Simulation of Hydrodynamic Performance of Stirred Extraction Column

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The hydrodynamic behavior of countercurrent extractors is described by balancing numbers of incoming and outcoming drops against those formed and destroyed by breakage and coalescence. The laws for these processes are still poorly understood, and no reliable experimental data exist. Special empirical equations were, therefore, derived from measurements in a stirred cell. Using a simplified model, a computer program for simulating the variation of the drop-size distribution along the height of a pilot-plant sized stirred column solved the equations for all column stages and preselected characteristic drop volumes. A comprehensive set of holdup and drop-size distribution profiles measured earlier with the same column and o-xylene dispersed in water has been simulated, with the average mean deviation of local drop-size distributions not exceeding 20%. Transitional phenomena such as the startup time necessary for obtaining a new steady state after changing the operating parameters and the flooding were successfully predicted.

Introduction

The hydrodynamic performance of stagewise liquid-liquid extraction columns with intensive input of mechanical energy is characterized by a variation of the local holdup of the dispersed phase along the column height. The drops are not uniform but display size distribution with varying broadness and mean diameter caused by variation of the drop velocity with holdup and drop diameter, and by drop breakage and coalescence. Small drops are affected by the local flow pattern of the continuous phase, so they may accumulate in some part of the column or even flow in the opposite direction, that is, concurrently with the continuous phase. The investigation of these phenomena is of considerable importance as they directly influence both the capacity and the mass-transfer performance of any extraction column. Especially the prediction of the flooding conditions is virtually impossible without a detailed knowledge about the variation of the drop-size distribution along the column height.

For quantitative description of these phenomena, the drop population balances may be written for each of the column compartments in the form of differential or finite difference equations and solved by suitable means. The balances should consider that drops enter the agitated stages from both sides, and are formed and destroyed inside the stages by breakage and coalescence. They may also transfer a solute with the surrounding continuous phase. Coulaloglou and Tavlarides (1977) were the first ones who wrote a consistent set of such equations for single-stage processes. Jiřičný et al. (1979) extended the treatment to multistage extraction columns. Recently, several groups have developed consistent simulation procedures for multistage countercurrent processes, including among others Dimitrova et al. (1989), Tsouris et al. (1994), Kronberger et al. (1995), or Zamponi et al. (1996). However, they concentrated on the development of mathematical procedures, assuming that the laws controlling the fundamental processes of drop movement, breakage, coalescence and mass transfer, used as subroutines in the simulation programs, are available in sufficient quality from the literature.

The breakage and coalescence rates are usually calculated from phenomenological equations written originally by Coulaloglou and Tavlarides (1977), modified by Sovová (1981), and thoroughly updated by Tsouris and Tavlarides (1994). They define the breakage rate as the collision frequency of drops and eddies of the continuous phase multiplied by the breakage efficiency, and the coalescence rate as a product of drop-drop collision frequency and coalescence efficiency. Equations for all particular terms based on general turbulence considerations were proposed and modified by empirical parameters. However, observing the following phenom-

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ena during our work with stirred tanks, we could not confirm the general applicability of such equations:

- In a closed cell, cleaned with chromsulfuric acid and thoroughly rinsed by deionized water, the mean drop size increased for about a day, remained constant for the next day and decreased again, well under the original value. No changes in the organic phase (o-xylene) were observed on a gas chromatograph, and the cell was thermostated to ±0.2°C.
- Replacing toluene of analytical purity with a technical one changed the specific interfacial area of the dispersion by up to 20%. The changes in the physical properties, including the surface tension, were much smaller than would be needed to explain the changes in the drop-size distribution using the equations from the literature.
- Adding a single drop of sulfuric acid into a 1,000-mL stirred tank changed the specific interfacial area by up to 15%.
- A pilot-plant column changed its mass-transfer performance from 9 to 5 theoretical stages by replacing the organic phase by a new batch of the same solvent.
- Rounding and smoothing-up the edges of the baffles in a standard stirred tank decreased the mean Sauter diameter of the drops by about 10%.
- The equations of Coulaloglou and Tavlarides define the influence of the physical parameters explicitly but contain four free parameters to allow for specific properties of the given equipment. Testing these equations on our stirred-tank data with toluene dispersed in water, the average deviation of the mean drop diameter was 35%. Optimizing the parameters lowered the deviation to 18%, but using the optimized constants for the same tank and o-xylene in water, the difference increased to 46%.

The above observations indicate that the breakage and coalescence rates strongly depend on experimental arrangement, presence or absence of mass transfer, purity of the phases and further, still undefined, parameters. Given the complexity of the available equations and the necessity to evaluate the free parameters for each situation separately, the use of simpler empirical equations seems to be more advantageous. This article shows that simple power functions for the breakage and coalescence rates enable a realistic simulation if the free parameters are evaluated from experiments with the same system as used in the final operation.

A proper description of the variation of drop velocity with drop diameter, holdup, and stirring intensity is of extreme importance for a reliable simulation of the column performance. Except for operating regimes close to flooding conditions, the drop movement influences the holdup variation along the column axis more than the drop interactions. In the majority of the above mentioned simulation procedures the drop velocity is calculated from equations originally derived for movement of single drops in the large extent of quiescent continuous phase, for example, those of Hu and Kintner (1955), modified by empirical constants. Weiss et al. (1995) proved that such equations do not apply to intensively agitated stirred columns, especially not at higher holdups. They proposed methods for evaluating the actual drop velocities from local measurements of holdup and drop-size distribution directly in the columns.

The mass-transfer rate is usually expressed either by a mass-transfer coefficient, sometimes even assumed to be independent of drop diameter and time, or computed from model equations, most frequently those of Handlos and Baron (1957). However, the transfer rate depends both on drop diameter and the drop age, and the model of Handlos and Baron has been developed for single drops moving in a large extent of quiescent liquid with constant concentration of the solute. It is not reliable even for these conditions, and its application for situations with a concentration gradient in the continuous phase is outright wrong. For an extraction column, the variation of mass-transfer rates with drop diameter and time may be evaluated from the concentration profiles by optimization but this requires a reliable model for the hydrodynamics. In this work we therefore concentrated on the hydrodynamic behavior and left the mass-transfer questions for a future study. We have developed a model which is applicable both to evaluate the holdup and the drop-size distribution profiles from known breakage and coalescence parameters and to evaluate these parameters from the profiles by optimization. We could simulate the performance of a pilot-plant column in its complete range of operational parameters, including the flooding conditions, with the average mean deviation from a set of measured profiles being under 20%.

