THE IMPORTANCE OF DROP COALESCENCE IN A STIRRED LIQUID–LIQUID EXTRACTION (KÜHNI) COLUMN

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ABSTRACT
Experiments performed at low agitation speeds, using the high interfacial tension toluene (dispersed)-water (continuous) system, in a pilot plant Kühni column, show that coalescence events are generally present and cannot be neglected in column-type liquid-liquid contactors, especially when the goal is the accurate modelling of the transient processes for control purposes. In this work, the features of this behaviour and the applicability to this situation of a new, fast and precise, recently developed algorithm,[1] that describes the dynamic behaviour of an agitated column, is illustrated. Experiments were performed at low agitation speeds, using the above high interfacial tension system. The simulations show reasonably good agreement with the experimental results and provide insight on the underlying behaviour mechanisms and important guidelines for the accurate modelling of the dynamics of the column.

Keywords: Kühni column, drop coalescence, drop breakage, column dynamics.

INTRODUCTION

Mechanically agitated devices for liquid-liquid contact in column extraction equipment have acquired great importance, thanks to their ability to create large interfacial areas, which promote mass transfer. The capacity and mass transfer efficiency of these columns have been shown to be greatly influenced by the hydrodynamic and mixing effects caused by drop interactions (breakage and coalescence).

In recent years, many authors have spent great effort in modelling these columns and in the experimental validation of their models. Models such as those of Cruz-Pinto,[2] Casamatta and Vogelpohl,[3] Laso *et al.*,[4] Tsouris *et al.*[5] and, more recently, Gerstlauer *et al.*[6] have been proved able to describe with reasonable accuracy the results of their authors' experiments, but most of them adopted significant simplifications, in order to make calculations feasible in reasonable time. The most important simplification commonly made is to ignore inter-drop coalescence, because the modelling of this effect is responsible for most of the computational effort required for the solutions.

However, even in strongly agitated columns, such as the Kühni type, coalescence is generally present, being responsible for mixing effects that promote higher mass transfer rates. Zamponi *et al.*[7] noted this fact, and explicitly recommended the inclusion of coalescence effects in future modelling efforts. More recently, Kentish *et al.*,[8] although using a simplified model, included coalescence effects to obtain good agreement with experimental data from their Kühni column. In addition, for control purposes, the agitation intensity may be an important manipulating variable and, in order to avoid flooding, its value may be lowered such as to adequately promote drop coalescence.

Recently, a simplified version of an algorithm previously developed by Ribeiro,[9] and further streamlined by Regueiras *et al.*,[10] has been proved able to accurately describe the transient behaviour of a continuous ideally agitated vessel, requiring only modest computer resources and very low computation times. Details of these models and corresponding algorithms are given elsewhere by Ribeiro *et al.*[11] and Regueiras *et al.*[10] A new powerful algorithm that combines the previous ones with a new transport model to describe the hydrodynamics of an agitated column has been developed by

Regueiras [1] and is used in the present work. Its application yields an encouraging agreement with the experimental results described below, which were obtained in a pilot plant Kühni column (University of Munich).

EXPERIMENTAL

The experimental work was carried out by Gomes[12] in a Kühni pilot plant column (150 mm inside diameter, active height of 2520 mm, divided into 36 stirred compartments). The test system was the equilibrated standard high interfacial tension toluene (dispersed phase)-water (continuous phase) system. Experiments at room temperature, under steady state conditions, were made both at normal and low levels of agitation. For flow capacities (superficial velocities) of about 16 $m^3/(m^2.h)$, for which the normal operating range is within 140-150 rpm for this column, the agitation range was extended down to 100 rpm. Drop size distributions (by a photoelectric technique) and local hold-ups (by an ultrasonic technique) were measured. Due to the low agitation intensity, local hold-ups were much lower than under normal conditions and, consequently, the sampling time needed for sucking in the required 1000 sample drops was much longer than usual.

The local hold-up response to step modifications of the agitation intensity was also measured, by bringing the agitation intensity down to 100 rpm, after steady state was reached at 140 rpm. Local hold-up measurements were also made when the agitation intensity was suddenly increased from 100 to 140 rpm.

