EFFICIENT COMPUTER SIMULATION OF THE DYNAMICS OF AN AGITATED LIQUID-LIQUID EXTRACTION COLUMN

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ABSTRACT

Given the difficulties associated with experimentation upon liquid-liquid extraction processes, computer simulation of these processes has recently acquired great relevance and usefulness as complement, if not an alternative, to direct experimentation. This is becoming increasingly attractive, as both the power and the availability of personal computing increase.

Recently, Ribeiro (1995), Ribeiro *et al.* (1997)) developed a fast and precise algorithm, adapting population balance models as Coulaloglou and Tavlarides' (1977) for the dispersed phase breakage and coalescence, and both the rigid and the oscillating drop models as proposed by Cruz-Pinto *et al.* (1983), for mass transfer. The algorithm enabled the simulation of the evolution in time of the trivariate joint distribution of drop properties in two-phase continuous mixers, in steady and transient states. A simplified version of this algorithm, that calculates the drop size distribution and the mean and standard deviation of solute concentration within each volume class, was further developed by Regueiras *et al* (1996).

In this work, the applicability of this simplified algorithm to the dynamic simulation of a Kühni liquid-liquid extraction column is illustrated. A transport model was developed from previous models of Cruz-Pinto (1979). Coulaloglou and Tavlarides' (1977) drop interaction model was again adopted.

It is shown that the algorithm is able to describe both the transient and the steady-state behaviour of a Kühni pilot column (Technical University of Munich), and the simulation runs that were made show good agreement with the experimental results. The present version of the algorithm predicts local hold-ups and drop size distributions at different column stages. So far, the results do not include mass transfer, but future work will contemplate this feature.

INTRODUCTION

In the last few decades, mechanically agitated devices for liquid-liquid contact in extraction equipment have acquired great importance, due to their capability of creating and maintaining large interfacial areas, which promote mass transfer. The efficient behaviour of these columns has been shown to be due, not only to these large interfacial areas, but also to the mixing effects caused by drop interactions - breakage and coalescence.

In recent years, many authors have spent great efforts in modelling these columns and attempting experimental validation of their models. Models such as those of Cruz Pinto (1979), Casamatta and Vogelpohl (1985), Laso *et al.* (1987) Tsouris *et al.* (1994) and, more recently, Gerstlauer *et al.* (1996), have been proved able to describe with reasonable accuracy the results of their authors' experiments, even if most of them adopted significant simplifications, in order to make calculations feasible in reasonable time. The most important simplification commonly adopted is to ignore drop coalescence, because the modelling of this effect is responsible for the most significant part of the computational effort required for the solutions, even when modelling concerns only the simplest case of an agitated vessel (Ribeiro *et al.*, 1995).

A simplified version of Ribeiro's algorithm developed by Regueiras *et al.* (1996) made possible the description of the transient behaviour of an agitated vessel, by means of a detailed

population balance including drop breakage and coalescence (such as the Coulaloglou-Tavlarides' model) and mass transfer effects, using the oscillating drop model, with only modest computer resources and very short processing times (computation time well below real process time).
In this work, we present a development of the previous algorithm to describe the case of an agitated column (Kühni type), whose results show excellent agreement with those obtained in experiments with a pilot plant Kühni column (University of Munich).

THE MODEL

The Kühni column may be adequately described as a sequence of agitated vessels, with backmixing and forward mixing effects on the movement of the dispersed phase along the column. The adopted model combines a population balance model as used by Coulaloglou and Tavlarides and their co-workers with the numerical solution technique developed by Ribeiro (1995) and further streamlined by Regueiras *et al.* (1996). This model describes the transient behaviour of a continuous ideally agitated vessel. Details of the model and of the algorithm are given elsewhere (Ribeiro *et al.*, 1995 and Regueiras *et al.* 1996).
The equations for this model are shown in Table 1. They include the hydrodynamic description of the dispersion within the vessel and the mass transfer of a solute between the continuous and the dispersed phases. In order to extend this model to the simulation of an agitated column, it was necessary to combine this model with a model for the motion of the drops and the continuous phase between stages.

