

## **Prediction of Mean Drop Size in Pulsed Packed Extraction Columns**

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### **Abstract**

*Sauter mean drop sizes have been measured in a pulsed packed extraction column for two liquid systems with and without mass transfer conditions. The effects of pulsation intensity, phase flow rates, and interfacial tension on drop size have been investigated under a variety of operating conditions. The drop size is influenced mainly by pulsation intensity and interfacial tension. Significant, but weaker, are the effects of continuous and dispersed phase flow rates. A precise correlation is proposed for predicting mean drop size in terms of operating variables, physical properties of the liquid systems and mass transfer direction. Good agreement between prediction and experiments is found for all operating conditions that were investigated.*

**Keywords:** *Pulsed Packed Column, Sauter Mean Diameter, Drop Size, Holdup*

### **1. Introduction**

Liquid-liquid extraction is one of the classical methods in separation technology and finds applications in the chemical and petroleum industry, hydrometallurgy, biotechnology, nuclear technology, food industry, waste management, and other areas. The efficiency of liquid-liquid contactors is primarily dependent on the degree of turbulence imparted to the system and the interfacial area available for mass transfer. The rate of mass transfer can be enhanced by pulsating motion imparted to the liquids by an external mechanical or electronic device.

Internal mechanical parts are eliminated, leakage is minimized, and the pulsator can be isolated [1].

Pulsing improves the rate of mass transfer by reducing the size of drops. Chantry and co-workers [2] have shown that the height of a packed column required to affect a given degree of extraction is reduced by a factor of three, under pulsation. The pulsed columns have a clear advantage over other mechanical contactors when processing corrosive or radioactive solutions, since the pulsing unit can be remote from the column. The absence of moving mechanical parts in such columns

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obviates repair and servicing. These advantages have led to the application of these columns in chemical, biochemical and petroleum industries besides nuclear fuel reprocessing.

A great deal of experimental effort has been expended principally for the purpose of evaluating column performance for design and scale-up. It is well known that the mean drop diameter is an important parameter in the study and design of the extraction column. In the modeling of liquid-liquid extraction columns, where a dispersed phase exists as discrete drops, an average volume-surface diameter is commonly used to predict the interfacial area for mass transfer, contact times and mass transfer coefficients [3-9]. In order to develop appropriate design procedures for a given type of extraction column, a knowledge of average drop size in terms of operating variables, liquid physical properties, and mass transfer direction is thus of paramount importance.

Although pulsed columns use sieve plates, packing, and discs and doughnuts, most investigations reported have dealt with the sieve plate type [10-12] and there is little experimental data available on drop size in pulsed packed columns.

The present study has examined the influence of operating variables including the pulsation intensity as well as continuous and dispersed phase flow rates on mean drop size in a pulsed packed column of 76.2 mm diameter. An empirical expression including the effects of operational variables, physical properties of the liquid systems and mass transfer direction on mean drop size is proposed.

## 2. Experimental

The present experimental work was carried out in a pilot plant pulsed packed column of 76.2mm diameter which was filled with 1.8m height of packing. Stainless steel Raschig rings with 1.9cm diameter were used as packing and the fractional packing voidage was 0.68. Compressed air was used as pulsation system. The inlets and outlets of the column were connected to four tanks, each of 100 liter capacity. The flow rates of two phases were indicated by two rotameters. The interface location of two phases 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 heavy phase. This valve received electronic signals from the optical sensor. When the interface location was going 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 allowed to leave the column via an overflow. A schematic diagram of the apparatus is given in Fig. 1.

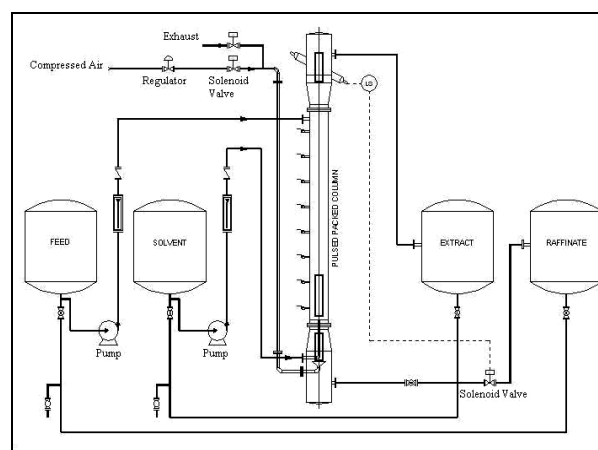


Figure 1. Schematic of the pulsed packed column

The experimental systems were toluene-water (high interfacial tension) and *n*-butyl acetate-water (medium interfacial tension). Acetone (4% wt) was used as solute in mass transfer conditions. These systems have been recommended by the European Federation of Chemical Engineering (E.F.C.E.) as official test systems for extraction investigations [13]. Distilled water was used as the continuous phase and technical grades toluene and *n*-butyl acetate of at least 99.5wt% purity were used as the dispersed phase. The physical properties of the liquid-liquid systems used in these experiments are listed in Table 1. It should be noted that, under mass transfer conditions, a degree of uncertainty surrounds the estimation of physical properties (particularly interfacial tension), since these vary not only with the inlet solute concentrations but also along the column. 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 1.** Physical properties of liquid systems at 20°C[13]

