

**Research note**

## **Mean Drop Size and Drop Size Distribution in a Hanson Mixer-Settler Extraction Column**

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### **Abstract**

*Drop size distribution and Sauter-mean drop diameters have been measured in a pilot scale Hanson mixer-settler extraction column for two different liquid-liquid systems. The experiments were carried out for both mass transfer directions. The effects of agitation speed and dispersed phase and continuous phase flow rates have been investigated under a variety of operating conditions. Mean drop diameter axial profiles show a strong nonuniformity, while drop size distribution does not change significantly with the column height. Sauter mean diameter and drop size distribution are strongly affected by agitation speed and interfacial tension. Significant, but weaker are the effects of continuous phase and dispersed phase flow rates. An empirical correlation for mean drop size as a function of Weber number, dispersed phase holdup, and viscosities ratio is suggested. In a further correlation, the dispersed phase holdup is replaced by flow rates which will certainly be known in a practical case.*

**Keywords:** *Sauter-mean Diameter, Drop Size Distribution, Hanson Mixer-settler, Dispersed Phase Holdup*

### **1. Introduction**

Liquid-liquid extraction is one of the classical methods in separation technology and finds applications in the chemical and petroleum industries, hydrometallurgy, biotechnology, nuclear technology, food industries, waste management, and other areas. Among various types of the extractor, the multistage mixer-settlers are widely used in industries because they give a stable operation and high stage efficiency. They do, however, occupy a disproportionately large

ground area and require a large solvent inventory. In addition, some mixer-settlers must be equipped with a complicated pipe system with pumps to transport the liquids between two successive stages. So, the type of extractor column of high performance is desirable. Their advantage is their high efficiency, low costs with respect to the number of stages, solvent inventory, settler area, site area, maintenance etc. According to the manner of putting the mixer and the settler side by side and how to separate the

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two phases in the settler, many types of mixer-settler extraction columns have been reported. Steiner et al. [1] used a three dimensional lattice as a partition of the mixing stages. Horvath and Hartland [2] achieved high stage efficiency with a mixer-settler extraction column in which the inter-stage mixing was extremely small, while the throughput of the column was also small. Takahashi et al. [3] proposed a mixer-settler extraction column (MS column) in which a mixer and a settler were in vertical arrangement in each stage, and showed that both the throughput and stage efficiency increased with agitation speed [4]. Wirz [5] proposed a column with a mixer at the center of the column and a large settling space around the mixer.

Knowledge of the drop size is of fundamental importance in the design of extraction columns. It affects the dispersed phase holdup, the residence time of the dispersed drops, and allowable throughput. Furthermore, the mean drop size is used to determine the interfacial area of mass transfer and affects both the continuous and dispersed phase mass transfer coefficients. It is therefore important to have the ability to predict the drop diameter as a function of the column geometry, agitation conditions, physical properties of the liquid-liquid systems, and the direction of mass transfer [6-8]. In reality a distribution of drop sizes exists which results in both a distribution of drop velocities relative to the continuous phase and variable mass transfer rates.

A number of relationships have been proposed to describe the effect of column geometry, agitation conditions, and physical

properties of the liquid-liquid system on drop size. Most researchers agree on the effect of Weber number on drop size, while the observed holdup effects of dispersed phase on the drop size are different, depending on investigators [9]. This fact suggests that the drop coalescence behavior depends on liquid-liquid system investigated, and physicochemical properties of liquid-liquid systems play an important role in controlling the drop size.

Hanson mixer-settler extractor is a rectangular mixer-settler with the horizontal arrangement of the mixer and the settler in each stage, i.e., each stage in the mixer-settler extractor is piled up in a vertical direction. This column is designed to obtain efficient contacting of two phases and a high degree of phase inter-stage separation [10, 11]. In order to develop appropriate design procedures for Hanson mixer-settlers, the knowledge of average drop size and its distribution in terms of the operating variables and liquid physical properties is, thus, of some importance.

In the present work, results are reported on drop size determinations carried out in a pilot scale Hanson mixer-settler extraction column for two different liquid-liquid systems. The effects of agitation speed and continuous phase and dispersed phase flow rates have been investigated. Finally, two different empirical correlations are proposed for prediction of Sauter-mean drop diameter.

