

## DROP SIZE DISTRIBUTION AND HOLDUP IN A ROTATING IMPELLER EXTRACTION COLUMN

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**Abstract**—A stagewise hydrodynamic model, applying drop population balance equations derived from models for breakage and coalescence of drops in a countercurrent liquid-liquid extraction system, was developed to predict the drop size distribution and the holdup of the dispersed phase in a rotating impeller extraction column. The drop size distributions were obtained by taking the photographs of the dispersions at the same locations through the rectangular shaped glass box filled with distilled water. The experimental variables were the impeller speed and flow rates of the continuous and dispersed phases. The solutions of the model equations were obtained by performing the computer simulation and the optimum parameter values were determined. The results predicted by the model were in good agreement with the experimental results obtained from the present rotating impeller extraction column.

### INTRODUCTION

Liquid-liquid extraction is an important separation operation which is much used now in the field of hydrometallurgy, nuclear fuel manufacture and reprocessing, pharmaceutical industry and prevention of water pollution, e.g., the removal of phenol from effluents, after the early days of limited application in the oil industry. In a number of extraction operation several types of equipments are employed, such as rotating impeller columns, rotating disc columns, reciprocating plate columns, pulse columns, spray columns, and constant flow stirred tank contactors. In these experiments, liquid phases of limited mutual solubility are contacted by dispersing one liquid phase in the other in the form of droplets. A relatively large specific contacting area of phases is required for the high efficiency of the countercurrent liquid-liquid extractor. A rational design of the dispersed phase contacting equipment should be based on the knowledge of the drop size distributions and the dynamics of the drop.

The drop size distribution is generally a function of time and position and can be considered as a result of breakage and coalescence. As the rate of this process is usually comparable with the rate of drop transport, consequently the drop size distribution and the dispersed phase holdup may vary along the liquid of the column. At a steady state condition the number

of the generation and disappearance of drops reaches equilibrium statistically. The breakage and coalescence of the drops greatly affect the interfacial area, concentration driving forces, and the extraction efficiencies. The mass transfer rate has been obtained mainly from the data based on the above and can be calculated by introducing the drop population balance equation.

The objectives of this work are to develop more adequate models of the hydrodynamics, applying drop population balance equations for predicting the drop size distribution in a countercurrent liquid-liquid extraction column and, to obtain the experimental data on a rotating impeller column, in order to compare the calculated results with the experimental data.

### THEORY

#### 1. Hydrodynamics

The basic hydrodynamic equation for liquid-liquid counter current flow, first applied to a pulsed packed column by Gayler, Roberts and Pratt [1], was defined as follows:

$$\frac{U_c}{\alpha(1-\phi)} + \frac{U_d}{\alpha\phi} = U_s \quad (1)$$

where  $U_c$  is the continuous phase velocity,  $U_d$  is the dispersed phase velocity,  $U_s$  is the slip velocity and

$\phi$  is the dispersed phase holdup. To account for the true countercurrent velocities of the continuous and dispersed phase at the planes of minimum cross sectional area in a column, an equipment factor  $C_R$ , sometimes called the constriction factor is introduced instead of  $\alpha$  [2]. Thus the basic hydrodynamic equation is expressed in terms of the overall phase flow rates,

$$\frac{U_c}{C_R(1-\phi)} + \frac{U_D}{C_R\phi} = U_S \quad (2)$$

According to Chartres and Korchinsky's assumption [3] the basic hydrodynamic equation can be applied to individual drop size fractions of size,  $d$ , and a form of that equation utilized is

$$\frac{U_c}{C_R(1-\phi)} - U_{D,i} = U_{S,i} \quad (3)$$

where  $U_{D,i}$  is the rise (or fall) velocity, relative to the column, and  $U_{S,i}$  is the velocity of the same drop (with diameter  $d_i$ ) relative to the continuous phase, in a dispersion with other drops of varying size.

