Research Article

Experimental Measurements of Drop Size Distributions in 30 mm Diameter Annular Centrifugal Contactor with 30% TBP-Nitric Acid Biphasic System

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For design and development of liquid-liquid extraction systems, it is essential to have an accurate estimation of hydrodynamic and mass transfer characteristics of the employed contactor. In the present study, experimental evaluations consisted primarily of determining the maximum solution throughput that could be processed without cross-phase contamination at a given rotor speed, O/A flow ratio, and organic-aqueous solution pair in a 30 mm bowl diameter centrifugal contactor. In addition, analysis included experimental drop size determinations as well as holdup determination. The experimental drop size distributions are expected to be helpful for modeling work.

1. Introduction

Centrifugal contactors represent an efficient class of solvent extractors as compared to conventional system of columns and mixer-settlers. Based on the construction, these can be classified as differential and discreet (staged) contactors. For details, user is referred to Laddha and Degaleesan [1]. For staged variants, essentially design of mixing as well as settling zones differ; for example, original (SRL) Savannah River Lab design was having a paddle mixer and a centrifugal settler, whereas contemporary (ANL) Argonne National Lab design is based on annular mixing zone coupled with a centrifugal settler. Advantages of centrifugal contactors include low floor area as well as low head space requirement, lower inventory, elimination of interstage pumping, higher mass transfer efficiencies, and a better settling due to high “ g ” separation. Design philosophy was explained by Leonard et al. [2], and contemporary research work was reviewed by Vedantam and Joshi [3]. In this work, hydrodynamics in a biphasic system were studied with 30 mm bowl diameter centrifugal extractor, designed and developed indigenously. The salient dimensions of this extractor are listed in Table 1.

2. Importance of Drop Size Distribution Determination in Centrifugal Extractors

A better understanding of the drop breakage and coalescence phenomena inside solvent extractor is required for robust design. The rate of mass transfer in liquid-liquid dispersions created in solvent extractors solely depends on the interfacial area, mass transfer coefficient, and the degree of mixing of the two phases. The interfacial area is related to holdup by the following relation:

$$a = \frac{6\phi}{d_{32}}, \quad (1)$$

where d_{32} is the Sauter mean diameter and ϕ the volume fraction of the dispersed phase. It is commonly observed that d_{32} is proportional to the mean drop diameter (d_{mean}). Therefore, most of the investigators have attempted to predict (d_{mean}) as a function of operating parameters and physical properties of the system. During the generation of liquid-liquid dispersion in liquid-liquid contactors, there is a continuous breakup

TABLE 1: Dimensions of the annular centrifugal extractor used in the study.

Separation zone height	74.5 mm
Organic weir diameter	15.0 mm
Aqueous weir diameter	17.4 mm
Annular gap	4.5 mm
Stationary radial bottom vane height	4.0 mm
Rotor bowl entry hole dia	8.0 mm

and coalescence of drops occurring simultaneously. Given a sufficient time, a dynamic equilibrium is attained and resulting in a drop size distribution. The knowledge of maximum stable drop diameter (d_{max}) permits estimation of transfer rates in liquid-liquid dispersions generated in centrifugal contactors. As ϕ increases, a also increases causing increase number of droplets. This caused increase in mass transfer coefficients due to rapid coalescence and redistribution of drops. Thus, it is advantageous to use dispersion with larger holdup. However, there is a limit for holdup for uninterrupted operation, beyond which dispersion can invert. The continuous phase becomes dispersed phase and vice versa. This inversion phenomenon results in unstable operation of extraction equipment.

Very few experimental measurements of drop sizes and drop size distributions for centrifugal extractor operation are available. Arafat et al. [4] and recently Schuur et al. [5] have reported such data.

3. Experimental Setup and Procedure

3.1. Experimental Procedure for Centrifugal Extractor Operation. The centrifugal contactor was tested under a variety of conditions involving different flow ratios and rotor speeds to evaluate its hydraulic characteristics. Solution employed for these tests were nonradioactive. Tests were made with an aqueous phase of 0.1M HNO₃ and an organic phase of 30% TBP in normal paraffinic hydrocarbon (NPH). Speed of the three-phase miniature motor was controlled by a solid state frequency controller. Hydraulic performance was measured over a range of rotor speeds 2000 to 4000 rpm (33.33 rps to 66.67 rps) and for aqueous to organic flow ratios which are 0.1 to 4.0. Two different configurations of peripherals were used for two different modes of operation.

The procedure was as follows: (1) aqueous phase (continuous phase) pump turned on; (2) starting of centrifugal extractor motor; (3) organic phase (dispersed phase) pump was turned on. After through mixing, sampling was performed after 10 minutes, this interval being generally required for flow rate and temperature stabilization. Sample was taken in a 20 mL tube. It was then allowed to settle for 3 minutes. After the interface was clear for both the solutions, total level was noted and level of aqueous phase was also noted, thereby getting the volume of organic as well as aqueous. Then, holdup was found out by dividing the value of volume of dispersed phase by total volume taken in the tube.