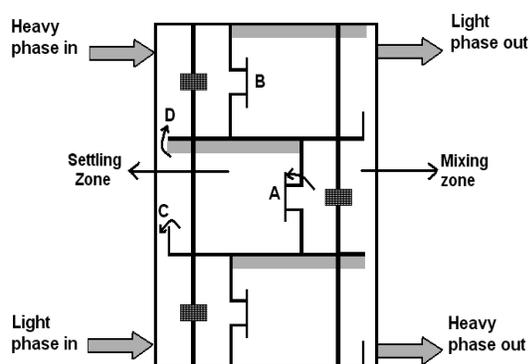
## **2. Experimental**

A pilot scale Hanson mixer-settler extraction column was used in these experiments. Two sides of the column were made of stainless

steel and the other sides were made of Plexiglas®. The inlets and outlets of the column were connected to four stainless steel tanks, each having a 200 liter capacity. The flow rates of the two phases were indicated by two rotameters; fine control being achieved by means of stainless steel needle valves.

The liquid level at the top of the column was automatically controlled by an optical sensor. A solenoid valve (a normally closed type) was provided at the outlet stream of the heavy phase. This valve received electronic signals from the optical sensor. When the interface location was about to change, the optical sensor sent a signal to the solenoid valve and the aqueous phase was allowed to leave the column by opening the diaphragm of the solenoid valve. The organic phase was also allowed to leave the column via an overflow.

Dispersion of the organic phase in each stage was achieved by means of a 6.5 cm diameter 6-bladed turbine agitator. The column consisted of seven stages, each 12 cm high. A schematic diagram of three stages is given in Fig. 1.



**Figure 1.** Schematic diagram of Hanson mixer-settler and flows of light and heavy phases

In each stage, the light and heavy phases were mixed in the mixing zone to provide initial contact. After that the unstable dispersion was transferred to a settler zone through the port 'A'. This port was located in the middle of a dividing wall and, in front of it, a vertical plate 'B' was placed to decrease the effect of centrifugal rotation in the settler so that the drop coalescence was improved. In the settler zone, the phases were separated, so the heavy phase was led to the previous stage mixer through a weir 'C' and the light phase was led to the next stage mixer through a port 'D'. The column geometry is listed in Table 1.

**Table 1.** Geometrical characteristics of the column used

Column Components	Dimensions (cm)
Mixer dimensions (length × width)	13×13
Settler dimensions (length × width)	26×13
Stage height	12
Column height	172
No. of stages	7
Agitator diameter	6.5
Blade height	1.3

The liquid-liquid systems studied were toluene-acetone-water (high interfacial tension) and n-butyl acetate-acetone-water (medium interfacial tension). These systems have been recommended by the European Federation of Chemical Engineering as official test systems for extraction investigations [12]. The content of acetone in dispersed or continuous phases was

approximately 3.5 (w/w %). Distilled water was used as continuous phase and technical grade of toluene and n-butyl acetate were used as dispersed phase. The physical properties of the liquid–liquid systems used in these experiments are listed in Table 2. In the present work, the values of physical properties have been assumed to correspond to the mean values of acetone concentration in the continuous and dispersed phases. The mean value of acetone concentration was obtained by averaging the values obtained at the inlet and outlet of the column.

**Table 2.** Physical properties of liquid systems at 20°C [12]

Physical property	Toluene-acetone-water	n-Butyl acetate-acetone-water
$\rho_c (kg/m^3)$	994.4–995.7	994.3–995.8
$\rho_d (kg/m^3)$	864.4–865.2	879.6–881.4
$\mu_c (mPa.s)$	1.059–1.075	1.075–1.088
$\mu_d (mPa.s)$	0.574–0.584	0.723–0.738
$\sigma (mN/m)$	27.5–30.1	12.4–13.2

