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Simulation of packed extraction columns with the REDROP model

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1 Introduction

For the design of extraction columns earlier methods utilized layer models like e.g. the plug–flow model and axial back–mixing models. All these models assume that both phases can be regarded as continuous. This requires that all elements of each phase behave statistically similar and that the dispersed phase represents a monodispersion where all drops within each height element of the column have identical solute concentration.

Recent models, so-called drop-population models, take into account that the dispersed phase shows drop-size distribution due to splitting and coalescence of drops. As a disadvantage, these models are mathematically difficult to handle. Only with various simplifications it is possible to use the drop-population models with acceptable effort. E.g. the drop-size distributions are separated into classes and restrictions concerning the splitting and coalescence of the drops are applied. Up to now the influence of mass transfer on the coalescence, which was verified experimentally, has not been considered in the simulations.

The aim of this paper is to propose a different approach to the simulation of packed extraction columns, the representative-drops model (REDROP model). In the broadest sense this approach can be understood as a Monte-Carlo simulation of the solution of drop-population balances. The basic idea is to model the development of each individual drop within a representative fraction of the cross section of a packed column. Each drop is traced during its entire lifespan, in the extreme case from the inlet to the outlet of the column. Changes of position, concentration, and size of the drop influenced by fluid dynamics, mass transfer, and splitting, as well as coalescence processes are monitored.

2 Theory

During its lifespan a drop is subject mainly to influences of fluid dynamics, mass transfer, as well as coalescence and redispersion. In the next three subsections (2.1 – 2.3), these influences are dealt with seperately and in detail, before their interactions are explained in section 2.4.

2.1 Fluid Dynamics

Fluid dynamics in an extraction column is determined by movements of both, dispersed and continuous phase. Starting point of the calculations is a single drop, for which several authors proposed calculation methods of sedimentation rate and rising velocity, depending on drop size.

Here, computations of relative drop velocity (v_e) in an empty tube were made, using the formulas of Ishii, and Zuber [1] for rigid and circulating drops, as well as Hu, and Kintner [2] for oscillating drops. Hence, the transition from circulating to oscillating drop, is determined by the intersection of both correlations.

The relative drop velocity v_e has to be adjusted to account for the influences of packing bodies. Mersmann [3] presented a suitable correlation:

$$v_0 = \frac{v_e}{1 + C_1 \left[\frac{v_e^2 \cdot \rho_d}{(d_F - d_E^*) \cdot \Delta\rho \cdot g} \right]^{C_2}}, \tag{1}$$

where d_F is the diameter of a packing body, and d_E^* that of the largest stable drop with $d_E^* < d_F$. The constant C_2 equals 0.667 and C_1 equals 1.4.

Swarm behaviour is accounted for by the correlation

$$v'_{rel} = v_0 \cdot (1 - \varepsilon)\,, \tag{2}$$

proposed by Gayler, Roberts, and Pratt [4]. In this correlation v_0 is the velocity determined by equation 1 and ε is the hold–up in the column.

Finally, the influence of coalescence on fluid dynamics has to be considered. Here the model proposed by Míšek [5] is used:

$$v_{rel} = v'_{rel} \cdot e^{a \cdot \varepsilon} \tag{3}$$

Míšek proposed the factor a to be $a = 0$ for inhibited coalescence and $a > 0$ for heavily coalescing dispersions.

However, in order to calculate the vertical position of the drop in a column its absolute velocity v_d is significant. The absolute velocity can directly be calculated from the velocity of the continuous phase,

$$v_c = \frac{\dot{V}_c}{1 - \varepsilon}\,, \tag{4}$$

and the relative velocity v_{rel} with

$$v_d = v_{rel} - v_c. \tag{5}$$

2.2 Mass Transfer

Mass transfer per unit area across a drops phase boundary, $\dot{m}''_A$, is proportional to the overall mass–transfer coefficient K_d and the concentration difference $y^* - y = mx - y$:

$$\dot{m}''_A = K_d \cdot (y^* - y)\,. \tag{6}$$

The overall mass–transfer coefficient is composed of mass–transfer coefficients β_d and β_c on the inside and outside of a drop, respectively. Thus, the overall mass transfer coefficient is calculated from:

$$\frac{1}{K_d} = \frac{1}{\beta_d \rho_d} + \frac{m}{\beta_c \rho_c}, \tag{7}$$

where m is the partition coefficient of the component transferred.