THE MODEL

The Kühni column may be adequately described as a sequence of agitated vessels with back- and forward mixing effects on the movement of the dispersed phase along the column, using a population balance formulation, with Coulaloglou and Tavlarides's drop interaction frequencies.[13] The resulting equations were solved for each compartment by the numerical technique developed by Ribeiro[9] and further streamlined by Regueiras *et al.*[10]

A new transport model was developed to describe the drop and continuous phase motions between stages. This transport model is based on the one described by Cruz-Pinto[2] and directly calculates the slip velocities, taking into account the density effects and the drag relationships for the drops, as previously modelled by Barnea and Mizrahi.[14,15]

The backmixing effect was also described and modelled as a random deviation superimposed on the terminal velocities of the drops, taking into account the influence of the agitation intensity, as a function of the rotor peripheral speed. A geometrical constriction factor, C_R, which controls the drop passage between stages, was calculated as a linear function of the fractional free area of the separators, as suggested by Goldmann.[16] The equations for this model are given elsewhere by Regueiras.[1]

RESULTS AND DISCUSSION

Figure 1 shows experimental and simulated local drop size number distributions along the column height, for the indicated continuous (F_C) and dispersed (F_D) phase flows, at moderate (140 rpm) and low (100 rpm) agitation speeds. The values for the parameters of the drop population balance model used (which have not yet been fully optimised) were identical in both simulations and very similar to those used in previous work by Guimarães,[17] concerning agitated vessels, as well as (to within less than 20%) to the ones used by Coulaloglou[12] and Bapat,[18] which have been claimed to be nearly "universal constants" for uniformly agitated contactors.

At the lowest agitation speed, a marked effect of coalescence may be identified, even in the upper stages of the column. The larger drops obtained overall, and the corresponding larger drop diameters at the upper stages, are justified (and can be approximately predicted by the algorithm) by the increased coalescence relative to breakage, combined with the more rapid motion of such large