If the dispersed phase static volume fraction of drops of size  $d_i$  is  $f_i$ , then equation (3) may be summed over all drop size fractions by Cruz-Pinto and Korchinsky [4], thus

$$\frac{U_D}{\phi} = \sum_i f_i C_R U_{D,i} = C_R \left[ \sum_i f_i U_{S,i} - \frac{U_c}{C_R(1-\phi)} \right] \quad (4)$$

or, upon arrangement

$$(C_R \sum_i f_i U_{S,i})^2 + (U_c - U_D - C_R \sum_i f_i U_{S,i})\phi + U_D = 0 \quad (5)$$

This equation may be solved iteratively for the holdup, flow rates, and drop size distribution data.

Real column conditions are far from stagnant, and to account for it the slip velocity should be defined as a function of holdup and a characteristic velocity  $U_K$ . Olney [5] and Rod [6] emphasized the polydispersed nature of dispersion when considering drop hydrodynamics. Misek and Marek [7] developed the theory accounting for coalescence effects. The introduction of drop size distribution into  $U_K$  is made on the assumption that all drops moved countercurrently to the continuous phase. In this work Misek's correlations for the slip and terminal velocity are used.

## 2. Drop Population Balance Equations

Presently modelling of countercurrent liquid-liquid extraction columns has proceeded from two basic models:

- (1) a differential model, with plug flow of the phases through the column; and
- (2) a stagewise model, with flows completely mixed stages.

In this work a modified stagewise column model is considered and it is assumed that a section of this column is regarded as a CSTR and within each section an isotropic field of turbulence applies. It is further assumed that, within each section, the dispersed phase holdup is uniform. In this column, the above assumptions will be applied to each of the sections although at least there must exist some non-uniformities. Then a set of unsteady state nonlinear integro-differential equation is obtained:

$$\begin{aligned} \frac{d[N(t)A(v,t)]}{dt} + N(t)A(v,t)[f(v) + g(v) + w(v,t)] \\ = N(t)A(v,t)f(v) + \int_0^{v_{max}} \beta(v',v)v(v')A(v,t)dv' + \int_0^{v/2} \\ \lambda(v-v',v')h(v-v',v')N(t)A(v-v',t)N(t)A(v',t)dv' \end{aligned} \quad (6)$$

where

$$w(v,t) = \int_0^{v_{max}-v} \lambda(v,v')h(v,v')N(t)A(v',t)dv' \quad (7)$$

At steady state, equation(6) reduces to

$$\begin{aligned} NA(v) = [n_0 a(v) + \int_v^{v_{max}} \lambda(v',v)v(v')g(v')NA(v')dv' \\ + \int_0^{v/2} \lambda(v-v',v')h(v-v',v')NA(v-v') \\ NA(v')dv'] / [f(v) + g(v) + w(v)] \end{aligned} \quad (8)$$

where

$$w(v) = \int_0^{v_{max}-v} \lambda(v,v')h(v,v')NA(v')dv' \quad (9)$$

and  $f$  is the escaping frequency,  $g$  is the breakage frequency,  $w$  is the coalescence frequency,  $NA$  is the drop size distribution in a section and  $n_0 a$  is the drop size distribution in the feed.

The various functions appearing in the above equations should be obtained before the solution is made. In this work the four functions introduced by Coulaloglou and Tavlarides [8] with assumption of  $v=2$  will be used. They are

$$g(v) = C_1 v^{-2.9} D^{*2/3} \frac{N^*}{1+\phi} \exp\left[-\frac{C_2 \phi(1+\phi)^2}{\rho_D v^{5.9} D^{*4.3} N^{*2}}\right] \quad (10)$$

$$\beta(v,v') = \frac{2C_3}{v'\sqrt{2\pi}} \exp\left[-\frac{(2v-v')^2 C_3^2}{2v'^3}\right] \quad (11)$$

$$h(v,v') = C_4 (v'^3 + v'^{2.3}) (v^{2.9} + v'^{2.9})^{1.2} \frac{N^*}{1+\phi} \quad (12)$$

$$\lambda(v,v') = \exp\left[-\frac{C_5 \mu_c \rho_c D^{*2}}{\sigma^2}\right] \frac{N^*}{(1+\phi)^3} \frac{v^{1.3} v'^{1.3}}{v^{1.3} + v'^{1.3}} \quad (13)$$

where  $g$  is the breakage frequency,  $\beta$  is the breakage distribution function,  $h$  is the collision frequency and  $\lambda$  is the coalescence efficiency of drops.