Table 1 – Drop interaction model equations

BREAKAGE EQUATIONS:

With the power number $N_P = 1.08 + \frac{10.94}{\mathrm{Re}_R^{0.5}} + \frac{257.37}{\mathrm{Re}_R^{1.5}}$,

after Fischer (1973), *in* Kumar and Hartland (1995), the power input to each stage is

$$P = N_P n^3 . D_R^5 . \rho_C$$

and the power density is

$$\varepsilon = \frac{4P}{\pi D_C^2 . H . [\rho_C . (1-\varphi) + \rho_D . \varphi]}$$

where D_C = inner diameter of the column and H = stage height.
The breakage frequency is

$$g(d) = C_1 . \frac{\varepsilon^{1/3}}{(1+\varphi) . d^{2/3}} . \exp\left[-C_2 . \frac{\sigma . (1+\varphi)^2}{\rho_D . d^{5/3} . \varepsilon^{2/3}} \right]$$, after Guimarães (1989).

The daughter drop size distribution is normal, with mean volume equal to half the mean volume of the parent drop.

COALESCENCE EQUATIONS:

$$c(d,d') = h(d,d') . \lambda(d,d')$$, where

$$h(d,d') = C_3 . \frac{\varepsilon^{1/3}}{1+\varphi} . \left(d^2 + d'^2\right)\left(d^{2/3} + d'^{2/3}\right)^{1/2}$$ = the collision frequency and

$$\lambda(d,d') = \exp\left[-C_4 . \frac{\rho_C . \mu_C . \varepsilon}{\sigma^2 . (1+\varphi)^3} . \left(\frac{d . d'}{d + d'} \right)^4 \right]$$ = the coalescence efficiency of the collision,

after Guimarães (1989).

This transport model was based on the one described by Cruz-Pinto (1979) and directly calculates the slip velocities, taking into account the density effects and the drag relationships for the drops, as previously used by Barnea and Mizrahi (1973 and 1975).
The dispersion effect is taken into account and is modelled as a random deviation superimposed on the terminal velocities of the drops, to take into account the agitation intensity effect, as a function of the rotor peripheral speed.
The constriction factor, C_R, that controls the drop passage between stages, was calculated by a linear function of the fractional free area of the separators, as suggested by Goldmann (1986).
The equations used in the transport model are grouped in Table 2.

Table 2 – Transport model equations

TRANSPORT EQUATIONS:

The slip velocity is obtained, after Cruz-Pinto (1979), from

$$v_s = \begin{cases} \dfrac{g.\Delta\rho.d^2}{18.\mu_C} & for \quad \mathrm{Re} \le 1 \\ 0.249.d.\left(\dfrac{g^2.\Delta\rho^2}{\rho_C.\mu_C}\right)^{1/3} & for \quad 1 \le \mathrm{Re} \le 60 \\ \mu_C.P^{0.15}.\dfrac{J-0.75}{\rho_C.d} & for \quad 60 \le \mathrm{Re} \end{cases}$$

with $\Delta\rho = (1-\varphi).(\rho_C - \rho_D)$, and $P = \dfrac{\rho_C^2.\sigma^3}{g.{\mu_C}^4.\Delta\rho}$

and $J = \begin{cases} (23.26\,H)^{0.422} & for \quad H > 72.708 \\ (0.75\,H)^{0.7843} & for \quad H \le 72.708 \end{cases}$

where $H = C_D.We.P^{0.15} = \dfrac{4.g.d^2.\Delta\rho}{3.\sigma}.P^{0.15}$

EFFECTIVE FLOW AREA:

$S_e / S = 0.75.e + 0.25 = C_R$, where e is the fractional free area, after Goldman (1986)

DIFUSION EQUATION:

The mean effective vertical velocity is

$$v_e = \frac{(v_s - v_C) + \sqrt{(v_s - v_C)^2 + \frac{v_d^2}{4}}}{2}$$

with $v_d = k_3.n.D_R$ where k_3 is a constant.

RESULTS AND DISCUSSION

In this first implementation, the algorithm describes the hydrodynamic behaviour of a Kühni column and calculates the local hold-ups and the drop size distribution in each stage of the column. Because our experiments were performed in the pilot plant Kühni column of the

University of Munich, the algorithm was adapted to simulate this particular column, considering its 36 agitated stages, dimensions and other characteristics, as described elsewhere (Gomes, 1998).

In order to validate the model and its algorithm, calculations were performed using the same values of the flow rates, agitation intensity, and start-up conditions of the laboratory experiments. The same values of the drop population balance model parameters used in our previous work, concerning agitated vessels (Guimarães, 1989), which are compatible with those used in the Tavlarides team's works (Bapat 1982, Coulaloglou 1975, etc.), were also adopted here. Those values are claimed by the above authors to be nearly "universal constants", for agitated vessels.