In the literature many correlations for calculating mass–transfer coefficients are proposed. The selection of the proper correlation depends on the flow behaviour of the drops. In table 1 the correlations selected by Arimont [6] which are also used in the simulations are listed.

	inside of the drop	outside of the drop
rigid surface	$Sh_d = 0.332 \cdot \frac{1}{Fo} + 6.58$ [7]	$Sh_c = 2 + 0.76 \cdot Re_c^{\frac{1}{2}} \cdot Sc_c^{\frac{1}{3}}$ [8]
circulating drops	$Sh_d = 0.284 \cdot \frac{1}{Fo} + 17.9$ [9]	$Sh_c = 3.62 \cdot Re_c^{\frac{1}{2}} \cdot Sc_c^{\frac{1}{3}} - 178$ [12]
oscillating drops	$Sh_d = 0.00365 \cdot Pe' + \frac{0.3}{Fo}$ [10, 11]	

Table 1: Correlations for the mass–transfer coefficients used in this work. For the definition of the dimensionless numbers Sh, Re, Sc, Fo, and Pe' see the list of symbols.

The transition from rigid to circulating drops for the mixture of n–butyl acetate/water was appointed to a drop diameter of 1.5 mm. The transition point from circulating to oscillating drops is the same as used in sedimentation calculations.

2.3 Splitting and Coalescence

Polydispersity of a drop–size distribution is the result of interaction of drop–drop coalescence and drop splitting. The REDROP-algorithm (see section 2.4) calculates the interactions in time intervals Δt. Hence a coalescence probability p_K and a splitting probability p_S in a time interval Δt have to be determined for every drop in the column.

The splitting probability of a drop within a time interval is the ratio of time interval Δt and mean life time τ_S of the drop until splitting. The mean life time is set inversely proportional to the square of drop diameter. The proportionality factor or splitting factor K_S accounts for all influencing variables (exept drop diameter) like pulsation, operating state, and geometric size:

$$p_S = \frac{\Delta t}{\tau_S} \qquad \text{with} \qquad \tau_S = \frac{K_S}{d^2}. \tag{8}$$

Similar to τ_S a mean coalescence time τ_K is calculated. The coalescence probability of each drop is formulated analogous to the splitting probability. Additionally, influence of mass transfer rate on coalescence as found by Rohlfing [13] is accounted for. Proportionality factors are K_K and K_{Km}:

$$p_K = \frac{\Delta t}{\tau_K} + \frac{\varepsilon \cdot f(\dot{m}_A'') \cdot \Delta t}{K_{Km}} \tag{9}$$

$$\text{with} \qquad \tau_K = \frac{K_K \cdot d}{\varepsilon} \qquad \text{and} \qquad f(\dot{m}_A'') = \tanh(1.0 \cdot 10^4 \, \frac{\mathrm{s\,m^2}}{\mathrm{kg}} \cdot \dot{m}_A'').$$

The parameters K_S, K_K and K_{Km} were adapted to experimental data. $f(\dot{m}_A'')$ is an empirical formulation valid for the mass-transfer direction dispersed to continuous ($d \rightarrow c$).

The formulations for splitting and coalescence probabilities (eq. 8–9) are ad hoc assumptions and have to be verified experimentally. They show, however, that the developed mode of computation, which uses ordinary formulations for independent variables, is suitable to simulate different operating states of an extraction process in pulsed packed columns. Calculations using formulations for p_S and p_K from literature [14, 15] did not lead to usable results in our simulations; the drop-size distribution was to narrow.