The constants  $C_1$ – $C_5$  in equations (10)–(13) are parameters to be obtained, which were lumped into in deriving the four functions which can be found in the literature [8]. Some authors [9] took the value of the constant  $C_3$  to be 3.0 for convenience and for comparing the experimental results of CSTR and others with those of their own equipment. In this work the value of constant  $C_3$  was fixed to 3.0 in Table 5 for the same reason.

It is considered that Coulaloglou and Tavlardides' four functions, equations (10)–(13) can not be applied to other dispersion systems without changing the values of the five parameters, because every system has different characteristics in the apparatus such as geometry and thickness of baffles, and size and shape of impeller, etc.. Thus optimum values must be found for the five parameters to obtain minimum differences between the experimental and calculated results. For this purpose a nonlinear optimization program is used [10]. Analytical solutions of the integral equation are not known and a numerical solution can be obtained by reduction with an integration formula as proposed by Valentas et al. [11].

## EXPERIMENTAL EQUIPMENT AND PROCEDURE

### 1. Experimental Equipment

A countercurrent rotating impeller column of conventional arrangement was designed [12,13] and used to obtain drop size data under various conditions. The overall schematic system for the experiment is shown in Fig. 1. The heavy phase and the light phase are fed to the top and the bottom of the column respectively from the head tanks equipped with the overflow outlet to maintain the liquid level constant. The continuous phase outlet is equipped with an adjustable limb to maintain a constant interface level. Likewise the dispersed phase outlet is equipped with an overflow. The rotating unit is connected to top of the main column. The solenoid valves are installed at the inlet and outlet of the column in order to stop the flows simultaneously.

The column inside diameter is 6.5 cm, and the working height is 210 cm. For most of the runs, the same layout of the column intervals was maintained, i.e. 30 compartments, each 7 cm height. The top and bottom sections of the main column are made of 13.5 cm inside diameter lucite column segments of 23 cm and 27 cm length respectively. The main column is provided with sampling ports at 50 cm intervals for measuring the local holdup. The organic phase was introduced

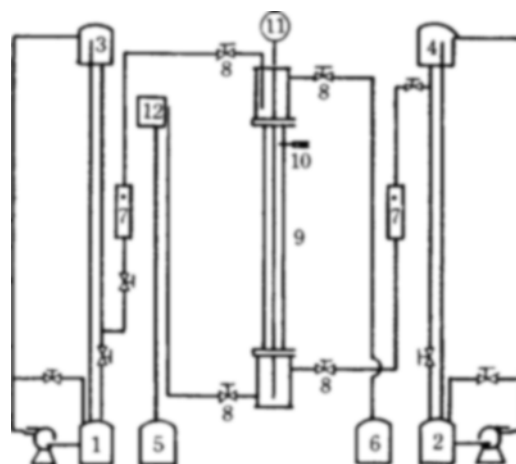


Fig. 1. Schematic flow diagram.

- |                      |                      |
|----------------------|----------------------|
| 1. Aqueous storage   | 2. Organic storage   |
| 3. Aqueous head tank | 4. Organic head tank |
| 5. Aqueous reservoir | 6. Organic reservoir |
| 7. Rotameter         | 8. Solenoid valve    |
| 9. Main column       | 10. Sampling port    |
| 11. D.C motor        | 12. Level adjuster   |

ed through a cylindrical distributor located centrally in the column and in close proximity to the first compartment. The organic phase distributor plates made of stainless steel contained 80 holes of diameter 2.4 mm. The opening diameter of distributor is 2 cm.

The doughnut plates and rotating impeller are made of 1 mm thick SUS 304 stainless steel plates. The opening diameter of the doughnut plate is 4.5 cm and the impeller diameter is 4.4 cm. The doughnut type plates are supported in the main column by means of three 3.5 mm diameter 220 cm length rod. Stainless steel sleeves around the rod serve as spacers for the plates. The spacer plays a role to adjust compartment height. The impellers are fixed by a 10 mm diameter 296 cm length stainless steel rod. Four 101410 little bearings are installed so as not to vibrate the rod. The entire rod assembly can be set by lifting it from the open top of the column. The shaft rod is driven by pulley attached to an RPM adjuster driven by a 1/4 hp 110 volt DC motor. The RPM could be adjusted by the tachometer. Drop size data were obtained photographically in this work.