Description of Model

To simulate the variations of the holdup and the drop-size distribution with the height of a countercurrent extraction column, the drop population balances should be written and solved for all column stages. The in- and outflows of drops of different sizes, as well as their formation and destruction by breakage and coalescence, must be considered. Though these balances may be written quite generally, simplifications are feasible to speed up the computation and to allow for limited knowledge about the variation of drop velocity and the interaction rates with drop size, holdup, and physical properties of the liquid system. A possible way of simplifying the procedure is the application of the geometric model introduced by Laso et al. (1987) and Steiner (1988). The drops are allocated into a small number of geometric classes, the characteristic volume of the next class being twice the volume of the previous class. The only interactions considered are the splitting of a drop into two identical halves and a coalescence of two identical drops into one. After a breakage, the products belong to the next lower class; after coalescence to the next higher class, the smallest drops do not break, and the greatest ones do not coalesce.

For the description of a countercurrent column, the geometric model is combined with the model of Jiřičný et al. (1979). The column is divided into a number of stages, and the balance equations are solved for each drop class and each stage. The following treatment assumes that the stages are real and limited by stators with identical free areas ϕ, and that the column works with light phase dispersed. The stages are numbered from the bottom to the top of the column.

Velocity profile in continuous phase

The actual mean velocity (ms^{-1}) inside the typical stage i is defined as

$$\bar{u}_i = -\frac{b_c}{1-\epsilon_i} \tag{1}$$

The negative sign corresponds to the coordinate system with positive direction from the bottom to the top of the column. The local velocity fluctuations considerably affect the column behavior. In the absence of better possibilities it is assumed with Jiřičný that the velocity profile in the continuous phase may be represented by two characteristic values, each of them being valid for one-half of the horizontal cross-section of the column. It is assumed that both these velocities may be derived from the mean velocity adding or subtracting a fixed term as adopted earlier in the classical stage-wise backflow model

$$u_{i,k} = \bar{u}_i (1 \pm f) \tag{2}$$

Index k refers to the two halves of the column cross-section, and f is the dimensionless backflow coefficient in the continuous phase. It is not identical with the back-flow coefficient from the backmixing theory, and its value must be obtained by optimization. In the stator openings the available area is smaller and, thus, the velocity is higher. The velocity between the stages i and $i+1$ separated by a stator plate with a relative free area ϕ is given by

$$u^s_{i+1,k} = -b_c \frac{1 \pm f}{\phi (1 - \epsilon_{i+1})} \tag{3}$$

The index $i+1$ denotes the stage where the flow originates.

Velocity of drops

The relative drop velocity (the velocity of a drop relative to the surrounding continuous phase) is a function of drop size, system properties, purity of the interface, holdup and intensity of the energy input. It is an essential variable that must be known accurately before any simulation is attempted. Weiss et al. (1995) have shown that it is not identical with the velocity of a single drop ascending in a large volume of quiescent continuous phase modified by some holdup correction, but that it strongly depends on the column construction and the intensity of the agitation. They proposed two methods for its evaluation, either from tracer experiments or by recalculation from local values of holdup and drop-size distribution in a column. The second method has been applied here, the final equation being

$$v^r_{i,j} = 15.50 \exp(-1.172 r)(1 - \epsilon_i)^{1.53} d_j^{1.07 \exp(-0.214 r)} \tag{4}$$

The coefficients of this equation are close to the coefficients obtained by Weiss et al. for a similar system. The drop velocity in the stator openings relative to fixed coordinates is obtained by vector addition of Eqs. 3 and 4

$$v^s_{i,j,k} = v^r_{i,j} + u^s_{i,k} \tag{5}$$

The result may be both negative and positive; in the former case the drops are entrained with the continuous phase and enter the stages from the ''wrong'' side.

Drop movement between the stages

Assuming that the drop can enter and leave the stages in both directions, there are four possible streams associated with a typical stage i, which may but need not carry drops

i— Entering from stage i − 1 (normal flow direction)

$$\dot{n}^{\text{up}}_{i-1,j,k} = n_{i-1,j} v^s_{i-1,j,k} \ldots v^s_{i-1,j,k} > 0 \tag{6a}$$

ii— Entering from stage i + 1 (backflow)

$$\dot{n}^{\text{down}}_{i+1,j,k} = n_{i+1,j} v^s_{i+1,j,k} \ldots v^s_{i+1,j,k} < 0 \tag{6b}$$

iii— Leaving to stage i + 1 (normal flow direction)

$$\dot{n}^{\text{up}}_{i,j,k} = n_{i,j} v^s_{i,j,k} \ldots v^s_{i,j,k} > 0 \tag{6c}$$

iv— Leaving to stage i − 1 (backflow)

$$\dot{n}^{\text{down}}_{i,j,k} = n_{i,j} v^s_{i,j,k} \ldots v^s_{i,j,k} < 0 \tag{6d}$$

If the conditions at the righthand side are not fulfilled, the corresponding streams do not carry drops.

Drop formation and destruction

According to the geometric model restrictions, the drops of j-class are formed by breakage of drops from class $j+1$ and by coalescence of drops from class $j-1$. They are destroyed by breakage into drops of $j-1$ class and by coalescence into drops of $j+1$ class. The breakage is assumed to be a first-order process, with its rate being proportional to the drop concentration. The coalescence is of second order, with its rate being proportional to the product of concentrations of both coalescing drops. With the geometric model, only identical drops are allowed to coalesce so the rate is proportional to the square of the concentration of the original drops. Though the proportionality parameters are called breakage and coalescence constants, respectively, they are functions of system parameters, drop size, holdup, and intensity of the energy input. The total number of drops of class j produced or destroyed in a unit time in stage i (in a column with unit cross-sectional area) is

$$q_{i,j} = h_s \left[K^c_{i,j-1} n^2_{i,j-1} + 2 K^B_{i,j+1} n_{i,j+1} - K^B_{i,j} n_{i,j} - 2 K^C_{i,j} n^2_{i,j} \right] \tag{7}$$