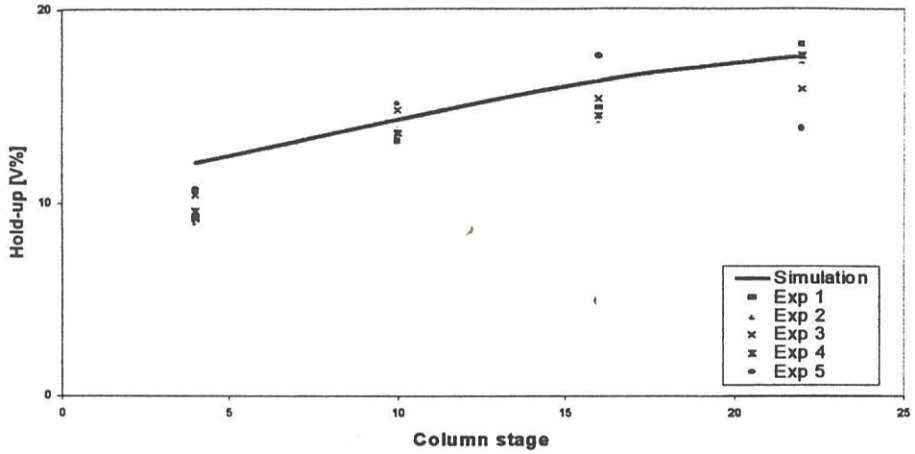

Figure 1 – Hold-up profiles at steady state - $Q_C = 125L/h$, $Q_D = 160L/h$, $n = 140r.p.m.$

The correlations used to calculate the power input were those developed by Kumar and Hartland (1996), in their valuable attempt at obtaining unified correlations for some of the more common agitated liquid-liquid extraction columns, and show good agreement with experimental results.

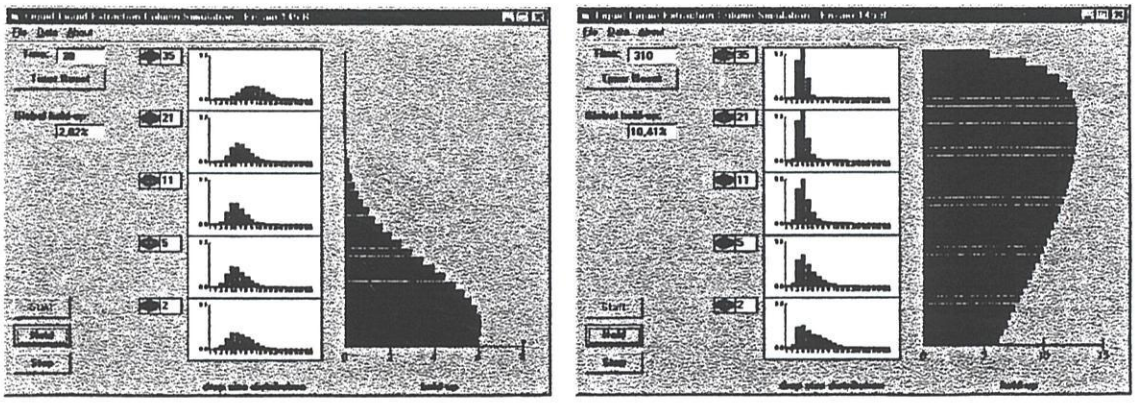

Figure 2 - Column simulation 30s after start-up and after reaching the steady state.

In the absence of experimental drop size measurements at the distributor outlet, the mean value and the standard deviation of the feed drop size distribution (which we assume to follow a log-normal law in the drop diameter), were tentatively adjusted. Further optimisation of these parameters is planned, and the Marquardt algorithm to perform these calculations has already

been implemented. In the present work, the values adopted for these parameters are those used in our simulation runs, with a view to obtaining plausible starting guesses for the Marquardt algorithm.