Before the experiments, both phases were mutually saturated by circulation through the column. After filling the column with the continuous phase, the rate of agitation and continuous phase flow rate were set on desired values and the dispersed phase gradually admitted to enter into the column up to the selected volumetric flow rate. The interface position was then maintained at the desired height by using the optical sensor, and the system was allowed to reach steady state, which necessitated 2-3 changes of column volume. A photographic technique was used to measure the drop diameter,

since it proved to be more accurate and reliable than other techniques. When the conditions became steady, photographs were taken at different positions along the column. The drops were photographed using a Sony DSC-F828 digital camera. Drop dimensions were then determined using AutoCAD software. Drop sizes were converted to absolute dimensions by comparing measured value with packing size. In the case of non-spherical droplets, the major and minor axes,  $d_1$  and  $d_2$ , were measured and the equivalent diameter,  $d_e$ , calculated from Eq. (1).

$$d_e = (d_1^2 d_2)^{1/3} \quad (1)$$

The Sauter mean diameter was then calculated from Eq. (2).

$$d_{32} = \frac{\sum_{i=0}^n n_i d_i^3}{\sum_{i=0}^n n_i d_i^2} \quad (2)$$

where  $n_i$  is the number of droplets of mean diameter  $d_i$  within a narrow size range  $i$ . For each case, at least 400 drops were analyzed to ensure good statistical accuracy. In order to fulfill the overall experimental objective, more than eighty runs were carried out. The holdup of dispersed phase was measured by rapid withdrawal of a 100 ml sample from the mixers.

### 3. Results and discussion

The effect of agitation speed on the drop size distribution is shown in Fig. 2. In each Figure, distributions corresponding to the

different column heights have been included. It can be seen that the shape of the drop size distribution histograms does not change significantly along the column height. Drop size distributions are, on the other hand, found to be dependent on the agitation rate and interfacial tension. As can be seen in this Figure, higher agitation speed

leads to a narrower and more symmetric distribution around the mean value. Moreover, drop size distributions are found to be narrower and more evenly distributed about the mean size for the system of lower interfacial tension (Note that the scales in the following Figures are different).