In a preliminary study with a single-stage cell dimensioned identically with the column stage (Balmelli, 1998) the breakage and the coalescence parameters were evaluated from experimental measurements of steady-state variations of the drop-size distribution with the speed of agitation and the holdup, and from unsteady-state variations of the specific interfacial area with time after an abrupt change of the speed of agitation. Assuming that the above parameters may be approximated by power functions and obtaining the coefficient and the exponents on drop volume, holdup, and speed of agitation by curve fitting, the following equations were obtained

$$K_{i,j}^{B} = 1.063(10^{-2})\epsilon_i^{0.464} r^{5.52} \upsilon_j^{0.2} \tag{8}$$

$$K_{i,j}^{C} = 0.499\epsilon_i^{0.88} r^{-20.5} \upsilon_j^{0.514-0.234r} \tag{9}$$

Balance equations for a typical stage

Combining the equations for drop movement and interaction the final differential equations for the typical stages are

$$h_s \frac{dn_{i,j}}{dt} = 0.5 \sum_{k=1}^{2} \left[\dot{n}^{\text{up}}_{i-1,j,k} + \dot{n}^{\text{down}}_{i+1,j,k} - \dot{n}^{\text{up}}_{i,j,k} - \dot{n}^{\text{down}}_{i,j,k} \right] + q_{i,j} \tag{10}$$

End stages

Extraction columns are usually limited on both sides by settlers with diameters considerably larger than that of the active section of the column. Assuming that the light phase is dispersed, the lower settler reduces the entrainment into the continuous phase while in the upper one the drops emerging from the top stage are coalesced at the main interface. The simulation of the upper settler is easy as it may be assumed that all drops leaving the active section are removed from the column so there are no downstreams into the last working stage. This stage, however, works under a different hydrodynamic regime, as there is no agitator above it, so the flow patterns around the upper stator plate are not symmetrical. Though not applied in this work, a different value of the backflow coefficient f may be necessary.

The conditions under the first stage at the bottom of the column may be more complicated. In intensively agitated columns with low phase throughputs the dispersed phase is usually introduced without a distributor so it may be assumed that all entering drops belong to the largest class. They are rapidly broken down in the first stage so it is not important how large they actually were. The holdup in the lower settler is small and the drops do not leave the column with the continuous phase. This corresponds to the assumption of zero downstreams from stage 1. However, with large throughput of the continuous phase and in the neighborhood of the flooding point this is not true and a more detailed simulation of the lower settler is necessary. Without an agitator, there is no breakage but the small entrained drops may proceed with the stream of the continuous phase out of the column, coalesce and return into the column, or accumulate under the lowest stator plate to form a layer of a denser coalescing dispersion (which is the most common cause for the flooding). For a comprehensive simulation, the velocity of the continuous phase and the holdup under the first stage must be estimated and the lower settler treated as a special stage with a special set of equations.

In our column there was a piece of regular packing under the bottom stage, which broke the large incoming drops into smaller ones. The drops leaving the lowest stage were assumed to coalesce and be incorporated into the feed drops inside of the packing and recycled into the column. A small experimental study was carried out to derive an empirical equation for the mean diameter of the incoming drops, which was used as the initial condition in the simulations

$$\bar{d}_{\text{in}} = 1.932 + 7.695(10^{-2})b_c + 3.756/b_c - b_d\big[6.679(10^{-2}) + \exp(-0.571 b_c)\big] \tag{11}$$

The result (mean diameter of the incoming drops) is in millimeters.

Computation

A C++ computer program has been written to solve the set of differential Eqs. 10 for all column stages and drop classes, the structure of which is given in Figure 1. After defining the constants and variables, the geometrically distributed drop classes were defined and the initial conditions specified. As a rule, the simulation started with the column filled completely with the continuous phase; at the beginning of the time counting the dispersed phase was introduced at the bottom. The program solved the set of some 400 differential equations (19 stages times 22 to 24 drop classes) using the modified Runge-Kutta-Fehlberg procedure. The main termination criterion was the stationarity of the local holdups in

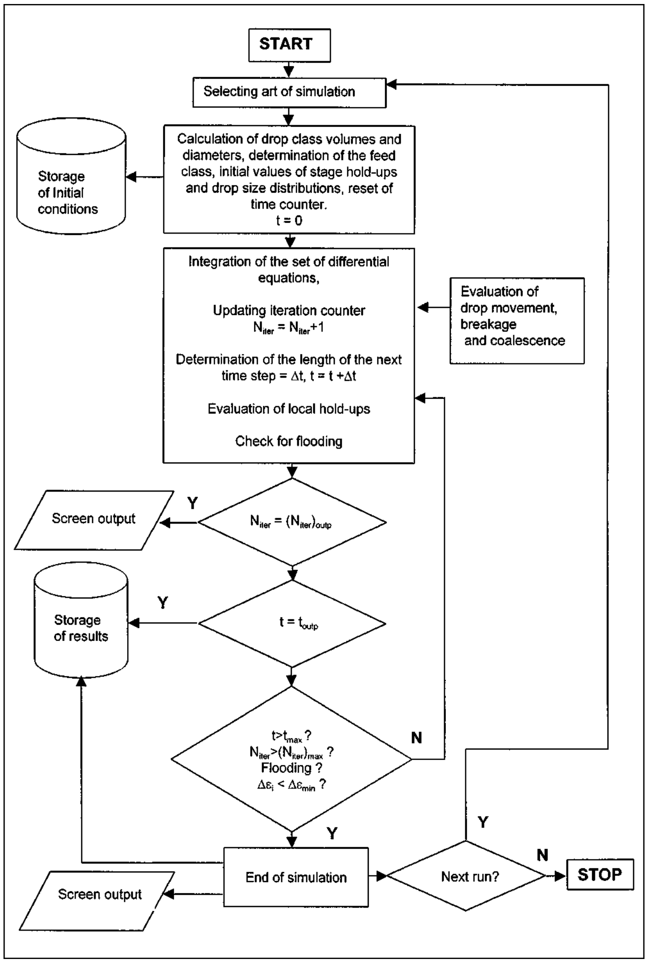

Figure 1. Simulation program.

all stages of the column, which should not change by more than the prescribed maximum difference. In test runs it was found that this value must be kept rather small; with $\Delta\epsilon_{\max} = 10^{-6}$, the accuracy of the calculated mean drop diameter was about 0.2% different from the one calculated with an extremely high accuracy. From $\Delta\epsilon_{\max} = 10^{-9}$ down, no difference was observable. The main simulations reported here were therefore terminated when none of the stage holdups changed by more than 10^{-9} between two successive evaluation steps. To prevent breakdown of the simulation at flooding, an additional criterion stopped the program when some of the stage holdups exceeded 0.75. The maximum allowable number of iterations was limited to prevent infinite runs in oscillatory regimes without obtaining perfect steady state. However, this was a very exceptional situation, which did not occur since the program was properly debugged. In the final version the program was provided with a graphical interface to enable its application in typical situations without first understanding the code.