Physical property	Toluene/ water	<i>n</i> -Butyl acetate/ water
$\rho_c$ [kg/m <sup>3</sup> ]	994.4-995.7	994.3-995.8
$\rho_d$ [kg/m <sup>3</sup> ]	864.4- 865.2	879.6-881.4
$\eta_c$ [mPa.s]	1.059-1.075	1.075-1.088
$\eta_d$ [mPa.s]	0.574-0.584	0.723-0.738
$\sigma$ [mN/m]	26.5-36.0	11.7 -14.1

Before carrying out the experiments, both phases were mutually saturated by repeated circulation through the column, after which solute was added to the dispersed or continuous phase to give a concentration of about 4wt% acetone. The amplitude and frequency of pulsation were next adjusted to the desired values and, after filling the column with the continuous phase, the dispersed phase was introduced. The interface position was then maintained at desired height, and the system was allowed to reach steady state, which necessitated 3-4 changes of column volume. The sizes of drops were measured during steady state conditions.

A photographic technique was used to measure the drop diameter, since it proved to be more accurate and reliable than other techniques. 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) [14].

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

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

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

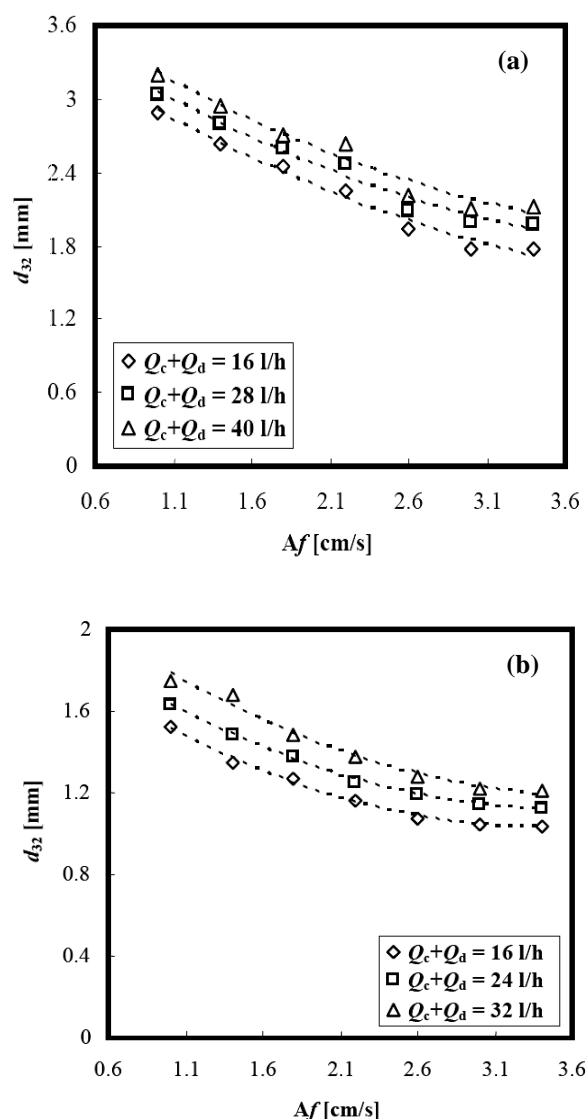
where  $n_i$  is the number of droplets of mean diameter  $d_i$  within a narrow size range  $i$ . At least 150 drops were analyzed for each determination.