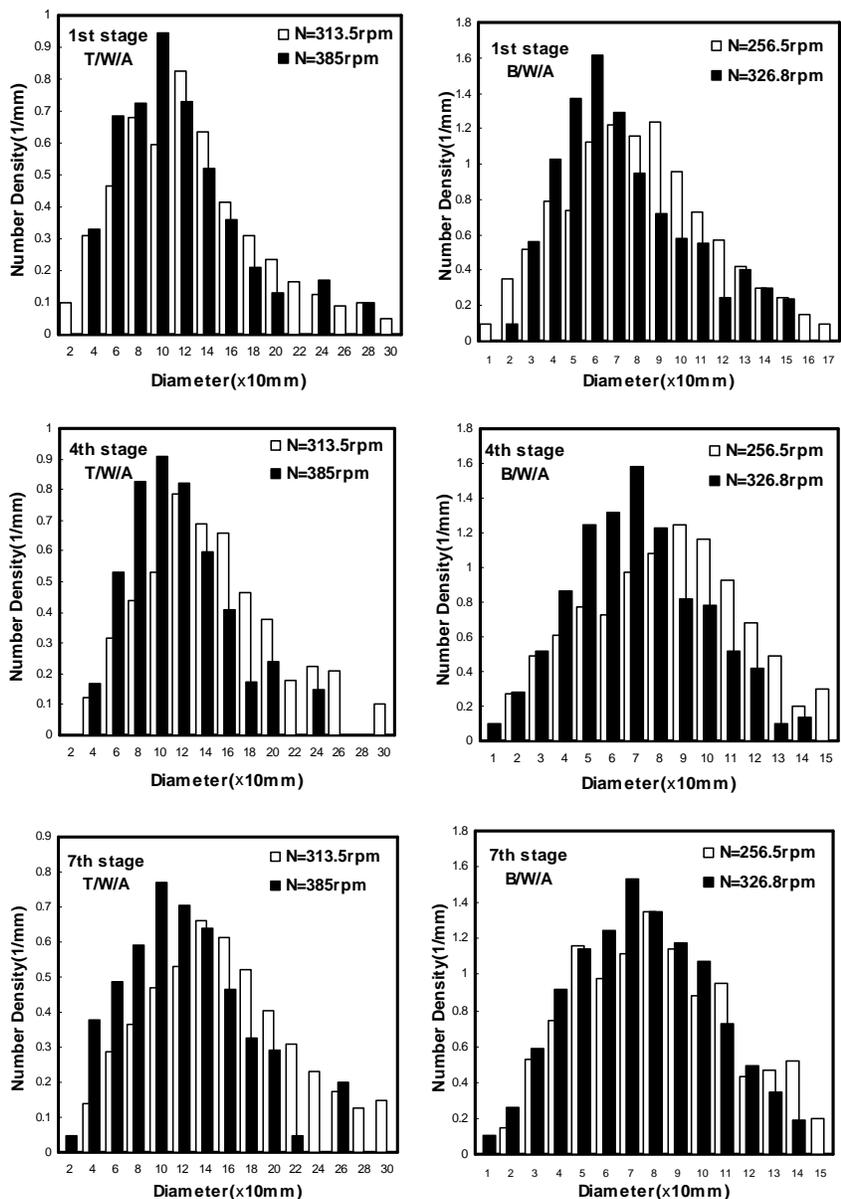


Figure 2. Effects of stage numbers on drop size distributions

The effect of continuous phase and dispersed phase flow rates are shown in Fig. 3. As can be seen in this figure, continuous phase and dispersed phase flow rates do not have a significant effect on the shape of drop size distribution. Changes in these variables, however, affect drop sizes by shifting the

distribution towards larger drops. These results are in agreement with the observations of Rincon-Rubio *et al.* [13] and Takahashi *et al.* [14]; their experiments show a similar trend in other types of mixer-settler extraction columns.