The primary result of the calculation was the distribution of the drop concentrations in all stages of the column, defined as the number of drops of a given size per unit volume of the column (n_{ij}). A summation over all drop classes provided the local holdup of the dispersed phase.

Experimental Data

A comprehensive set of experimental data measured earlier by Kumar (1985) was used as a basis for the simulation. It contained results from 93 runs performed with a Kühni column with a diameter of 150 mm and 18 agitated stages, the main dimensions of which are collected in Table 1. The column was constructed of glass, stainless steel and Teflon so no contaminating materials were in contact with the liquid system. It was supported by a tank system with 4 storage vessels of 500 L each, and the flows were automatically stabilized. The experimental runs involved steady-state measurements of local holdups in 6 positions along the column height and measurements of drop-size distribution in 3 positions. The holdup was measured by rapid withdrawal of 100 mL of the agitated dispersion through special taps, the drop-size distributions were obtained by sucking the drops into capillary tubes with diameters of about 2 mm and photographing the flowing dispersion outside the column. The evaluations were carried out manually on a digitizing tablet connected to a desktop computer. The position of the sampling points is shown in Figure 2. The column was operated with o-xylene dispersed in water. The system was kept reasonably clean by using deionized water, frequently discharged; the organic phase was distilled about once in a week. The experiments used in this work have been run without mass transfer, the total specific loading varied between 1.4 and 11.7 $m^3/m^2 \cdot h$, phase ratio (b_d/b_c) between 0.4 and 2.5, and holdup between 0.02 and flooding (> 0.6).

Table 1. Column Specifications

Column diameter	0.15 m	
Active column height	1.26 m	
Number of compartments	18	
Compartment height	0.07 m	
Stator plate (fractional free area)	0.235	
Shrouded six bladed turbines	diameter	0.085 m
	height	0.010 m
End sections	length	0.600 m (upper) 0.550 m (lower)
	diameter	0.200 m
Packing in bottom section	Melapack (Sulzer of Switzerland)	

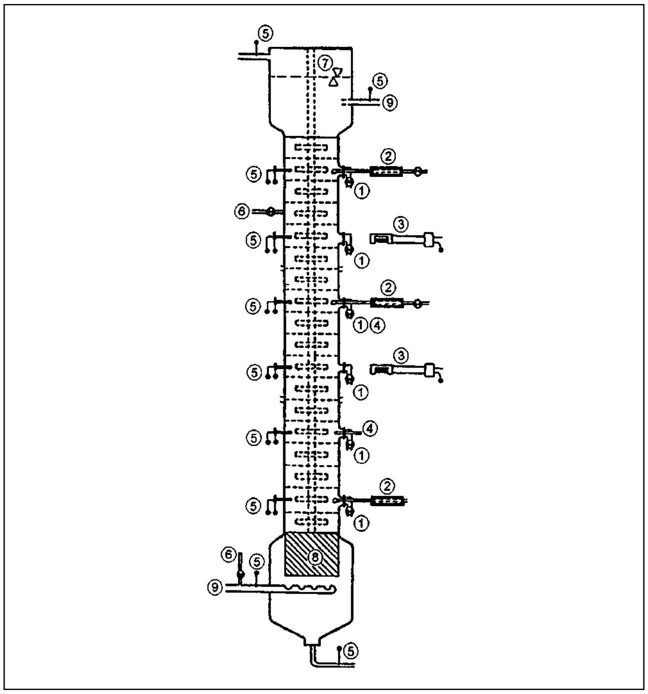

Figure 2. Experimental column with arrangement of measuring devices.

(1) Holdup; (2) drop-size distribution; (3) backmixing in dispersed phase; (4) backmixing in continuous phase; (5) concentration; (6) tracer injection; (7) interface control; (8) special packing; (9) feed line.

Results

Free parameters

The only unknown parameter in the model equations is the backflow coefficient f. However, it was found by test simulations that the difference between the predicted and measured mean drop diameters may be up to 50% when the breakage and coalescence constants are evaluated from Eqs. 8 and 9. Though these equations were derived from measurements with a stirred cell dimensioned as the column stage and with the same system as used in the column there were still some differences:

- The top and the bottom plates of the cell were smooth and there were no connecting rods between them. In the column there were stators with sharp edged perforations and three connecting rods.
- Working without phase throughput, it was impossible to have uniform dispersion along the cell height with lower intensities of the agitation. While the column operated regularly already at 100 rpm, the cell needed at least 180 rpm to prevent vertical segregation of the drops.

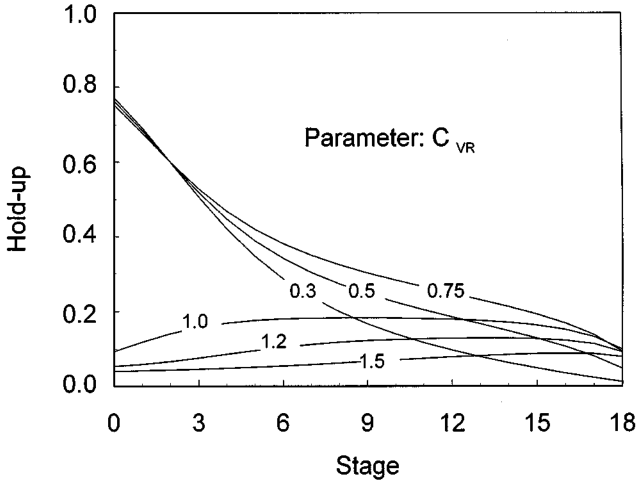

Figure 3. Variation of local holdup with the value of C_{VR}.

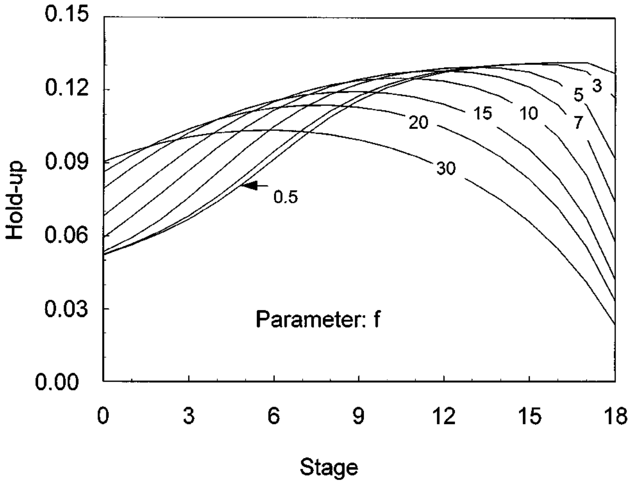

Figure 5. Variation of local holdup with the value of backflow coefficient *f*.