After calculating splitting and coalescence probabilities for each drop, the decision has to be made if the drop splits, coalesces or encounters neither event during Δt. The decision is made by utilizing random numbers as shown schematically in figure 1. Splitting and coalescence probabilities for an exemplary drop are represented by lines of corresponding length. The value of a generated random number $0 \leq s < 1$ is then compared with the probabilities as shown. As a result s lies in one of the three indicated sections and determines the event encountered by the drop during Δt.

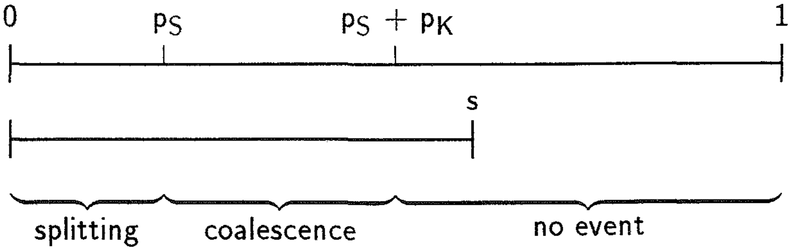

Figure 1: Calculation of drop splitting and drop coalescence

The sum of the probabilities $p_S + p_K$ has to be less than unity, since otherwise not all possible events are accounted for properly. As these probabilities describe quantities relative to the choosen time interval, the above mentioned condition fixes the maximum lenght of the time interval in the calculation.

2.4 Calculation Method

The interaction of fluid dynamics, mass transfer, splitting, and coalescence determine the drop–size distribution, as well as the hold–up in the column. Moreover, the hold–up in turn influences the interaction between the different influences. Thus a dynamic equilibrium is established at steady state. Figure 2 illustrates the REDROP algorithm with which the transient and steady–state behaviour of a column is calculated.

While the dispersed phase is calculated drop by drop, the concentration of the continuous phase and the local hold–up of the dispersed phase are determined for small height elements which are moved with the continuous phase through the column. The number of height elements and the operating conditions as well as the geometry of the column are set in the initialization procedure. At the start the entire column is filled with continuous phase.

In the time loop of the REDROP algorithm the necessary calculations are done for each time intervall Δt. Drop feeding is done in accordance with the given phase load, a drop size at the distributor and the choosen time interval.

Subsequently, in the so-called drop loop, the velocity (section 2.1), the mass transfer (section 2.2), as well as splitting and coalescence probabilities (section 2.3) are calculated for every single drop. If the splitting and coalescence calculations show that the drop should split or coalesce, an according flag for the drop is set. However, coalescence ist not yet possible, since the partner drop for coalescence can not yet be determined.

After a complete run of the drop loop, the drops are sorted according to their height in the column and splitting as well as coalescence of the drops is performed if the according flag is set. A drop marked for splitting separates into two drops. Concentration and height of the generated drops is adapted from the original drop. The daughter drops' splitting flag and their life span are reset. Coalescence is carried out for two drops flagged for coalescence, the height of which is closest to each other. After the coalescence, the new drops life span is set to zero. If mass transfer occurs from dispersed in the direction to the continuous phase, secondary drops are generated by following scheme: If one of the drops after splitting or coalescence is larger than 1.5 mm in diameter, a secondary drop is separated. The diameter of the secondary drop is chosen randomly between 0.1 mm and 1 mm.

Finally, concentrations of both phases and the hold–up are calculated for the height elements before the time is incremented by Δt and computation continues with the next drop feeding.