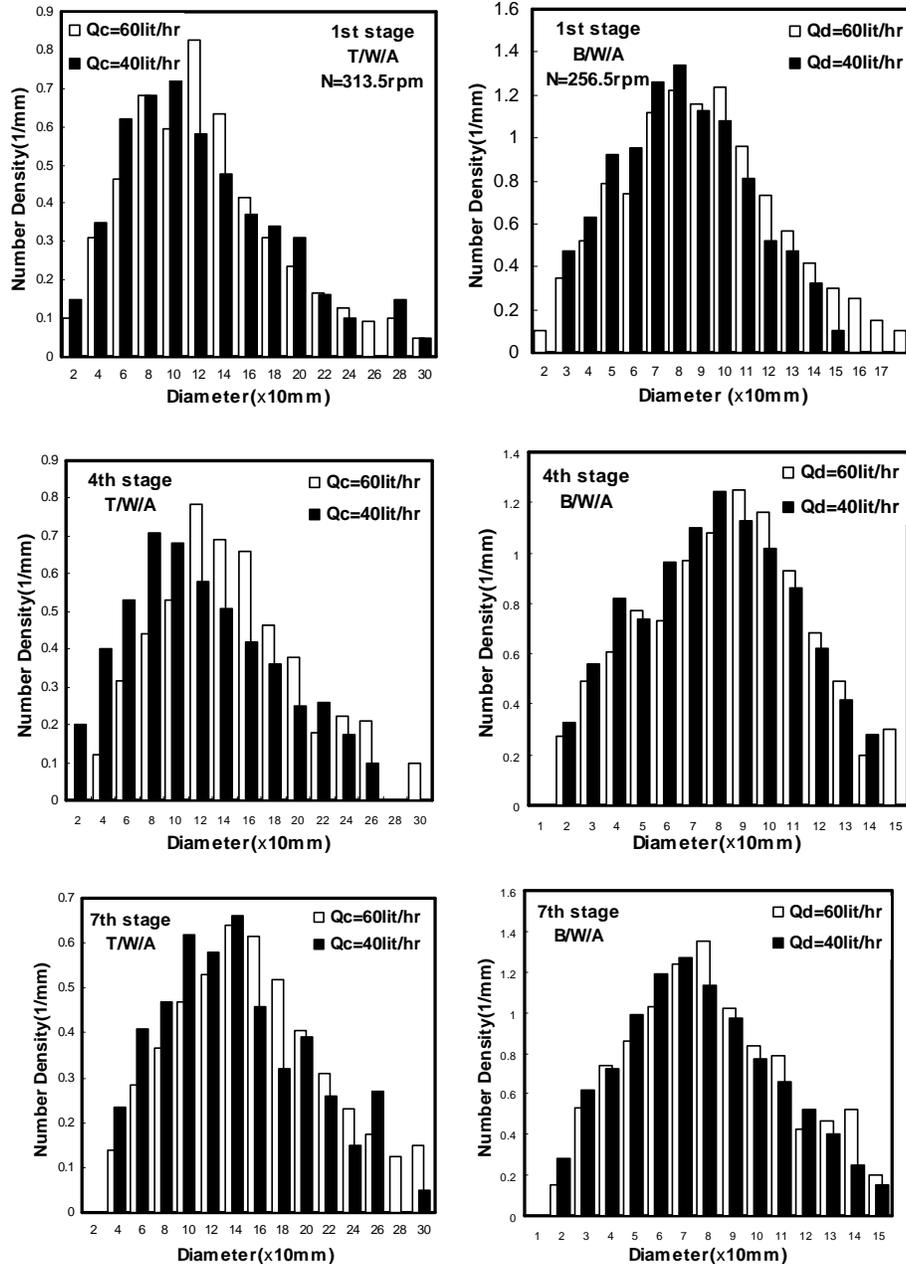


Figure 3. Effect of continuous phase and dispersed phases flow rates on drop size distributions

Typical variations in the mean drop size with the column height are given in Fig. 4. Three different curves, corresponding to three different rates of agitation, are given for each mass transfer direction. For  $c \rightarrow d$  direction, the large drops formed at distributor disappeared rapidly. This rapid decrease of the Sauter mean diameter along the column is due to the predominance of the drop breakage mechanism. On the contrary, for  $d \rightarrow c$  direction, the profiles show that the drops are still growing in the first part of the column and the mean size of drop population remains nearly constant after the second or third stages, due to the competition of coalescence and breakage. This result exhibits the influence of coalescence in a case where it becomes predominant. It is also observed that the mean drop size does not change significantly above the middle of the extractor. This observation indicates that, at a certain height inside the column, the rate of coalescence becomes equally important with drop breakage for both mass transfer directions.

Fig. 5 shows the effect of agitation speed on mean drop size for both systems and both mass transfer directions. As can be seen in this figure, agitation has a strong effect on the Sauter mean diameter. For both systems, it was found that as the agitation speed is increased, the mean drop size strongly decreases. The rise of agitation rate carries to higher shear stress and to intense drop breaking and consequently the Sauter mean diameter will decrease. However, at high agitation speed, the fall in the values of Sauter mean diameter is gradual and it can be conjectured that further breakup of dispersed phase drops into smaller ones is limited.

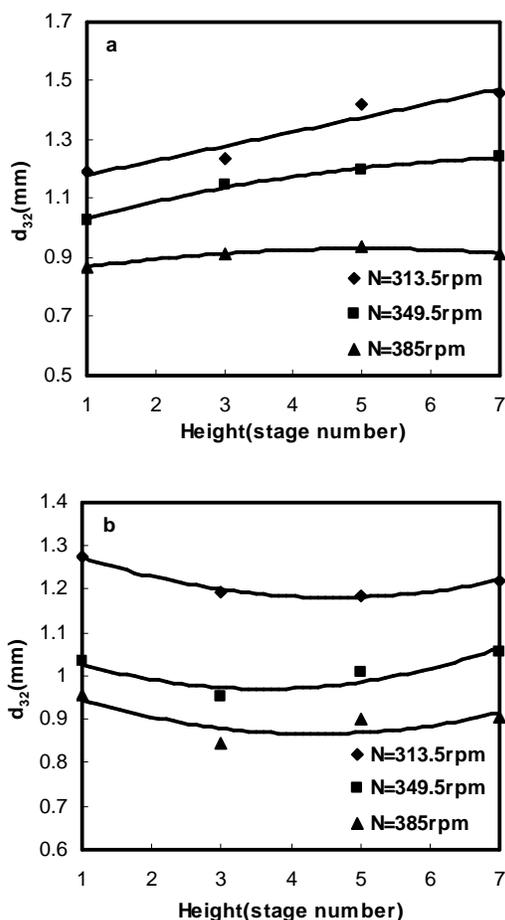
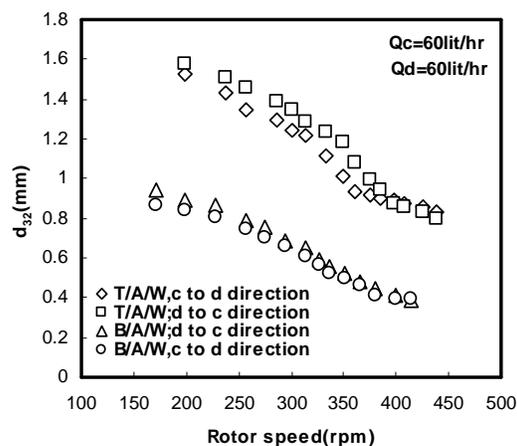