### 2. Experimental Procedure

The main aim of the experimental procedure was to obtain drop size distribution data and holdup in a countercurrent extraction column. Experiments were performed with odorless kerosene as dispersed

**Table 1. Description of the column**

Column	Dimension
Working height	210 cm
Inside diameter	6.5 cm
Opening diameter of stator	4.5 cm
Impeller diameter	4.4 cm

**Table 2. Physical properties of the system**

Properties	Value
Viscosity of dispersed phase	$\mu_D: 0.011 \text{ g cm}^{-1} \text{ sec}^{-1}$
Density of dispersed phase	$\rho_D: 0.805 \text{ g cm}^{-3}$
Density of continuous phase	$\mu_C: 1.000 \text{ g cm}^{-3}$
Interfacial tension	$\sigma: 28.20 \text{ dyne cm}^{-1}$

**Table 3. Experimental parameters**

Parameters	Range
Impeller speed( $N^*$ )	180-340(rpm)
Compartment height(H)	7(cm)
Velocity of water( $U_C$ )	7.42-28.60( $\text{cm min}^{-1}$ )
Velocity of kerosene( $U_D$ )	7.42-25.64( $\text{cm min}^{-1}$ )

phase and distilled water as continuous phase. Description of the column and the physical properties of the system are listed in Tables 1 and 2 respectively.

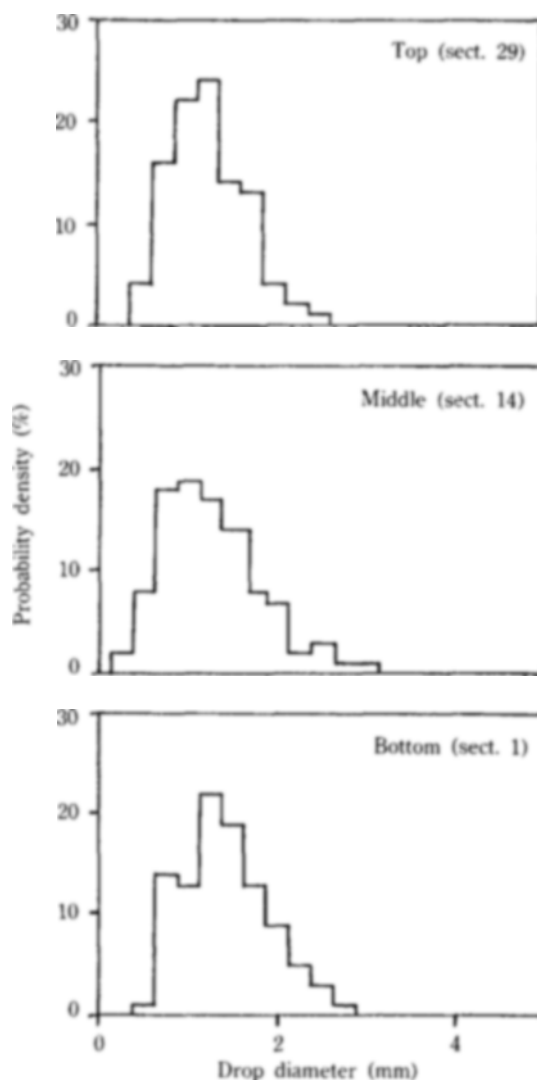
The experimental column is initially filled with distilled water and the water flow rate was set at the desired level. The agitator was then started and its speed adjusted at a proper RPM. With the organic outlet line from the column open, the organic feed pump was started and the rate was increased gradually to the desired level. The agitator speed was checked at regular intervals using a tachometer. Periodically, the flow rates were checked during the course of each experiment. The column was operated at the experimental settings until steady state conditions were established. Experimental conditions are listed in Table 3.

Holdup measurements were made when two phases have been completely separated after the solenoid valves were shut off in order to stop the flows. Local holdup was measured for three selected sections (1, 14, 29). The sample probes were positioned equidistantly between the edge of the disc and the column wall. The holdup was determined from the volume composition of the two-phase samples.