### 3. Results and discussion

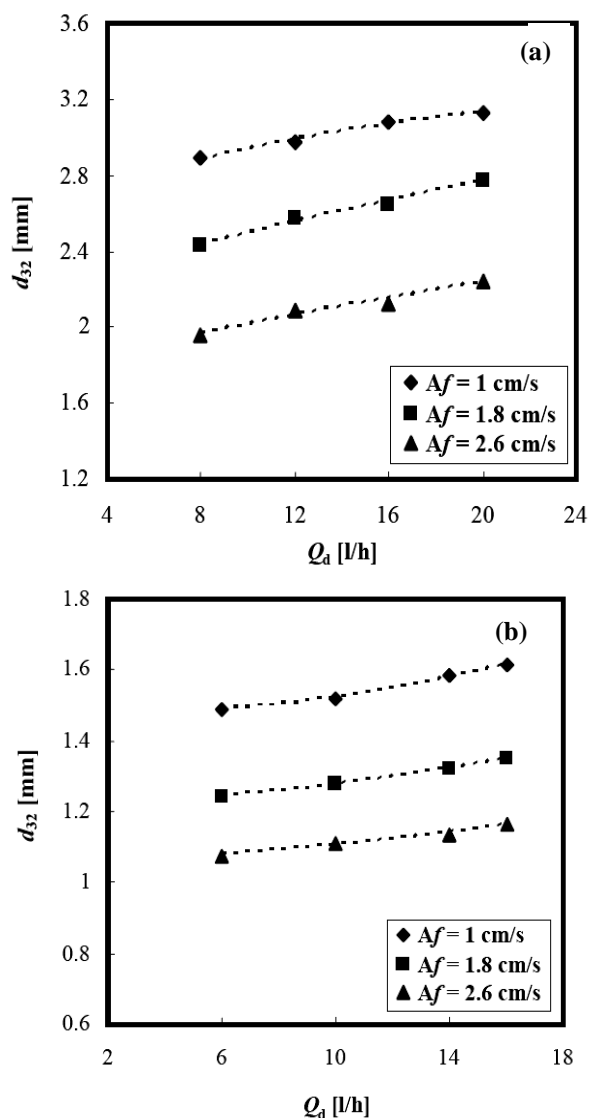
The main operating parameters found to affect mean drop size of the column were the pulsation intensity and phase flow rates. The effect of pulsation intensity on Sauter mean drop diameter is shown in Fig. 2. As expected, smaller drops are found at higher pulsation intensities. The rise of the pulsation intensity leads to intense drop breaking and consequently the Sauter mean diameter will decrease. However, at high pulsation intensity, the fall in the values of Sauter means the diameter is gradual. Droplet coalescence is enhanced at high pulsation intensity by the increasing probability of droplet collision. Thus at high pulsation intensity, the enhanced rate of coalescence overcomes the increased tendency for droplet breakdown and the drop size apparently stabilized. Fig. 2 also shows that larger drops are produced from the high interfacial tension system (toluene-water) than are generated from the lower interfacial tension system (n-butyl acetate-water). It can also be found that the effect of pulsation intensity on mean drop size of toluene-water system is larger than that of n-butyl acetate-water, because the breakup of dispersed phase drops into smaller ones is limited for the latter system due to its lower interfacial tension.

The effect of dispersed phase flow rate on mean drop size is shown in Fig. 3. Increasing the flow rate of dispersed phase tends to increase the average drop size. As can be seen in Fig. 3, the effect of dispersed phase

flow rate is weaker than the effect of pulsation intensity. 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.

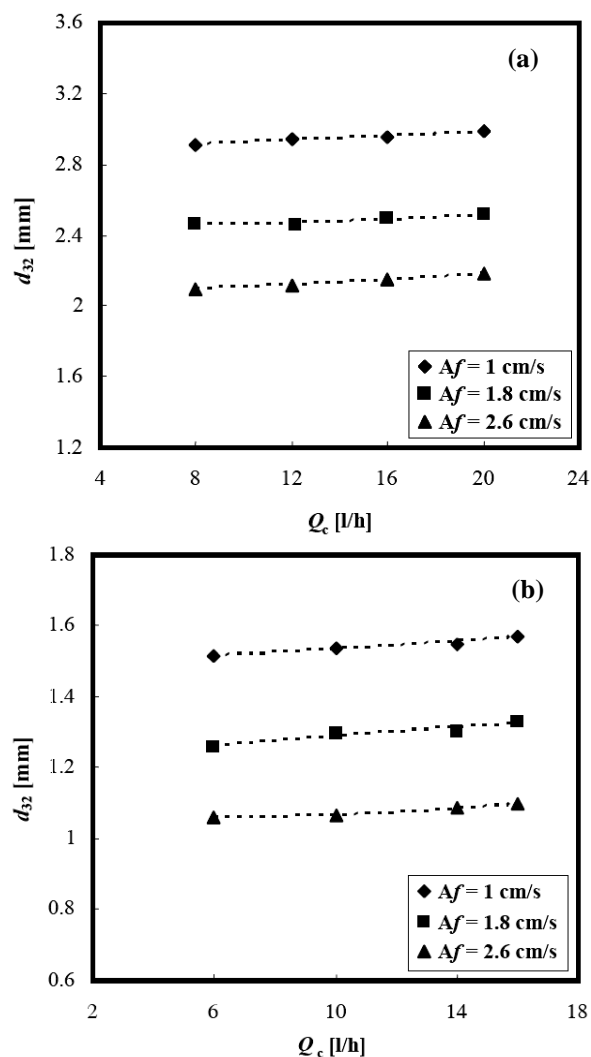


**Figure 2.** Effect of pulsation intensity on mean drop size (a) toluene-water (b) *n*-butyl acetate-water ( $Q_d/Q_c=1$ )