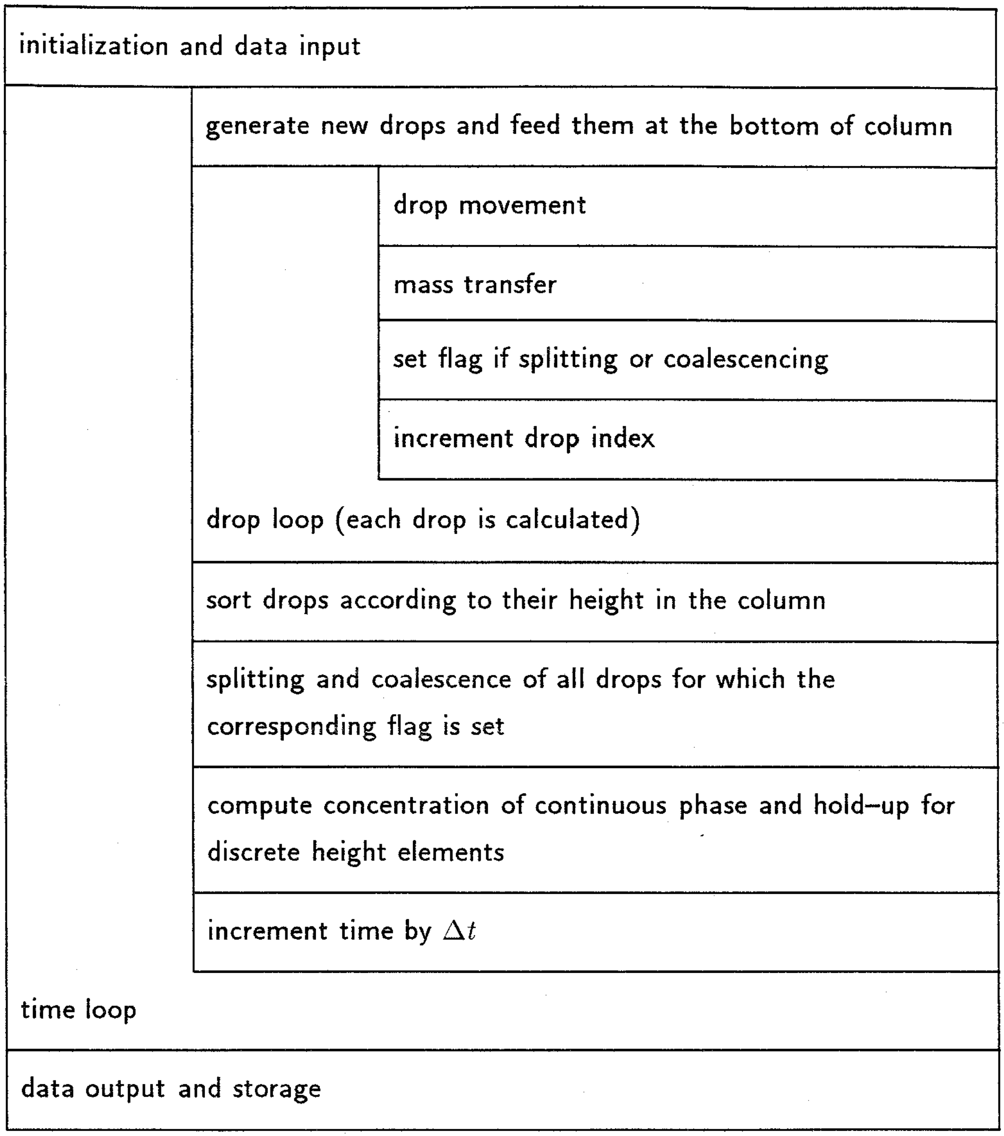

Figure 2: Algorithm of the REDROP model

3 Results

Calculations were done evaluating the experimental data of Rohlfing [13]. The measurments of Rohlfing were carried out in a pulsed packed column, equipped with various measuring methods to determine drop–size distributions of dispersed phase and concentration profiles of both phases. Moreover, dynamic and static hold–up were determined. A detailed description of these measuring devices and of the experimental procedure can be found in [13].