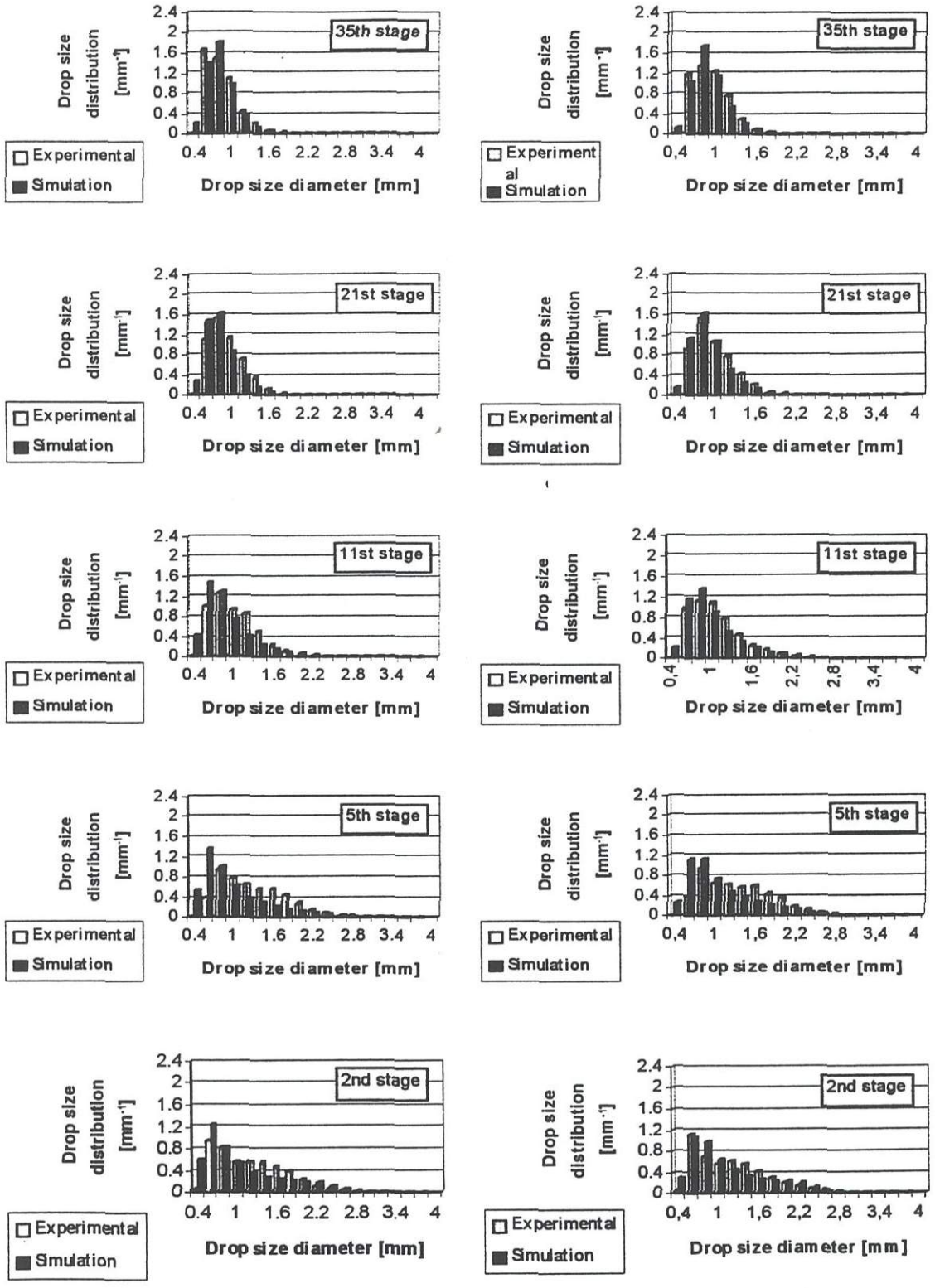

Figure 3 – Drop size distribution profiles for the equilibrated toluene-water system: $Q_C = 125 L/h$, $Q_D = 1160 l/h$; left: $n = 150 r.p.m.$, right $n = 140 r.p.m$