To allow for these differences the following free parameters were defined and evaluated by optimization from the column data:

- backflow coefficient f;
- multiplicative factor of the breakage constant C_{KB};
- multiplicative factor of coalescence constant C_{KC}; and
- multiplicative factor of the relative drop velocity C_{VR}.

C_{KB} and C_{KC} were found to be mutually dependent so it was possible to keep the value of C_{KC} at 1 and reduce the number of the free parameters to three. The influences of the remaining parameters on a typical holdup profile are shown in Figures 3 to 5.

The optimized mean value of C_{VR} was 1.24, practically independent of the speed of agitation. This parameter has been determined with a high accuracy, the standard deviation being 0.01. Its deviation from unity may be explained by inaccuracies in the column construction and by the uncertainty about the velocity profile in the continuous phase. The increase of holdup on the lefthand side of Figure 3 with $C_{VR} < 1$ indicate that at these values the column would flood and no stable operation would be possible.

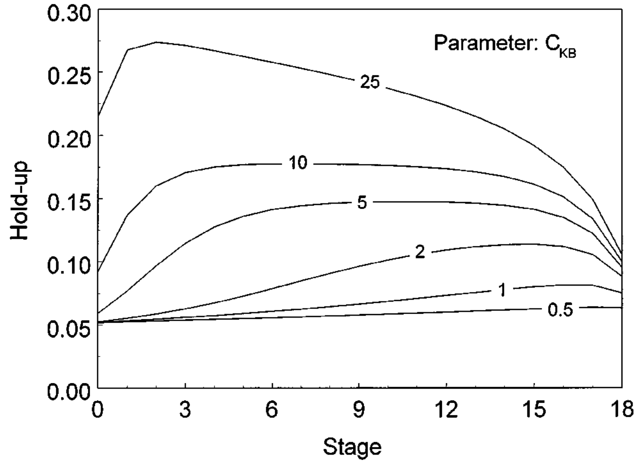

Figure 4. Variation of local holdup with the value of C_{KB}.

The value of the backflow coefficient varied from 3.67 for the low stirring speed of 140 rpm to 4.9 at 300 rpm, but no decrease of accuracy was observed when a mean value of 4.58 was used for all experiments. The standard deviation was 0.12.

Figure 6 indicates that the correction factor for the breakage constant C_{KB} was a function of the stirring intensity, approaching unity at high revolutions. It could be correlated by the following function

$$C_{KB} = 383.5 r^{-3.89} \tag{12}$$

This could be incorporated into Eq. 8 so the breakage constant for the column is given by

$$K^{B}_{i,j} = 4.07 \epsilon_i^{0.464} r^{1.63} v_j^{0.2} \tag{13}$$

The three correction factors were evaluated by optimization

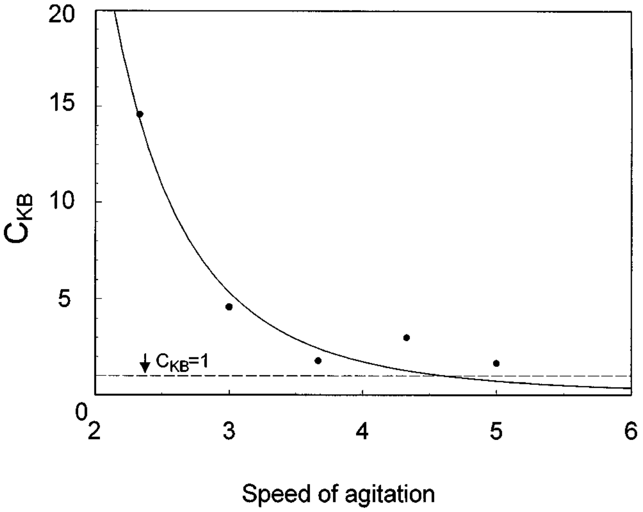

Figure 6. Variation of factor C_{KB} with speed of agitation.

Table 2. Results of Simulation

rpm	b_c ($m^3/m^2\cdot h$)	b_d ($m^3/m^2\cdot h$)	Mean Deviation of Local Holdups* (%)	Mean Deviation of Mean Diameter d_{10}** (%)	Mean Deviation of Sauter Diameter d_{32}** (%)
140	2.38	2.60	27.1	34.0	12.0
	2.38	5.32	19.0	6.0	14.9
	3.40	2.60	17.6	14.5	13.8
	5.77	3.96	21.5	17.2	16.2
	8.38	3.28	26.1	21.6	23.0
180	1.92	2.21	13.9	16.5	18.5
	1.92	3.34	6.9	14.2	8.1
	1.92	4.47	17.0	21.1	12.0
	2.53	2.21	8.3	15.7	11.7
	2.53	2.83	7.5	9.0	5.0
	3.40	2.21	17.3	11.0	19.0
220	1.92	1.47	13.7	9.6	5.0
	1.92	1.92	7.0	12.3	12.7
	1.92	2.60	22.6	27.2	24.0
	1.92	3.40	15.4	Flooding	
	2.38	1.00	5.0	4.9	4.7
	2.38	1.92	20.1	21.8	18.2
	3.40	1.00	16.1	15.3	12.1
260	1.58	0.60	6.3	16.0	21.0
	1.58	1.00	5.8	12.3	9.6
	1.58	2.72	19.0	Flooding	
	1.92	0.60	8.2	21.2	23.1
	1.92	1.00	23.0	15.3	19.8
	2.38	0.60	16.9	21.2	45.0
	3.40	1.87	26.1	Flooding	
300	0.79	0.60	13.5	20.4	30.8
	0.79	1.87	22.4	27.3	29.0
	1.81	1.00	18.3	21.6	23.8
mean			**15.8**	**17.1**	**17.3**

*Stages 2, 5, 8, 11, 14 and 17.
**Stages 2, 19, 17.

of a selected set of 23 typical experiments carried out under characteristic operational regimes: The experiments close to the flooding conditions and those with extremely low-phase throughputs were excluded as not sufficiently accurate. The series with the lowest stirring speed of 100 rpm was left out as the stirred cell could not be operated under these conditions due to insufficient dispersion and segregation of the liquid phases. All optimized parameters were well defined, the t-values (ratios of parameter values to standard deviations) being frequently above 20. With the optimized parameters, another set of 28 randomly selected experiments was simulated with the accuracy shown in Table 2.