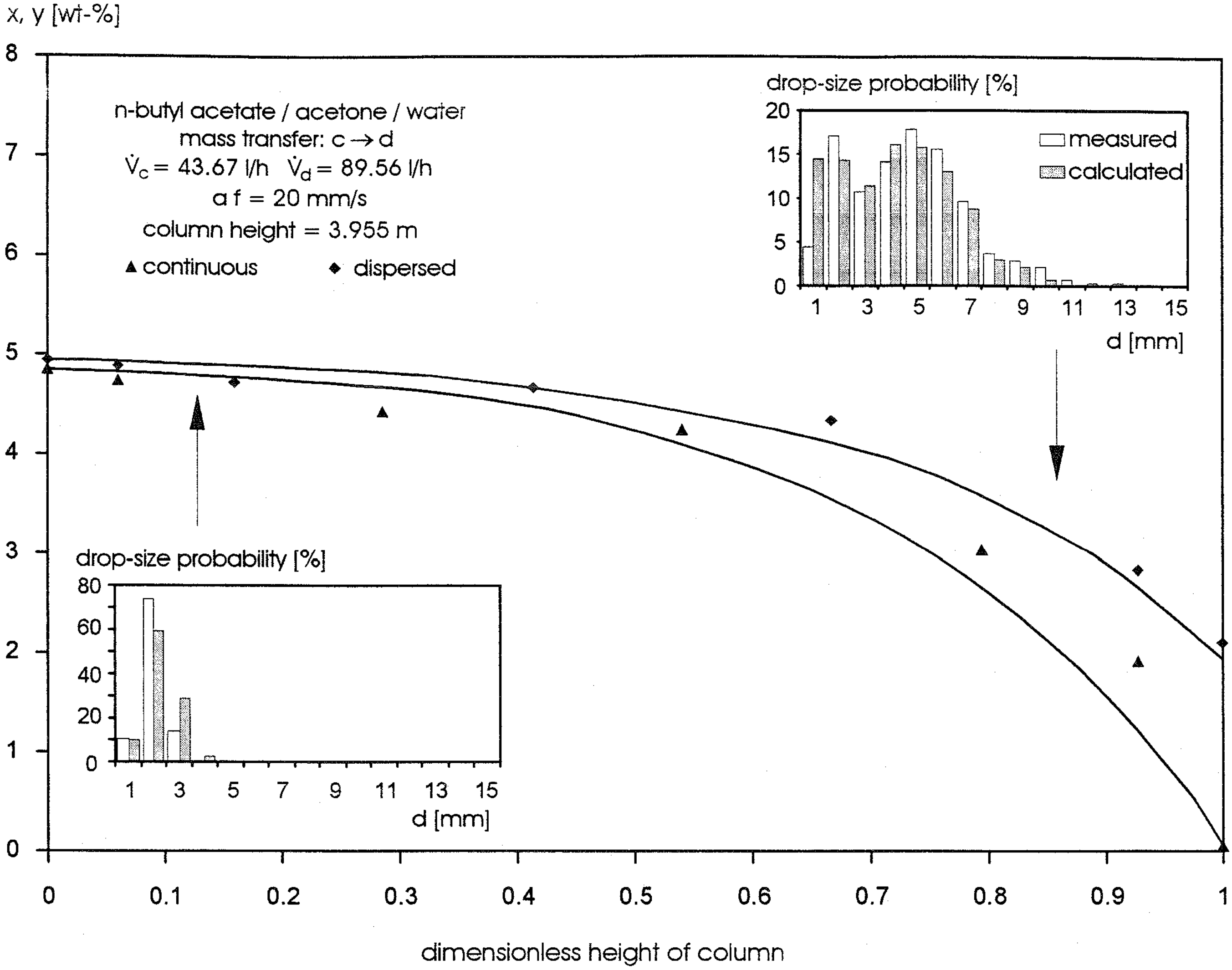

Figure 3: Measured (symbols) and calculated (lines) concentration profiles, as well as drop–size distributions (arrows indicate the height in the column where the distributions were measured); parameters for calculation: $K_S = 0.7 \cdot 10^{-5} \text{m}^2\text{s}$; $K_K = 1.6 \cdot 10^2$ s/m; $K_{Km} = 0.5 \cdot 10^{-5}$ s; $a = 2$

All experiments were conducted using a mixture of n–butyl acetate/water/acetone. Also in all experiments, n–butyl acetate was used as dispersed phase, and acetone as transfering component. Material properties of this mixture are documented in [16].

In figures 3 and 4 calculated concentration profiles and drop–size distributions are compared with experimental results. The calculated concentration profiles agree well with the measurements.