## RESULTS AND DISCUSSIONS

### 1. Experimental

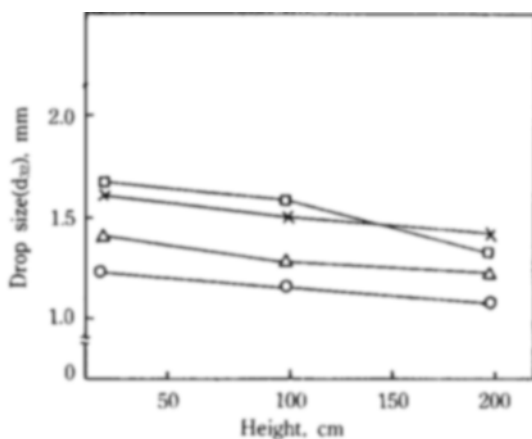
Measurements of drop size distribution in a rotating



**Fig. 2. Drop size distribution at  $N^* = 180 \text{ rpm}$ ,  $U_C = 14.53 \text{ cm/min}$ ,  $U_D = 14.53 \text{ cm/min}$ .**

impeller extraction column was performed for the various experimental conditions listed in Table 3. In order to avoid the effect of the interfacial mass transfer due to mutual solubility of two phases, mutually saturated phases were used. To achieve this, two phases were recycled through the recycle system of the apparatus.

Fig. 2 shows the typical drop size distribution of three sections of this column at a impeller speed and dispersed and continuous phase flow rates, which were compared with the calculated results. It was found that the drop size distribution in a rotating impeller extraction column shows a unimodal form and



Symbol	RPM	$U_c$ (CPM)	$U_l$ (CPM)
○	260	14.53	14.53
△	260	14.53	25.64
×	260	24.51	17.99
□	260	24.51	25.64

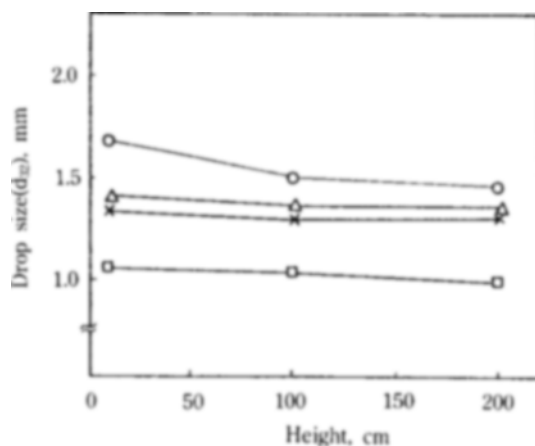
Fig. 3. Effect of the phase flows on the drop size.

the drop size gradually decreases as the column height increases under the same operating condition. This is believed to occur because the drops further break up by agitation as they move up the column while the dynamic equilibrium between the breakage and the coalescence is maintained. It is also observed that the skewed profile of the drop size distribution increases and the variance decreases at higher impeller speeds.

The effects of the column height and the phase flows on the drop size distribution are shown in Fig. 3. The drop size increases, with increasing the dispersed phase flows and decrease as the column height increases. This trend is in accordance with the previous data for the rotating disc contactor [8,9].

An increase in impeller speed from 180 to 340 rpm caused a considerable decrease in the drop size. While the variation of phase flows affect the drop size a little, the impeller speed plays an important role for the drop size determination. Fig. 4 shows the effect of the impeller speed on the drop size.

The overall dispersed phase holdup in the column was measured in each case by shutting off the flows activating the solenoid valve in the feed. The typical plots of the dispersed phase holdup data with the dispersed phase flow rate and the continuous phase flow rate are shown in Fig. 5. It was found that the holdup of the dispersed phase depends largely on the dispersed phase flow rate. Local holdup was measured by



Symbol	RPM	$U_c$ (CPM)	$U_l$ (CPM)
○	180	14.53	17.09
△	260	14.53	17.99
×	300	14.53	17.99
□	340	14.53	14.53

Fig. 4. Effect of the impeller speed on the drop size.