Table 3. Summary of Simulation Example

Operational Regime	b_c	2.38 ($m^3/m^2\cdot h$)	
	b_d	1.00 ($m^3/m^2\cdot h$)	
	r	220 (rpm)	
		Optimized	Generalized equations
Free parameters	C_{VR}	1.20	1.24
	C_{KB}	1.80	2.45
	f	5.0	4.58
Mean deviations (%)	Holdup	3.9	5.0
	d_{10}	5.1	4.9
	d_{32}	4.7	8.6

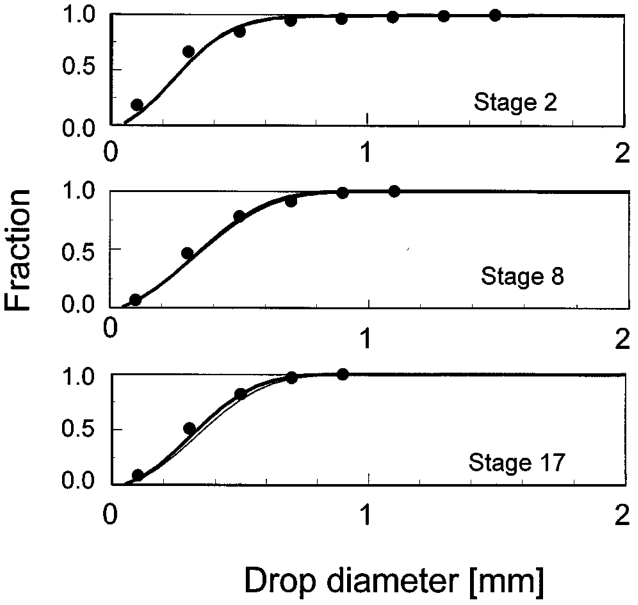

Figure 7. Cumulative drop fractions in stages where experimental data is available.

Simulations both with generalized equations and optimized constants are shown (——; ——, respectively).

Example of simulation

Complete simulation results of a randomly selected experiment are shown in Table 3 and Figures 7 to 9. The free parameters were both optimized for this special experiment and calculated from the generalized Eqs. 4, 9 and 13. For this special evaluation, 22 drop classes covering drop diameters from 0.05 to 6.4 mm were used, leaving a safe margin of at least two empty classes at the top and the bottom of the distribution. After reaching the steady state, the overall material balances of both phases were perfectly fulfilled. Each stage input equaled output with accuracies better than 10^{-10}

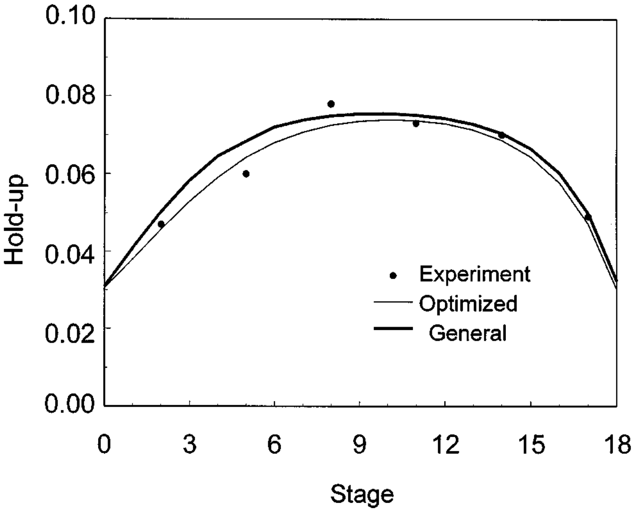

Figure 8. Variation of holdup with column height.

Comparison of simulated and experimental profiles.

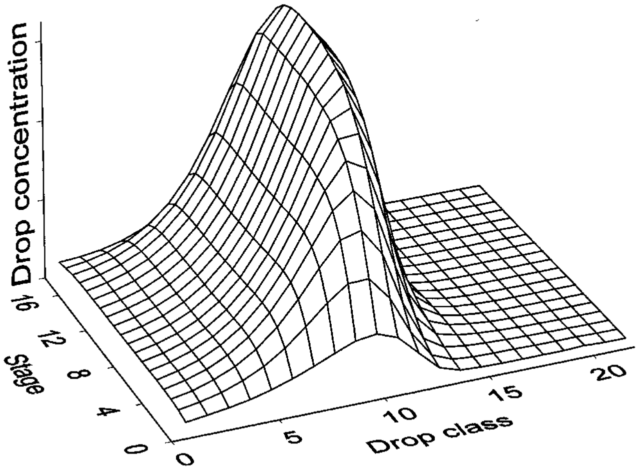

Figure 9. Variation of drop concentration with column height.

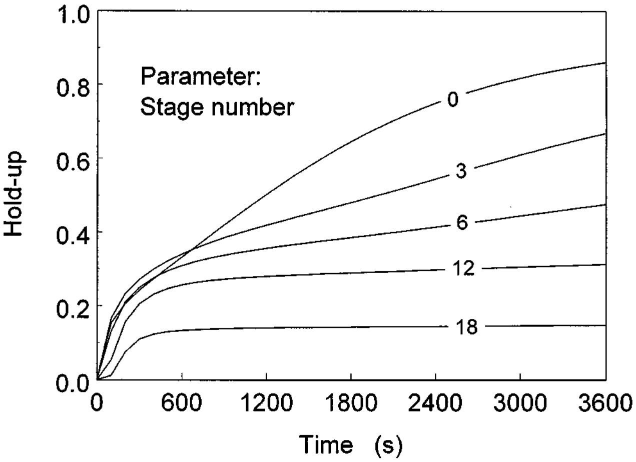

Figure 10. Variation of local holdup with time at flooding conditions.

