused by the disk vibration. The factor η can be assumed to be the ratio S_h/S_i . Substitution of Eq.(10) into Eq.(11) gives the back flow ratio α_{η} :

$$\alpha_{v} = \frac{F_{v}'}{F} = \frac{F_{v}'}{u_{l}S_{i}} = \left(-\frac{1}{2} + 2\nu\beta + \frac{2A\nu\varepsilon}{u_{l}}\frac{S_{h}}{S_{i}}\right)\frac{S_{d}}{S_{i}}$$
$$= \left\{-\frac{1}{2} + 2\nu\beta + \frac{2A\nu\varepsilon}{u_{l}}\left(\frac{d_{h}}{d_{i}}\right)^{2}\right\}\left(\frac{d_{d}}{d_{i}}\right)^{2}$$
(12)

The correlation between the observed ratio α_v and the dimensionless parameter, $(2A_{\nu\varepsilon}/u_l)(d_h/d_i)^2$, are shown in **Fig. 8** for the case of $\nu = 100$ cpm and in **Fig. 9** for the case of 200 cpm. In these figures, the solid lines are based on Eq.(12). The agreement is fairly good, and the case of 200 cpm is better than that of 100 cpm. It is noted, however, that Eq.(12) may not be valid at small values of $(2A_{\nu\varepsilon}/u_l)(d_h/d_i)^2$ for the gas-liquid systems $(u_g \neq 0)$ where the back flow ratio never approaches zero.

Conclusion

The liquid-phase mixing characteristics of the multistage vibrating-disk column with concurrent gas-liquid flow have been investigated by means of the step response method with electrolytically generated bromine tracer, and the following conclusions have been obtained:

1) The mixing characteristics can be expressed by the back flow model which consists of equi-volume perfect mixers of twice the number of actual stages.

2) The back flow ratio α is considerably influenced by the hole diameter d_h of the partition plate and is slightly affected by the vibrating frequency of the disk ν and the liquid velocity u_l . The gas velocity u_g has little effect on the ratio except at u_g closer to zero.

3) The effects of the vibration and the gas flow on the back flow ratio are nearly the same within the present experimental conditions.

4) The back flow ratio ascribed to the disk vibration α_v , can be predicted by the following dimensionless equation:

$$\alpha_v \left(\frac{d_i}{d_d}\right)^2 = \Psi \left\{ \frac{2A\nu\varepsilon}{u_l} \left(\frac{d_h}{d_i}\right)^2 \right\}$$

where the function Ψ is dependent on the wave form of the disk vibration.

5) The electrolytic generation of a tracer is proved to be useful for the flow model identification.

Acknowledgment

All the numerical calculations were carried out at the Computer Center, University of Osaka Prefecture.

Nomenclature

			T
A	= amplitude of vibration	[cm]	
d_d	= diameter of vibrating disk	[cm]	u_{α}
d_h	= diameter of hole in partition plate	[cm]	u_l
d_i	= inner diameter of column	[cm]	u,
f(t)	= tracer step response	[]	ı
F	= volumetric main liquid flow rate	[cm ³ /sec]	V
F'	$= F'_v + F'_q$, back flow rate	[cm ³ /sec]	v
F'_q	= rate of back flow produced by gas flow	[cm ³ /sec]	x
F'_{v}	= rate of back flow produced by vibration	[cm ³ /sec]	
l	= defined in Fig. 7	[sec]	
Ν	= number of equi-volume perfect mixers in		α
	series in the back flow model	[]	α_g
N_{eq}	= number of equivalent stages of perfect		α_v
	mixers in series, defined by Eq. (4)	[—]	β
R_s	= ratio of the area of the disk to that of		ε



Fig. 8 Observed back flow ratios in comparison with the predicted ones by Eq.(12)



Fig. 9 Observed back flow ratios in comparison with the predicted ones by Eq.(12)