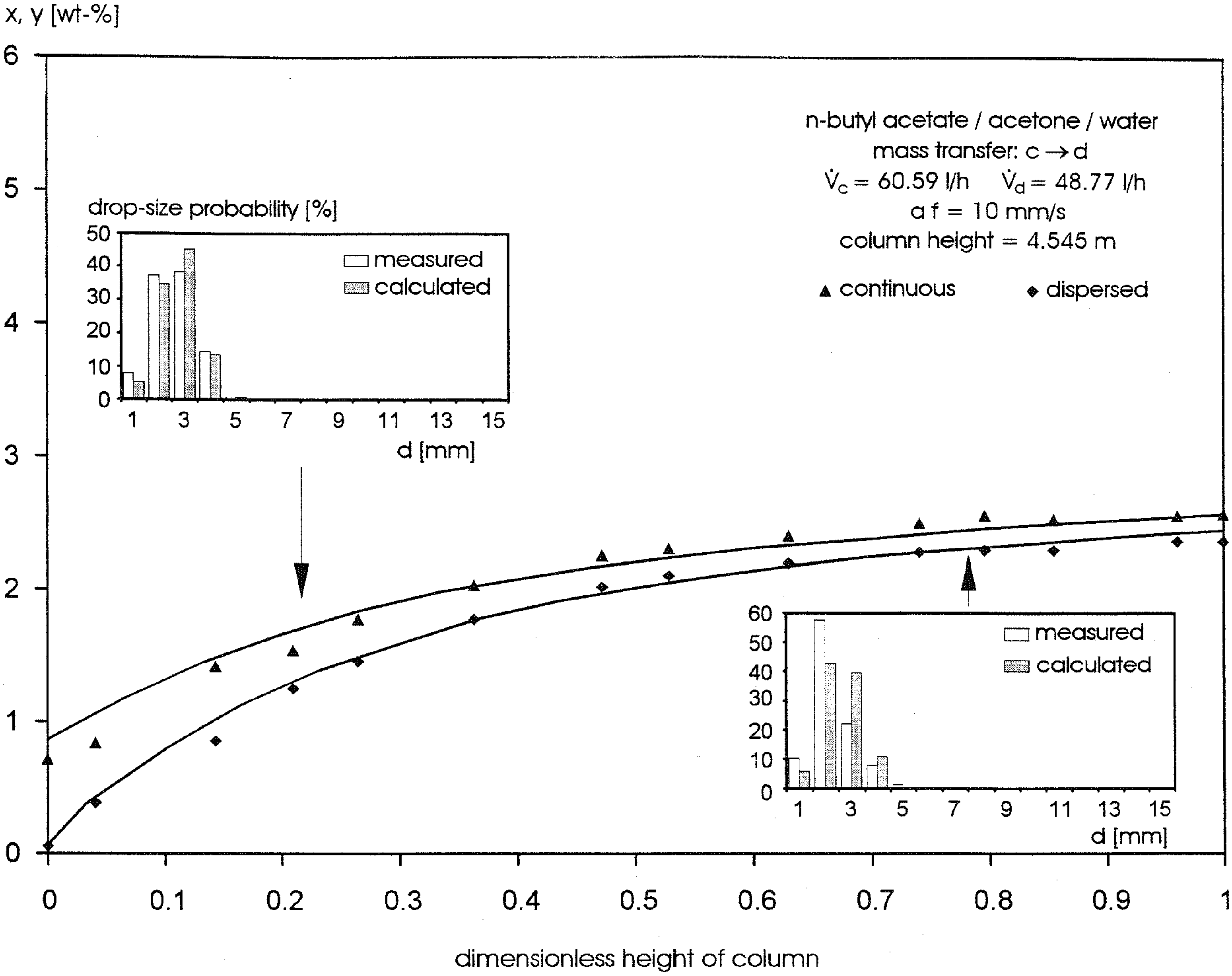

Figure 4: Measured (symbols) and calculated (lines) concentration profiles, as well as drop–size distributions (arrows indicate the height in the column where the distributions were measured); parameters for calculation: $K_S = 4 \cdot 10^{-5}$ m^{2}s; $K_K = 1.4 \cdot 10^2$ s/m; $K_{Km} = 0.5 \cdot 10^{-5}$ s; $a = 0$