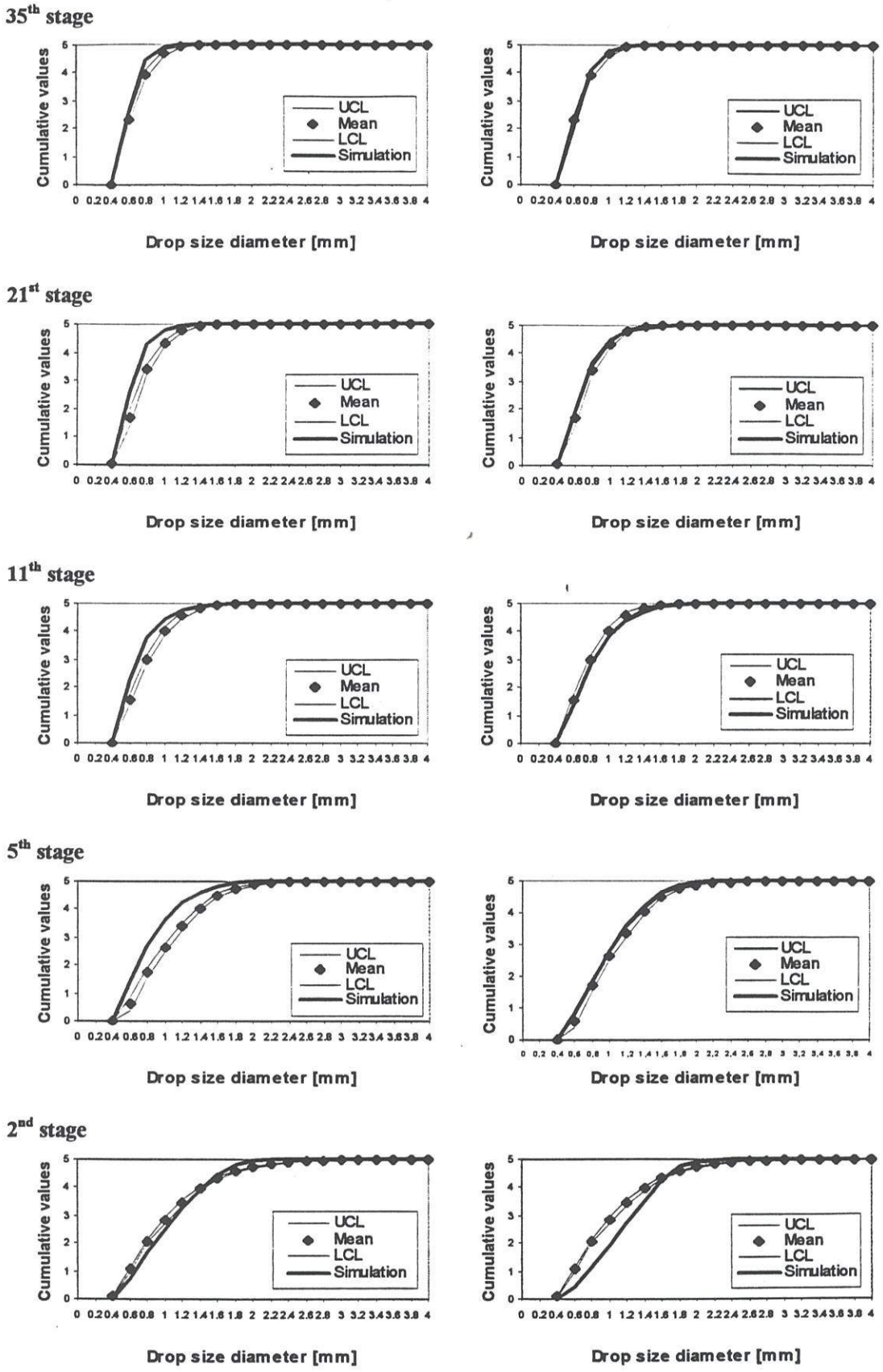

Figure 4 – Simulation results *vs.* confidence limits for the experimental cumulative drop size distributions along the column ($Q_C = 94L/h$), $Q_D = 120L/h$, $n = 170r.p.m.$); a - left: original Bapat parameters; b - right: modified Bapat parameters

Results of experimental (repeat runs) and simulated local hold-ups are shown together in Fig. 1, for comparison. They are in good agreement, and runs made for the start-up period of the column show plausible responses of the model in dynamic conditions (Fig.2), as well as for the steady state. Local drop size distributions, experimental and simulated, are shown in Fig. 3, for two different conditions of agitation intensity. The agreement obtained stands for the capacity of the model to describe, stage by stage, the drop size distributions.
The mean and the confidence interval, obtained by Student's t-technique over 6 experimental cumulative distributions, is compared with the correspondent simulation results using two different values for the population balance constants. As may be seen, the deviation between calculated and experimental distributions are not significantly outside this confidence interval. The poorest agreement was obtained for the fifth stage distributions, as illustrated in Fig 4a, where Bapat's values for the breakage and coalescence parameters were used; global hold-up values are also in poor agreement (Gomes). However, a minor modification in the breakage constants values (about 20%), used in another simulation run, leads to the results shown in Fig 4b. In this case, the poorest agreement for the drop size distributions was obtained at the lowest stage of the column, where experimental data for the distributor outlet are lacking, the first known data being precisely at the second stage; the agreement obtained for the hold-up is, in this case, satisfactory (Gomes). This fact, resulting from an insufficient instrumentation of the particular laboratory column, is the major difficulty that we shall have to overcome in the final adjustment of the model parameters.