FIGURE 1: Operable zone of 35 mm dia centrifugal extractor used in this study. Rotational speed was 50 rps.

FIGURE 2: Plot of total throughput versus O/A ratio at 50 rps for flooding conditions.

FIGURE 3: Variation of holdup with organic flow rate at 66.7 rps.

FIGURE 4: Variation of holdup with organic flow rate at 50 rps.

FIGURE 5: Variation of holdup with organic flow rate at 66.7 rps.

3.2. Measurement of Size Distribution of Drops. Drops of dispersed phase are formed during mixing. To get an accurate estimate of mass transfer area generated during mixing, drop size distribution is to be measured. In this study, drop size distribution measurement was carried out only for organic dispersion. Equipment used for drop size measurement was a laser-based drop size analyzer (Model CIS-100 coupled with LFC-100, from M/S Galai, Israel, 0.6–3000 μm range). For drop size measurement, a particular flow rate of aqueous phase (continuous phase) was fixed, and the dispersed phase (organic phase) was varied from low to high till inversion occurred. For each run, the dispersion was captured (about 2–3 mL) in 10–15 mL sodium dodecyl

FIGURE 6: Variation of drop sizes for varying holdup values at 33.7 rps rpm.

FIGURE 7: Variation of drop sizes for varying holdup at 2945 rpm.

sulphate (3% w/w in de-mineralized water) in a beaker. The surfactant hindered the coalescence of the dispersed phase drops, and this mixture was transferred to drop size analyzer compartment. Additional surfactant solution was added to the analyzer compartment. The stirrer speed of the analyzer was maintained at 15 rpm, and the flow rate through the cell was maintained at 10 mL/min. There may be some coalescence in the process of measurement. However, by keeping the amount of extraneous agent added constant and by completing the analysis in the roughly same time, the amount of errors can be assumed to be constant. The measurement of the drop sizes continued till 100% of the drops were processed ultimately resulting in d_{32} and other statistical parameters.

FIGURE 8: Variation of drop sizes for varying holdup at 3960 rpm.

FIGURE 9: Experimental drop size distribution for acidified water 30% TBP/commercial dodecane system at O/A ratio = 0.666 with CE bowl speed of 33.33 rps. X-axis is drop size in μm . Y-axis is in arbitrary units.

4. Result and Discussions

4.1. Establishment of Operability Zone. The individual phase flow rate was varied to achieve a maximum flow without flooding for each combination. Figure 1 shows operable limits of limiting aqueous and organic flows. The limiting throughput, as sum of organic and aqueous phase flow rates, has been shown in Figure 2, which may be taken as an ultimate capacity at the given O/A ratio.

4.2. Holdup. Dispersed phase holdup is defined as the fraction of volume occupied by the dispersed phase. The interfacial area available for mass transfer in a countercurrent extraction depends upon the volume fraction of dispersed phase as well as mean droplet size. It is therefore important at the design stage to be able to predict these quantities for any given system, contactor geometry, and set of operating conditions. From operational point of view, knowledge of

TABLE 2: Selected results of experimental drop size measurements for 0.01 N/30% TBP/commercial dodecane solvent aqueous pair.

O/A ratio	Speed rpm	d_{32} μm	d_{max} μm
0.162	2000	34.64	31.45
0.205	2000	60.25	58.80
0.333	2000	42.43	40.56
1	2000	50.32	49.86
0.143	3000	33.50	29.59
0.143	3000	73.51	71.41
0.7	3000	50.47	50.95
0.162	4000	37.06	36.00
0.24	4000	35.12	30.59
0.443	4000	33.60	31.41
0.613	4000	39.65	37.56

FIGURE 10: Variation of d_{32} with varying O/A ratio.

the dispersed phase holdup is also essential for inventory purposes. Variation of holdup is shown in Figures 3, 4, and 5 for different rotational speeds. Variation of drop sizes with holdup is shown in Figures 6, 7, and 8 for different rotational speeds.

4.3. Results for Drop Size Distribution Measurements and Drop Sizes Estimation. The aqueous phase consisted of acidified demineralized water. Organic phase was 30% TBP + 70% commercial dodecane. Figure 9 shows the representative experimental drop size distribution for one combination of operating parameters like different O/A ratios and rotor speeds. The numerous graphs for other combinations could not be included due to limitation of space. During these tests, variation of d_{32} and d_{max} versus O/A ratios is shown in Figures 10 and 11. Selected data are also listed in Table 2. These data could be approximated as a weak function of O/A ratio

FIGURE 11: Variation of $d_{\max}$ with varying O/A ratio.

with small effect of rotational speed as shown in Figures 10 and 11. The experimental drop size distributions are expected to be helpful for modeling work.

5. Conclusions

Experimental drop size measurements and drop size distributions are reported for a centrifugal extractor of 30 mm bowl size for 30% TBP/dodecane/nitric acid solvent/aqueous pair. For no mass transfer regime, drop size distribution shows a weak dependence on phase flow ratio. However, estimated drop sizes are strong functions of holdup as evident from the variation graphs.

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