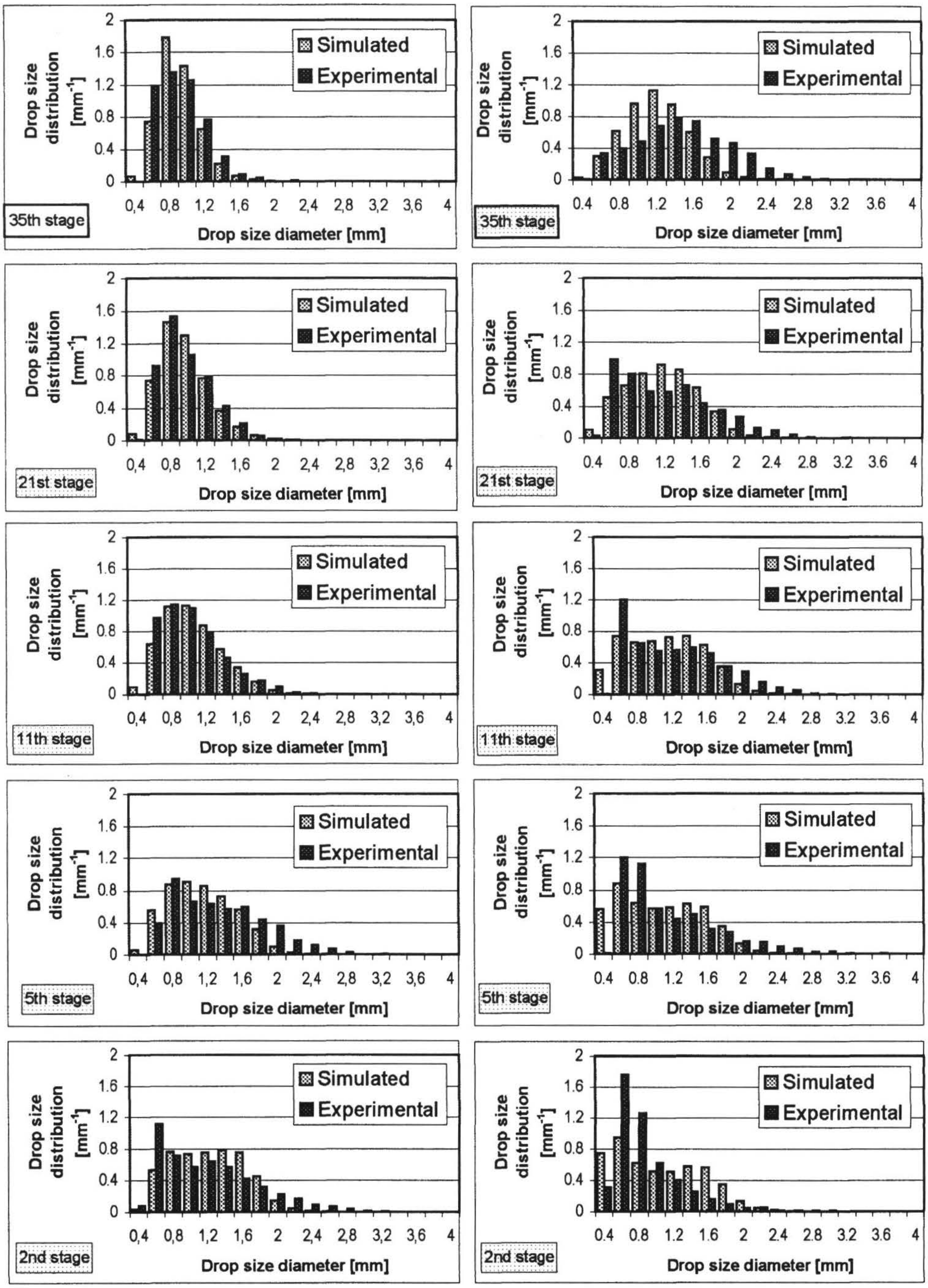

Figure 1. Drop size distributions for the equilibrated toluene-water system: F_C = 125 L.h^{-1}; F_D = 160 L.h^{-1}; left: I = 140 rpm, right: I = 100 rpm.

drops up along the column. The poorest agreement between simulated and experimental drop size distributions was obtained at the lowest instrumented stage (2nd) of the column (also the closest to the inlet dispersed phase distributor, for which experimental drop size data could not be measured, thus having to be grossly estimated).

Additionally, it may be noted that, at 100 rpm, drop coalescence combined with less intensive breakage is responsible for the bimodal shape of the drop size distributions (as also approximately predicted by our model), which dies out only at the highest stages of the column, while the distributions are unimodal at 140 rpm, except at the lowest stages.

It is somewhat intriguing that, at the column's 2nd stage, the experimental drop size distribution is wider and has higher average drop size at 140 rpm than at 100 rpm. At this stage it is also bimodal at 140 rpm, although this feature is quickly lost higher up in the column, whereas such bimodal character appears only at higher stages at 100 rpm, finally to disappear at the highest stages. Here, the average drop size stabilises close to the higher modal size. The same features were found in various other experimental runs at low agitation rates.[12]

These observations, and the way the darker (experimental) size distribution shifts and widens its shape along the column, can only be explained by increased drop coalescence relative to breakage at 100 rpm.

It is also reasonable to presume that, at the indicated flows, the distributor produces static drop size distributions (detectable by photography or any stage sampling method, like the one used) closer to those found at the 2nd stage for the lowest agitation rate. These, for more intensive agitation, given the higher local hold-up, will quickly tend to coalesce, yielding the bimodal distribution found at the bottom stages. Higher up in the column, however, it will quickly become unimodal, with a clearly decreasing average drop size.

Considering that these are as yet non-optimised simulations, the agreement obtained for the local hold-ups may also be considered satisfactory, within the variation range of repeated experimental runs, as indicated in Figure 2.