Figure 4. Variations in mean drop size with column height (a)  $d \rightarrow c$  direction (b)  $c \rightarrow d$  direction (Toluene-Acetone-Water System)

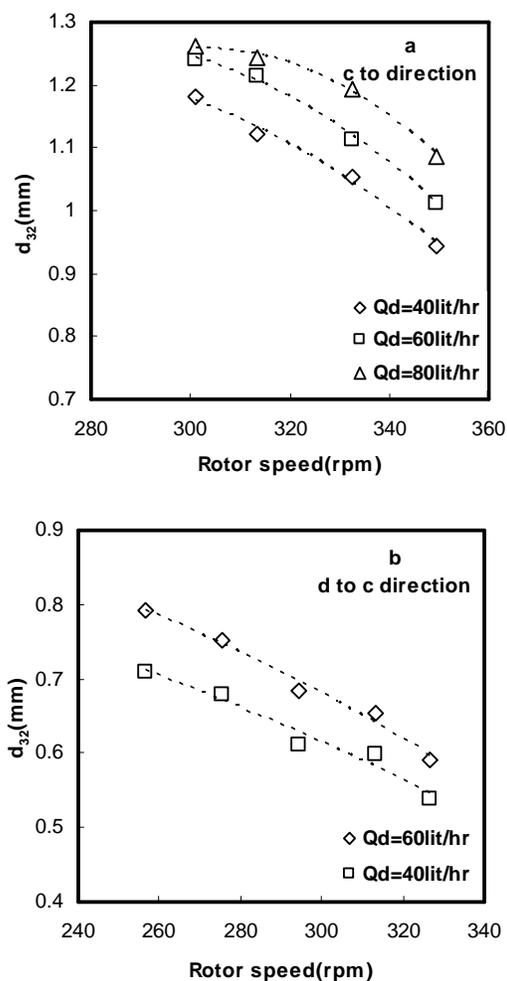
Possibly drop stabilization occurs due to the high turbulence in the continuous phase. The results confirmed that the drop size in this column changes with the direction of mass transfer. This fact has been well known for other types of the extraction columns. When the mass transfer occurs from the continuous to the dispersed phase, the concentration of the solute in the draining film between two approaching drops will be lower than the surrounding continuous liquid. For mass transfer in the opposite direction, the concentration will be correspondingly higher.

The resulting gradients of interfacial tension will retard drainage and inhibit coalescence in the former case and accelerate drainage and coalescence in the latter case. Therefore, this figure shows that  $c \rightarrow d$  transfer tends to produce smaller drops than the opposite direction. For all runs, the mean drop diameters are strongly affected by system properties. Smaller drops are generated from the medium interfacial tension system (n-butyl acetate-acetone-water) than are produced from the higher interfacial tension system (toluene-acetone-water).