Figure 3 shows results for an example where the coalescence behaviour is influenced by mass transfer for mass–transfer direction dispersed to continuous. At high mass–transfer rates coalescence increases. As a result, a wide drop–size distribution in the upper part of the column builds up in spite of pulsation. The drop–size distribution with its two maxima is typical for this mass–transfer direction. In the lower part of the column, where the mass–transfer rate is extremly low, a drop size distribution with a small mean diameter establishes. However, for mass–transfer direction continuous to dispersed (figure 4) an influence of mass transfer on the drop–size distribution in the column can hardly be detected.

4 Conclusions and Significance for Practice

The REDROP model for simulation of the operating characteristics of a packed extraction column was developed and a corresponding computer program implemented. Within the program the main influences on the extraction process are taken into account: mass transfer, fluid dynamics as well as splitting and coalescence. The operating characteristics of an extraction column in pilot–plant scale were simulated by the program in agreement with experimental data.

In summary tracing individual drops has the following advantages:

- The mathematical effort is comparatively small.
- Dynamic calculations can be carried out without additional effort.
- Any mathematical model for the description of fluid dynamics, mass transfer and splitting and coalescence processes can easily be tested and varied within the program in the appropriate modules. Interactions between the different processes acting on each drop can easily be taken into account (e.g. mass transfer influencing coalescence).
- Concentration distributions for each drop–size class evolve quite naturally.

Especially the two latter points represent a distinct progress of the REDROP model over previous approaches.

5 List of Symbols

A	$[\mathrm{m}^2]$	surface area
a	—	coalescence parameter
$a \cdot f$	[mm/s]	pulsation parameter: stroke · frequency
C_i	—	constants
d	[m]	drop diameter
d_F	[m]	diameter of packing bodies
d_E^*	[m]	diameter of largest stable drop
D	$[\mathrm{m}^2/\mathrm{s}]$	diffusion coefficient
$Fo = \frac{4\tau D}{d^2}$		Fourier number
g	$[\mathrm{m}/\mathrm{s}^2]$	gravitational acceleration
K	$[\mathrm{kg}/\mathrm{m}^2\ \mathrm{s}]$	overall mass–transfer coefficient
K_K	[s/m]	coefficient for drop coalescence
K_{Km}	[s]	coefficient for drop coalescence due to mass transfer

K_S	$[\mathrm{s\ m^2}]$	coefficient for drop splitting
m	—	partition coefficient of transferred component ($y^* = mx$)
$\dot{m}''_A$	$[\mathrm{kg/sm^2}]$	mass flow per unit area
p_K	—	coalescence probability
p_S	—	splitting probability
$Pe' = \dfrac{v_{rel}\,d}{D_d\,(1+\eta_d/\eta_c)}$		modified Peclet number
$Re = \dfrac{v\,\rho\,d}{\eta}$		Reynolds number
s	—	random number [0,1[
$Sc = \dfrac{\eta}{\rho\,D}$		Schmidt number
$Sh = \dfrac{\beta\,d}{D}$		Sherwood number
v_0	[m/s]	characteristic velocity
v_e	[m/s]	relative drop velocity in an empty tube
v_{rel}	[m/s]	relative drop velocity in the column
$\dot{V}$	[l/h]	volume flow rate
x	[wt–%]	mass fraction of transferred component in continuous phase
y	[wt–%]	mass fraction of transferred component in dispersed phase
y^*	[wt–%]	equilibrium mass fraction of transferred component in dispersed phase
β	[m/s]	mass–transfer coefficient
Δt	[s]	time interval
$\Delta\rho$	$[\mathrm{kg/m^3}]$	density difference
ε	—	hold–up of dispersed phase
η	[kg/(m s)]	dynamic viscosity
ρ	$[\mathrm{kg/m^3}]$	density
τ	[s]	actual life time of the drop
τ_K	[s]	drop–coalescence time
τ_S	[s]	drop–splitting time

Subscripts

c	continuous phase
d	dispersed phase

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