$b_c = 1.92$ m^3/m$^2 \cdot$h; $b_d = 6.06$ m^3/m$^2 \cdot$h, 180 rpm.

m^3/m$^2 \cdot$h, and the maximum holdup change at termination was 5.10^{-10}. The program found that there would be a downflow of small drops from the lowest stage to the settler, corresponding to an entrainment of the dispersed phase of 1.9% of the throughput. The holdup profile was simulated with a mean deviation of 3.9% using the free parameters optimized for this special experiment and 5.0% with the parameters from the generalized equations. Also, the drop-size distributions are well reproduced, with the main deviations occurring at small drop diameters. Figure 9 shows that the drop concentrations are changing tremendously along the column height. Though the changes in the holdup and the mean drop diameter are not very large, the drop-size distribution becomes much narrower with the column height as the large drops are broken down and the small ones are washed out into the lower part of the column.

Flooding

As a rule, the column floods at its lower end when large numbers of small drops are entrained by the continuous phase into the lower settler where the cross-sectional area is large and the velocity, therefore, is low. The drops accumulate under the first plate, coalesce, and a layer of dense dispersion or fully coalesced phase then grows into the column making any steady-state operation impossible. But, for very special combinations of phase throughputs and stirring speeds, it is difficult to start the flooding along the column height without changing the physical properties of the liquids by mass transfer or constructional errors (for example, smaller free area of stator plates, where the bearings are positioned). Being by definition an unsteady-state process, the flooding is difficult to monitor experimentally, and the operator must decide when to declare the column flooded and record the holdup at flooding conditions. The column starts to fill with the dispersed phase from the bottom and the layer of the dense dispersion grows continuously.

The process may be well simulated by the recent program and a dangerous situation could be recognized earlier than in an actual column from the shape of the holdup curves. Using the additional breakup criterion of holdup increase above 75%, the flooding could be automatically predicted. The simulation progress could be very slow with regimes close to the flooding conditions; the variation of local holdups with time shown in Figure 10 is typical for a straightforward situation. In this case the flooding progressed regularly and the final holdup profile agreed well with the experimental one (Figure 11). Though some holdup profiles at flooding conditions documented in the data set displayed irregular shapes, which could not be exactly reproduced, it was always possible to say that the corresponding regime was not within the normal range of operation.

Examples of further simulations

Figure 12 demonstrates the increase of the steady-state holdup when the throughput of the dispersed phase is in-

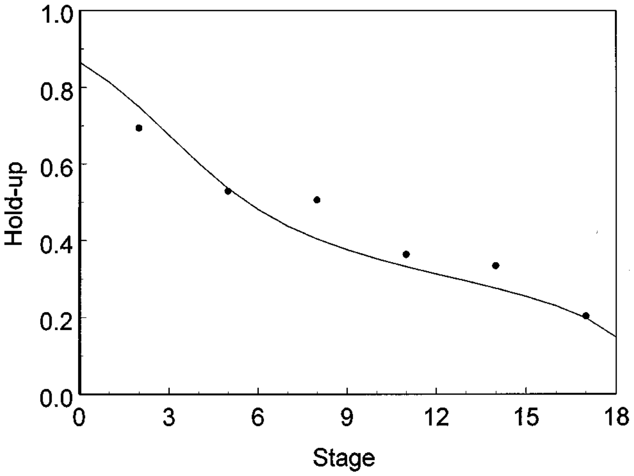

Figure 11. Comparison of simulated holdup profile with experimental data at flooding.

Same parameters as in Figure 10.

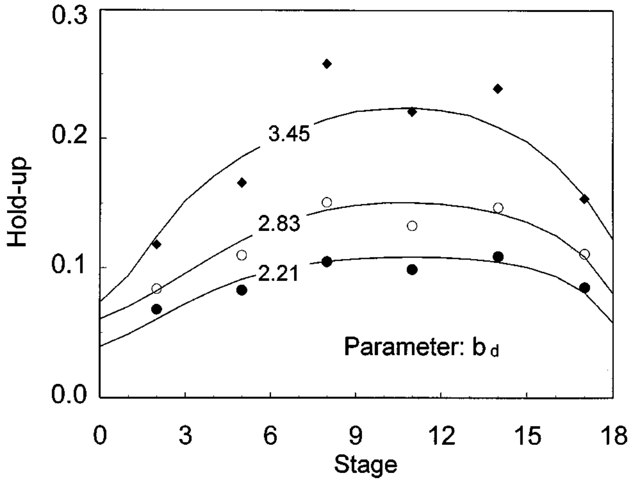

Figure 12. Variation of local holdup with loading of dispersed phase.

$b_c = 3.4\ \mathrm{m^3/m^2 \cdot h}$, 180 rpm.

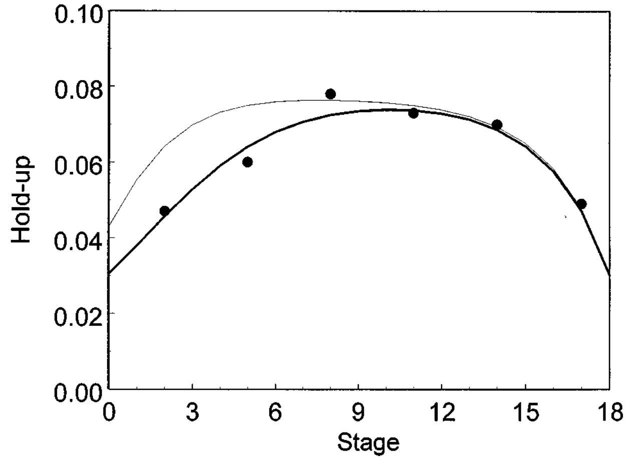

Figure 14. Simulated increase of stage height from 0.07 m (——) to 0.14 m (——).

creased while the throughput of the continuous phase and the stirring speed is kept constant. The upper curve is close to the flooding conditions, and a further slight increase of the throughput would cause a rapid increase of the holdup in the lower part of the column and the breakdown of the steady-state operation.

The approach to the steady state in Figure 13 shows how the column fills with the dispersed phase during a typical startup. The column was filled with the continuous phase, and at time zero the dispersed phase was introduced. Due to a relative high intensity of the agitation (220 rpm), numbers of small drops were produced which caused the maximum at the middle of the column. They could not proceed against the stream of the continuous phase to the top of the column and distributed along its lower part until equilibrium was found. Again, a small increase of either the speed of agitation or the phase loadings would cause a flooding. The simulation indicates that the transient period is rather long, though the change in holdup between 500 and 1,500 s is not very large. An inexperienced operator would probably not wait half an hour before carrying out steady-state measurements of holdup and drop-size distributions.