 S_d

 S_h

 S_i $\frac{t}{t}$

 t^2

	the partition plate, defined by Eq.(5)	[—]
=	$\pi d_d^2/4$, area of the disk	$[cm^2]$
	$\pi d_h^2/4$, sectional area of the hole in the	
	partition plate	$[cm^2]$
=	$\pi d_i^2/4$, sectional area of the column	$[cm^2]$
=	time	[sec]
	mean residence time	[sec]
=	squared mean time, defined by Eq. (3)	[sec ²]
=	the time at which $f(t)$ can be regarded as	
	being steady	[sec]
	gas velocity (superficial)	[cm/sec]
=	liquid velocity (superficial)	[cm/sec]
=	linear velocity through the partition plate	
	hole	[cm/sec]
-	effective volume of the column	[cm ³]
	velocity of the vibrating disk	[cm/sec]
• =	displacement of the vibrating disk from its	
	neutral position	[cm]
_	F'/F, back flow ratio	[—]
=	F'_{a}/F , back flow ratio caused by gas flow	[]
=	F'/F, back flow ratio caused by vibration	[]
=	defined in Fig. 7	[sec]
=	liquid holdup	[]
	1 1	

η	= effectiveness factor in Eq.(11)	[—]
ν ν	= vibrating frequency of disk	[sec ⁻¹]
σ^2	= variance of the residence time distribution,	
	defined by $Eq.(2)$	[]
τ	= reduced time $(=t/\overline{t})$	[—]

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EFFECT OF MASS TRANSFER ON THE SELECTIVITY OF (m, n)-(p, q) ORDER CONSECUTIVE GAS-LIQUID REACTIONS*

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Theoretical analysis of the selectivity of (m, n)-(p, q) order consecutive gas-liquid reaction is developed based on the film theory. The reaction scheme discussed is as follows: $B(l)+A(g)\rightarrow R(l)$, rate= $k_1A^mB^n$; $R(l)+A(g)\rightarrow S(l)$, rate= $k_2A^pR^q$. The relations between the yield of an intermediate product R and the conversion of B are numerically calculated for a continuous stirred-tank reactor, and it is shown that Hikita and Asai's approximate method is satisfactorily extended to this consecutive reaction system. The yield-conversion relations for (m, n)-(m, q) order reaction (i.e. m=p) are closely approximated by those of (1, n)-(1, q) order reaction using a given generalized parameter h_1 which corresponds to the Thiele modulus in solid-catalysed gas-phase reactions. In the case of $m \neq p$, the effect of diffusion appears even in the slow reaction regime, whereas in the case of m=p, diffusion limitation influences the yield only when the reaction in the liquid film is predominant. The reaction factor for (m, n)-(p, q)order reaction.

Introduction

Recently several theoretical and experimental studies^{1-3,5,6,8,10-13}) have been presented on the selectivity of consecutive gas-liquid reactions, and it was found that the yield of an intermediate product was lowered by the effect of diffusion limitation in the liquid film. In these studies, however, the reactions analysed were zero or first order with respect to the gaseous reactant and first order with respect to the liquid reactant.

In this paper, the theoretical studies of the present authors are extended to consecutive reactions of general order. Both numerical and approximate solutions for yield vs. conversion and reaction time vs. conversion

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The reaction factor for this reaction system is also presented.

1. Basic Equation

The consecutive reaction considered is

$$B(l) + A(g) \rightarrow R(l), \qquad R_1 = k_1 A^m B^n \tag{1}$$

$$R(l) + A(g) \rightarrow S(l), \qquad R_2 = k_2 A^p R^q$$
(2)

where A and B are feeds, R is a desired intermediate product and S is an undesired final product. Here it is assumed that reaction (1) is *m*-th order with respect to gaseous reactant A and *n*-th order with respect to B, while reaction (2) is *p*-th and *q*-th order with respect to A and R, respectively. Hereafter, reactions (1) and (2) are referred to as an (m, n)-(p, q) order reaction.

Differential material balance for each species in the liquid film gives the equations

^{*} Received on June 11, 1973

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