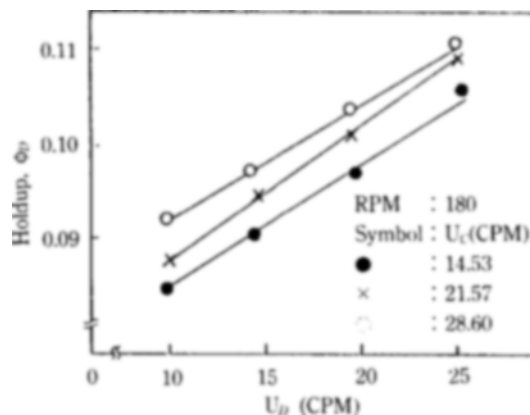


Fig. 5. Variation of the dispersed phase holdup with respect to the dispersed phase flow rate.

direct sampling. Fig. 6 shows the experimental data for local holdup at a impeller speed and various dispersed and continuous phase flow rates.

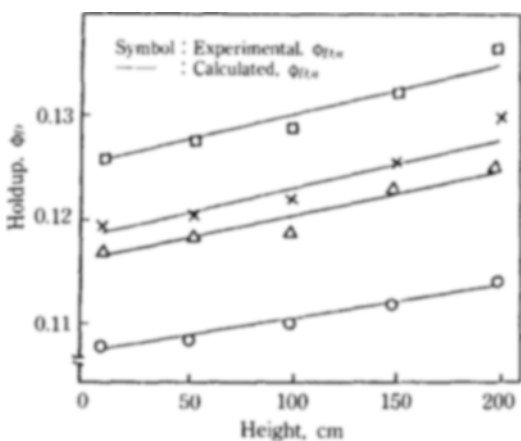
## 2. Theoretical

Equation(5) was iteratively solved to determine the dispersed phase holdup and the column constriction factor. Flow rates, overall holdup, and local drop size distribution data were used for the calculation.

Using the local drop size distributions obtained experimentally and the overall dispersed phase holdup measured, the local holdups with column height were predicted. The calculated holdups were subject to the

**Table 4. Predicted values for the local holdup and column constriction factor**

RPM	$U_c$ (CPM)	$U_D$ (CPM)	Drop diameter $d_{32}$ (mm)		Holdup		Constriction factor $C_R$
			SEC. 1	SEC. 29	SEC. 1	SEC. 29	
180	14.53	14.53	1.74	1.52	0.085	0.091	0.148
260	14.53	17.09	2.18	1.65	0.105	0.115	0.094
300	14.53	17.99	1.76	1.40	0.132	0.140	0.088
180	14.53	17.09	2.17	1.64	0.091	0.093	0.172
260	14.53	25.63	2.11	1.96	0.120	0.124	0.140
300	24.51	17.99	2.01	1.87	0.135	0.142	0.092
180	24.51	25.64	2.28	2.02	0.100	0.102	0.240
260	24.51	17.99	2.21	1.99	0.121	0.126	0.144
340	24.51	17.99	2.14	1.80	0.136	0.144	0.104



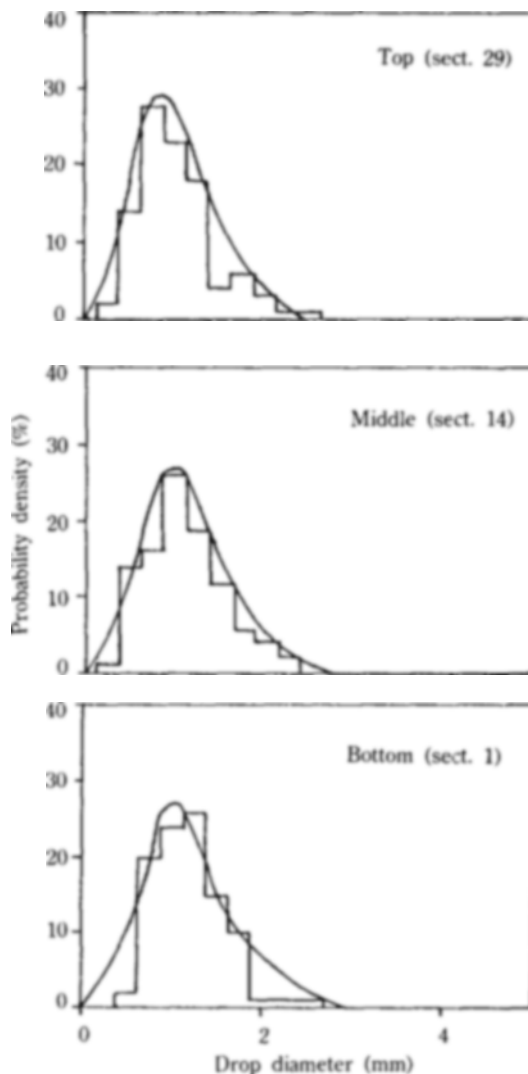
Symbol	RPM	$U_c$ (CPM)	$U_D$ (CPM)
○	260	14.53	14.53
△	260	14.53	17.09
×	260	24.51	17.99
□	260	24.51	25.64