**Figure 5.** Effect of agitation speed on mean drop diameter

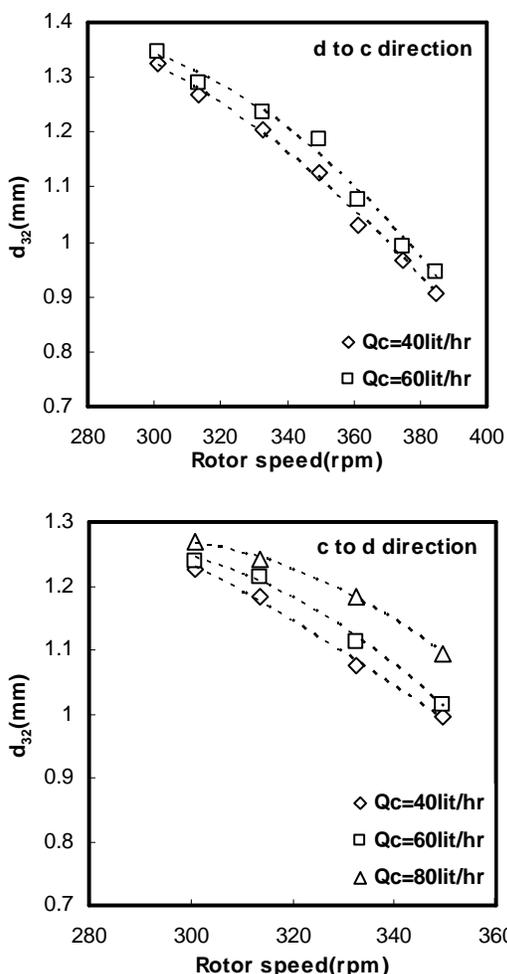
Fig. 6 illustrates the effect of dispersed phase flow rate on the average drop size. Increasing the flow rate of dispersed phase tends to increase the average drop size. As can be seen in Fig. 7, the effect of  $Q_d$  is weaker than the effect of agitation speed. For a 100% increase of  $Q_d$ , only a 10% increase in  $d_{32}$  was found. The increase in droplet size may be attributed to an increase in the coalescence rate due to the larger holdup values that are observed as  $Q_d$  increases.



**Figure 6.** Effect of dispersed phase flow rate on Sauter mean diameter (a)-Toluene-Acetone-Water (b)-Butyl acetate-Acetone-Water

As shown in Fig. 7, the flow rate of the continuous phase has a comparable influence on the mean drop diameter. Increasing the continuous phase flow rate was found to increase the Sauter mean diameter. This increase in the mean drop size may be attributed to a decrease in residence time accompanying the decrease in the holdup. Comparing Figs. 7 and 8, it can be seen that the effect of dispersed phase flow rate on  $d_{32}$  is larger than that of continuous phase flow rate. It should be noted that the dotted lines in

Figs. 6 and 7 are only a guide tool for better vision.



**Figure 7.** Effect of continuous phase flow rate on Sauter mean diameter (Toluene-Acetone-Water System)

### 3.1. Modeling of mean drop size

One of the main objectives in this study is to generate a reliable correlation to represent the mean drop diameter in Hanson mixer-settler columns.

Currently the basic theoretical concept of Hinze [15] is commonly applied to calculate the Sauter mean diameter in stirred liquid-liquid dispersion. The maximum stable drop

diameter  $d_{max}$  is calculated under the assumption that the turbulent kinetic energy of drop  $E_{kin}$  equals the interfacial energy  $E_{\gamma}$ . The Weber number  $We_{d_{max}}$  describes the ratio between  $E_{kin}$  and  $E_{\gamma}$ . This value marks a critical state where the drop is still stable:

$$We_{d_{max}} = \frac{E_{kin}}{E_{\gamma}} = \frac{\rho w'^2 d_{max}}{\sigma} \quad (3)$$

Under the condition that the energy dissipation rates in the stirred tank are spatially uniform, Eq. (3) results in an equation for the maximum drop diameter:

$$\frac{d_{max}}{D_i} = C_1 \cdot \left( \frac{N^2 D_i^3 \rho}{\sigma} \right)^{-0.6} = C_1 \cdot We^{-0.6} \quad (4)$$

According to several authors the maximum drop diameter correlates linearly with the Sauter mean drop diameter:

$$\frac{d_{32}}{D_i} = C_2 \cdot We^{-0.6} \quad (5)$$

Following the findings of previous investigations on drop size distribution in agitated vessels and extraction columns, the effect of coalescence drop size can be expressed in terms of dispersed phase holdup as:

$$d_{32} = C_4 (1 + C_3 \phi) We^{-0.6} \quad (6)$$

Therefore, the exponent -0.6 of the Weber number in relation to  $d_{32}$  is of great importance for scale up [16].