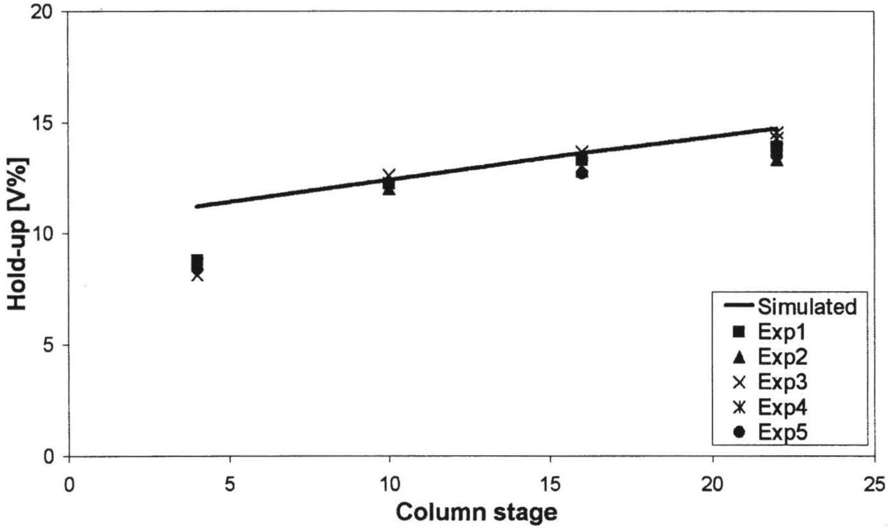

Figure 2. Hold-up profiles at steady-state – $F_C = 125$ L.h^{-1}, $F_D = 160$ L.h^{-1}, I = 140 rpm

The same coalescence effect becomes apparent when comparing the local dispersed phase hold-up values experimentally obtained before and after sudden step changes in the agitation rate (Figure 3). In this case, it may be noticed that the dispersed phase hold-up variation between the 16th and the 22nd stages is reversed, after an agitation rate change from 140 to 100 rpm. This may be understood as a result of increased coalescence/breakage frequency ratios, as the drops are broken down along the column height.

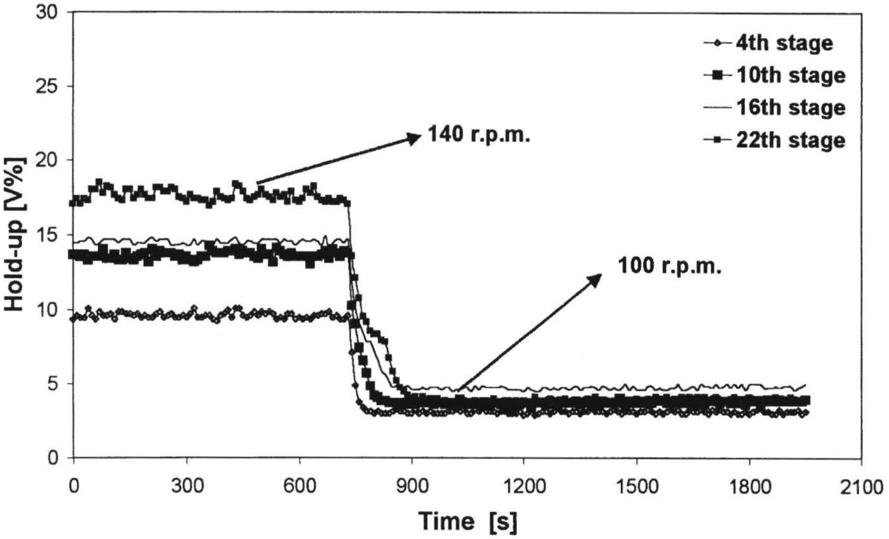

Figure 3. Hold-up profiles before and after sudden step changes in the agitation rate- F_C = 125 $L.h^{-1}$, F_D = 160 $L.h^{-1}$.

CONCLUSIONS

Although the operating range explored in this paper is not representative of normal steady state conditions, inter-drop coalescence cannot be neglected, when predicting (for control purposes) both the steady state and dynamic behaviour of liquid-liquid extraction columns.

A new, powerful, precise and fast algorithm to simulate the hydrodynamic behaviour of a mechanically agitated column (Kühni type) has been developed.

The algorithm is able to predict the local drop size distributions and the local hold-up profiles of a pilot plant Kühni column.

The agreement with the experimental data obtained in the pilot plant suggests that Coulaloglou and Tavlarides' population balance model may be appropriate, as well as, with minor modifications, the values for the breakage and coalescence parameters used by these authors for agitated vessels.

Further optimisation of the interaction constants and transport parameters is possible and is being pursued, along the lines of our previous and current work on mixers, for comprehensive equipment performance predictions.

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