**Fig. 6. Axial distribution of holdup at  $N^* = 260$  rpm.**

constraint that the overall predicted value of the holdup agreed with the experimental average value. The typical data for the calculated holdups are shown in Fig. 6.

The values of the constriction factor  $C_R$  were calculated using the hydrodynamic model and are listed in Table 4, along with the local holdup values. The results provide further support for the present approach employed to correlated the dispersed phase holdup with the flow rate, the drop size, and the column geometry.

The basic drop population balance equation (8) was applied to the dispersion at each column section. The drop size distribution and the average drop size( $d_{32}$ ) were calculated. The experimental data and the pre-

**Fig. 7. Drop size distribution at  $N^* = 260$  rpm,  $U_c = 14.53$  cm/min,  $U_D = 17.09$  cm/min.**

**Table 5. Values of parameters obtained from optimization**

Operating conditions	Parameter values
180 RPM	$C_1 = 0.24316E+0$
$U_C = 14.53$ CPM	$C_2 = 0.54165E-2$
$U_D = 14.53$ CPM	$C_3 = 3.0$
	$C_4 = 0.49307E-3$
	$C_5 = 0.30888E+7$
260 RPM	$C_1 = 0.11640E+0$
$U_C = 14.53$ CPM	$C_2 = 0.20009E-2$
$U_D = 17.09$ CPM	$C_3 = 3.0$
	$C_4 = 0.49207E-3$
	$C_5 = 0.30888E+7$
340 RPM	$C_1 = 0.83649E-1$
$U_C = 14.53$ CPM	$C_2 = 0.11051E-2$
$U_D = 24.51$ CPM	$C_3 = 3.0$
	$C_4 = 0.49207E-3$
	$C_5 = 0.30888E+7$

dicted values with the parameters obtained for each operation are in good agreement, which are illustrated in Fig. 7. The parameters are listed in Table 5.

If the stagewise hydrodynamic model used in this work were accurate, the value of the parameters,  $C_1$  through  $C_5$  should be a set of unique optimum values under varying operating conditions. But the parameters obtained by optimization in the present work show a minor dependency on the operating conditions as indicated in Table 5. Errors in measuring the flow rates, local and overall holdups and drop size distributions which are expected must have partly contributed to the parameter variations. Besides assuming binary, and homogeneous, drop breakage and coalescence, the model used in this work was developed for a local isotropic turbulent field, where drop breakage and coalescence were assumed to be the results of turbulent fluctuation at the drop surface. It may also contribute to the same effects. Moreover the possibility that the basic hydrodynamics and drop population balance equations do not accurately represent the experimental behaviour of the dispersion, may not be neglected and leaves a subject for further study. However at the present stage, as direct experimental data for the rotating impeller column are not available, this model can be useful to predict the holdup and the drop size distribution.

## CONCLUSIONS

A stagewise hydrodynamic model, applying drop population balance equations derived from models for

breakage and coalescence of drops in a countercurrent liquid-liquid extraction system, was developed to predict the drop size distribution and the holdup of the dispersed phase in a rotating impeller extraction column. To obtain the solution of a set of nonlinear integro-differential equations, computer simulation was performed using the backward iteration scheme.

The holdup data and the drop size distribution of kerosene-water systems measured experimentally by using a 6.5 cm I.D. and 210 cm high rotating impeller column were found to be in good agreement with the theoretical ones, from which the optimum values of parameter in the model equations were determined.