The simulation can also predict the column behavior after changing some construction parameters. Doubling the stage height would cause a change of the holdup profile to a shape shown in Figure 14. In this example the number of stages was kept constant, so the column height was doubled. The path of the drops in the column was now longer and, as the speed of agitation was rather high, the breakage process especially intensified. More small drops were formed and entrained from the upper to the lower part of the column, so the maximum on the holdup curve became broader. Such an influence could not be predicted intuitively as, with other operational conditions, completely different results might result.

Figure 15 shows the influence of the free area of the stators limiting the stirred stages. Decreasing the free area increases the holdup, as it is now more difficult for the drops to

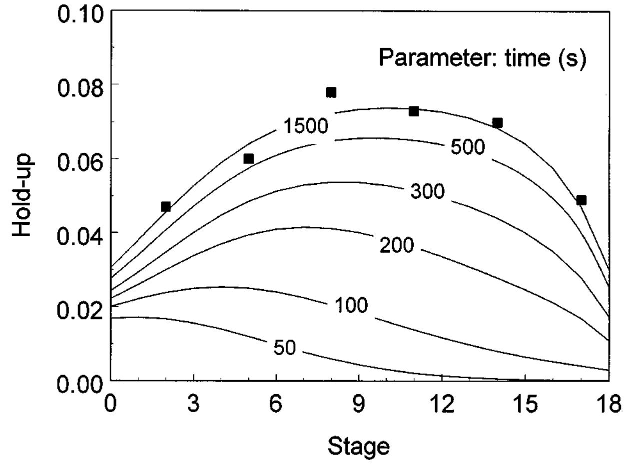

Figure 13. Variation of local holdup during a startup from empty column.

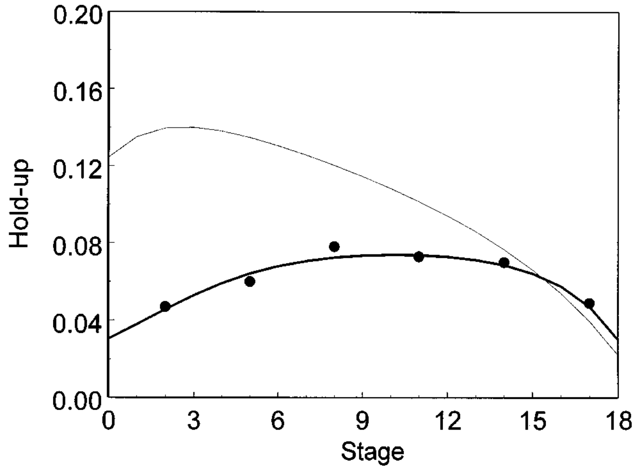

Figure 15. Simulated decrease of free area of the stator plates from 0.235 (——) to 0.15 (——).

overcome the counterstreams of the continuous phase in the stator openings. A decrease of the free area by 35% doubled the maximum holdup and brought the holdup profile close to the dangerous shape indicating that the regime is now close to the flooding. In Kühni columns stages of various free area are frequently used to make the holdup profiles more uniform. Without a simulation program, the plate arrangement must be optimized in experiments.

Discussion

The purpose of this work was to develop a workable simulation program for a given type of extraction column. It should be sufficiently precise to predict the extraction column's behavior in full range of the operational parameters, but at the same time, so simple that it would run fast on a medium desktop computer. Both simulation of the column performance from known parameters and evaluation of the parameters from known column performance should be possible. We have therefore combined the simplified geometric model with empirical equations developed and valid for the given system to obtain a very reasonable result.

The geometric model drastically simplifies the actual situation, but Laso (1986) has proved by numerous case studies that the deviation from the complete balance is less than 15% if the same equations for breakage and coalescence constants are used. With slightly modified constants, the results may be made identical with the full-scale model. So if the constants are evaluated from experimentally measured drop-size distributions using the same simplified model as used later in the simulation, the results are sufficiently accurate. Given the model simplicity, such an evaluation is easily possible.

A realistic description of the variation of the breakage and coalescence rates with drop diameter, speed of agitation, and holdup is essential for every serious simulation. Even the most sophisticated model degenerates to a precise sum of fancy numbers if these parameters are not defined properly. We have, therefore, evaluated the breakage and coalescence rates from measurements with the same liquid system in a single-stage stirred cell dimensioned as a stage of the column. This was only partially successful—without the phase throughputs, the dispersion in the cell was not homogeneous at low stirring speed. Transferring the resulting equations to the column, we had to introduce a correction factor that approached unity at high speeds of agitation but became large with insufficient stirring (Figure 6). This indicates that, at low levels of turbulence, the breakage in the column is more intensive than in the cell. We are trying to improve the cell construction, but it is hardly possible to simulate perfectly a flow system in cell without phase throughputs at low intensity of agitation. This problem may be bypassed if the breakage and coalescence parameters are obtained from measurement on a small pilot column and used in the scale-up.

The drop velocity related to fixed coordinates may be accurately evaluated by the methods described by Weiss et al. (1995). However, its recalculation to the relative velocity is difficult, as the velocity profile of the continuous phase is still unknown. A detailed study of local velocity and its fluctuations in the stator openings would be of considerable use, as the backflow coefficient in the continuous phase remains the least known parameter. It cannot be evaluated by the tracer methods used with the backmixing models but must be optimized from the column data.

The equations for the coalescence and breakage rates used in this work (Eqs. 9 and 13) are special equations valid for the given system and equipment only. However, the same procedure may be used with any equations so the simulation is generally applicable. It is sufficiently accurate for most applications in design and scale-up. It cannot replace the experimental work, as at least three parameters must be determined from measured data but it may save a considerable number of experiments and generalize data obtained on smaller columns and single-stage cells for applications on columns of different size and construction details.

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Notation

b = specific throughput, $m^3/m^2 \cdot s$
d = drop diameter, m
h_s = stage height, m
K^B = breakage constant, s^{-1}
K^C = coalescence constant, $m^{-3} \cdot s^{-1}$
n = drop concentration, m^{-3}
$\dot{n}$ = drop flow rate, $m^{-2} \cdot s^{-1}$
q = drop formation rate, $m^{-2} \cdot s^{-1}$
r = speed of agitation, s^{-1}
u^s = velocity of continuous phase in stator openings, ms^{-1}
v = velocity of dispersed phase, ms^{-1}
v^r = relative drop velocity, ms^{-1}
v^s = drop velocity in stator openings, ms^{-1}
ϵ = holdup of dispersed phase

Indices

c = continuous phase
d = dispersed phase
j = drop class
in = initial value

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