CONCLUSIONS

1- A new, powerful, precise and fast algorithm to simulate the hydrodynamic behaviour of an agitated liquid-liquid extraction column (Kühni type) has been developed.
2- The algorithm can accurately predict the local drop size distributions and the hold-up profile of the pilot plant Kühni column of the Technical University of Munich.
3- The agreement with the experimental data obtained in the pilot plant suggests that Coulaloglou and Tavlarides' population balance model is appropriate, as well as, with minor modifications, the values for the breakage and coalescence parameters used by these authors for agitated vessels.
4- Further optimisation of the interaction constants and transport parameters is planned, as well as the inclusion of the appropriate drop mass transfer model.

ACKNOWLEDGEMENT
Co-operation is gratefully acknowledged from Professor J. Stichlmair and his staff, at the Technical University of Munich.

NOMENCLATURE

C_D	drag coefficient
d, d'	drop diameters
D_R	rotor diameter
g	gravitational acceleration
n	rotor speed (RPS)
R_e	Reynolds number
S	column cross sectional area
W_e	Weber number

v_C	continuous phase velocity
v_s	slip velocity
φ	dispersed phase hold-up
μ_C	continuous phase dynamic viscosity
ρ	effective density of dispersion
ρ_C , ρ_D	continuous and dispersed phase densities
σ	interfacial tension

REFERENCES

Bapat P. M., Ph.D. Thesis, Illinois Institute of Tecnology, Chicago Illinois, 1982.

Barnea E. and Mizrahi J., Can. J. Chem. Engng., 53, 461, 1975.

Casamatta G. and Vogelpohl A., Ger. Chem. Engng., 8, 96, 1985.

Coulalouglou C. A, Ph.D. Thesis, Illinois Institute of Tecnology, Chicago, Illinois, 1975.

Coulalouglou C. A, Tavlarides L. L., Chem. Engng. Sci.32, 1289,1977.

Cruz-Pinto J. J. C., Ph.D. Thesis, The Victoria University of Manchester, 1979.

Cruz-Pinto J. J. C., Korchinsky W. J., Al Husseini R., Proceedings of ISEC'83 Conference, 207, Denver, Colorado, 1983.

Fisher E.A, Ph.D. Thesis, Swiss Federal Institute of Tecnology, Zurich, Switzerland, 1973.

Gerstlauer A., Mitrovic A., Gilles E., Zamponi G., Stichlmair J., Value Adding Through Solvent Extraction, Vol. 1, Proceedings of ISEC`96, edited by D. C. Shalcross, R. Paimin, L. M. Prvcic, University of Melbourne, 1031, 1996.

Goldman, G., Ph.D. Thesis, Technical University of Munich, Germany, 1986.

Gomes, M L. N., Ph.D. Thesis, in preparation .

Guimarães, M. M. L., Ph.D. Thesis, Universidade do Minho, Braga, Portugal, 1989.

Kumar A., Hartland S. A., Ind. Eng: Chem. Res. 34, 3925, 1995.

Kumar A., Hartland S. A., Ind. Eng: Chem. Res.35, 8, 2682, 1996.

Laso M., Steiner L., Hartland S., Chem. Engng. Sci., 42, 2429, 1987.

Regueiras P. F. R., Ribeiro L. M., Guimarães M. M. L., Cruz-Pinto J. J. C, Value Adding Through Solvent Extraction, Vol. 1, Proceedings of ISEC'96, edited by D.C. Shallcross, R. Paimin, L. M. Prvcic, University of Melbourne, 1031, 1996.

Regueiras P. F. R., Ph.D. Thesis, in preparation.

Ribeiro L. M., Ph.D. Thesis, Universidade do Minho, Braga, Portugal, 1995.

Ribeiro L. M., Regueiras P. F. R., Guimarães M. M. L., Cruz-Pinto J. J. C., Computing Systems in Engngr. 6, 4/5, 465, 1995.

Ribeiro L. M., Regueiras P. F R., Guimarães M. M. L, Madureira C. M. N., Cruz-Pinto J. J. C., Computers and Chem. Engng. 21, 5, 543, 1997.

Tsouris C., "Modelling and Control of Extraction Columns", Ph.D., Thesis, Syracuse University (1992).

Tsouris C. and Tavlarides L. L., AIChEJ, 40, 395,407, 1994.