## NOMENCLATURE

- $A_0(v,t)dt$  : number fraction of drops in the feed with volume between  $v$  and  $v+dv$  at time  $t$   
 $A(v,t)dv$  : number fraction of drops in a section  
 $C_1-C_5$  : parameters in Equation (10) to Equation (13)  
 $C_k$  : constriction factor  
 $d$  : drop diameter [cm]  
 $d_{32}$  : sauter mean drop diameter [cm]  
 $D^*$  : impeller diameter [cm]  
 $f(v)$  : escape frequency i.e. fraction of drops with volume between  $v$  and  $v+dv$  flowing out per unit time [ $\text{sec}^{-1}$ ]  
 $g(v)$  : breakage frequency of drop of volume  $v$   
 $h(v)$  : collision frequency of drops of volume  $v$  and  $v'$  [ $\text{sec}^{-1}$ ]  
 $N_0(t)$  : number rate of drops in the feed [ $\text{sec}^{-1}$ ]  
 $N^*$  : impeller speed [ $\text{sec}^{-1}$ ]  
 $N(t)$  : number of drops at time  $t$   
 $t$  : time [sec]  
 $v, v'$  : drop volume [ $\text{cm}^3$ ]  
 $U_C$  : superficial velocity of continuous phase [ $\text{cm sec}^{-1}$ ]  
 $U_D$  : superficial velocity of dispersed phase [ $\text{cm sec}^{-1}$ ]  
 $U_K$  : characteristic velocity [ $\text{cm sec}^{-1}$ ]  
 $U_S$  : slip velocity [ $\text{cm sec}^{-1}$ ]

## Greek Letters

- $\alpha$  : porosity  
 $\beta(v, v')$  : breakage distribution function defined by Equation (11)  
 $\gamma$  : numerical constant depending on the order of integration  
 $\delta$  : numerical constant depending on the order of integration

$\epsilon$	: energy dissipation per unit mass [erg/g/sec]
$\zeta$	: numerical constant depending on the order of integration
$\xi$	: numerical constant depending on the order of integration
$\lambda(v,v')$	: coalescence efficiency of drop of volume $v$ with a drop of volume $v'$
$\mu$	: viscosity [poise]
$\nu$	: kinematic viscosity [cm sec <sup>-1</sup> ]
$\rho$	: interfacial tension [dyne cm <sup>-1</sup> ]
$\tau$	: residence time [sec]
$\phi$	: dispersed phase holdup fraction

### Subscripts

C	: continuous phase
D	: dispersed phase
i	: species

### REFERENCES

1. Gayler, R., Roberts, N.W. and Pratt, H.R.C.: *Trans. Instn. Chem. Engrs.*, **31**, 57 (1953).
2. Strand, C.P., Olney, R.B. and Ackerman, G.H.: *AIChE J.*, **8**, 252 (1962).
3. Chartres, R.H. and Korchinsky, W.J.: *Trans. Instn. Chem. Engrs.*, **53**, 247 (1975).
4. Cruz-Pinto, J.J.C. and Korchinsky, W.J.: *Chem. Engr. Sci.*, **36**, 687 (1981).
5. Olney, R.B.: *AIChE J.*, **10**, 827 (1964).
6. Rod, V.: *Brit. Chem. Engr.*, **16**, 617 (1971).
7. Misek, T. and Marek, J.: *Brit. Chem. Engr.*, **15**, 202 (1972).
8. Coulaloglou, C.A. and Tavlarides, L.L.: *Chem. Engr. Sci.*, **32**, 1289 (1977).
9. Cruz-Pinto, J.J.C. and Korchinsky, W.J.: *Chem. Engr. Sci.*, **35**, 2213 (1980).
10. Jeon, Y.M.: Ph.D. Thesis, KAIST, Seoul Korea (1984).
11. Valentas, K.J., Bilous, O. and Amundson, N.R.: *I & EC Fund.*, **5**, 271 (1966).
12. Choi, Y.K.: MS Thesis, Ajou Univ., Suwon, Korea (1986).
13. Lee, Y.K. and Kim, C.: Proceedings of ISEC '90, in press (1991).