On this basis, the following correlations for each mass transfer direction are derived in

terms of Weber number, dispersed phase holdup, and viscosity ratio for two different systems (Toluene-Acetone- Water and Butyl acetate-Acetone-Water) by using the least squares method:

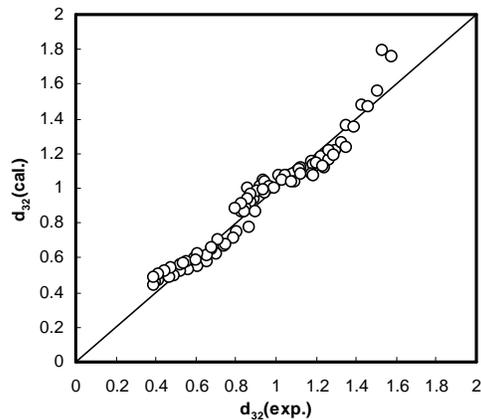
$$\frac{d_{32}}{D_i} = 0.197(1 + 3.04\phi)We^{-0.6} \left( \frac{\mu_d}{\mu_c} \right)^{-1.27}$$

(d→c direction) (7)

$$\frac{d_{32}}{D_i} = 0.23(1 + 2.24\phi)We^{-0.6} \left( \frac{\mu_d}{\mu_c} \right)^{-1.14}$$

(c→d direction) (8)

which are based on 95 data points. The average relative deviation in predicted values of  $d_{32}$  from experimental points is 7.78%. The comparison of experimental results with those calculated by Eqs (7) and (8) is shown in Fig. 8.



**Figure 8.** Comparison of experimental  $d_{32}$  with those calculated from Eqs. (7) and (8)

In practice, the dispersed phase holdup is not known and should be calculated from another correlation. If predicted values of the holdup are used in Eqs. (7) and (8) to calculate mean drop size, an additional error will be introduced. An alternative drop size

correlation in terms of  $N$ ,  $\sigma$  and continuous and dispersed phase superficial velocities is therefore developed. The results are:

$$d_{32} = 0.187N^{-0.83}\sigma^{0.89}V_d^{0.05}V_c^{0.15}$$

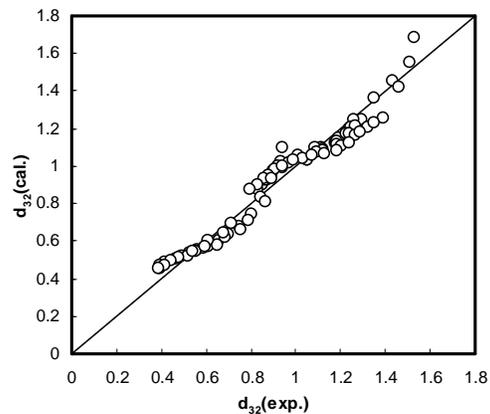
(d→c direction) (9)

$$d_{32} = 0.31N^{-0.51}\sigma^{0.81}V_d^{0.02}V_c^{0.13}$$

(c→d direction) (10)

which reproduce the experimental data with an average relative deviation of 7.09%. These equations show that agitation speed and interfacial tension of liquid-liquid systems are the variables having the largest effect on  $d_{32}$ . The effect of both continuous and dispersed phase superficial velocities is small and positive, that is, increasing either results in higher drop size.

Fig. 9 shows the agreement between experimental and predicted data, using correlations (9) and (10). Figs. (8) and (9) indicate that the suggested correlations can predict Sauter-mean drop diameter with high accuracy and both methods can be used for prediction of  $d_{32}$ , although equations (9) and (10) do not need any experimental values.



**Figure 9.** Comparison of experimental  $d_{32}$  with those calculated from Eqs. (9) and (10)

#### 4. Conclusions

The effects of agitation speed, dispersed phase and continuous phase flow rates, interfacial tension, mass transfer direction, and the height of column on drop sizes were studied experimentally in a pilot scale Hanson mixer-settler. The results showed that mean drop size and its distribution are drastically affected by agitation speed as well as interfacial tension, while they are only slightly dependent on phase flow rates. Drop size distribution shape varied little along the column height. The experiments revealed that mass transfer direction had a significant effect on drop behavior along the column height, although at a certain height above the middle of the column mean drop size became constant for both mass transfer directions. Additionally, in the present work, two empirical correlations are derived to predict the Sauter mean drop diameter from the measurement of more than 30000 drops. The predictive correlations can reproduce the experimental data with very high accuracy. The present work is useful to those that apply Hanson mixer-settler column and the results of this study can reduce the experimental work associated with the design of this type of extractor.

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