**Figure 3.** Effect of dispersed phase flow rate on Sauter mean drop size (a) toluene-water,  $Q_c=10$  l/h (b) *n*-butyl acetate-water,  $Q_c=8$  l/h

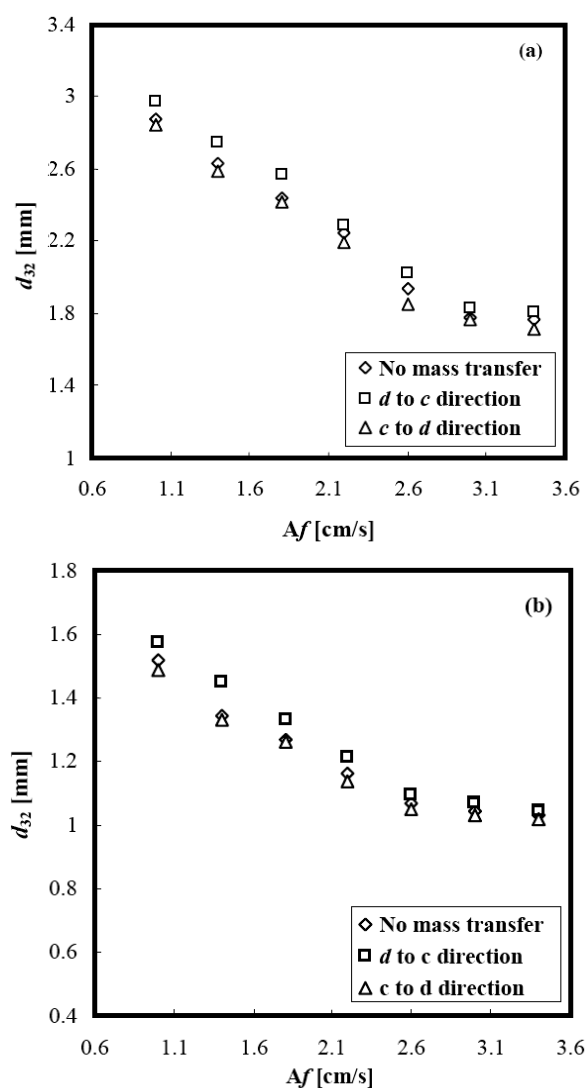
The effect of continuous phase flow rate on Sauter mean drop size is illustrated in Fig. 4. The mean drop diameter slightly increases with an increase in continuous phase flow rate. Increasing the continuous phase flow rate would be expected to increase residence time of drops due to the reduction in slip (relative) velocity between the drops and continuous phase. Increasing the residence time of drops enhances the probability for drop coalescence, leading to larger drops.



**Figure 4.** Effect of continuous phase flow rate on Sauter mean drop size (a) toluene-water,  $Q_d=10$  l/h (b) *n*-butyl acetate-water,  $Q_d=8$  l/h

The effect of mass transfer direction on mean drop size is given in Fig. 5. As can be seen in this figure, mass transfer of acetone from the dispersed phase to continuous phase results in a larger drop size. 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 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,  $c \rightarrow d$  transfer tends to produce smaller drops than the opposite direction.



**Figure 5.** Effect of mass transfer direction on Sauter mean diameter (a) toluene-water (b) *n*-butyl acetate-water ( $Q_c = Q_d = 8$  l/h)

#### 4. Prediction of Sauter mean drop diameter

One of the main objectives in this study is to generate experimental drop size data, and

then to derive from it, a reliable correlation to represent the mean drop diameter in pulsed packed columns.

Mean drop diameters in pulsed packed columns are usually predicted by the following semi-empirical correlation which has been obtained by Spaay *et al.* (1971) [15]:

$$\frac{1}{d_{32}} - 0.72 \left( \frac{\Delta \rho g}{\sigma} \right)^{0.50} = 6700 \left( \frac{1-\phi}{\phi} \right) \left( \frac{fA \rho_c}{\eta_c a_p} \right) \left( \frac{\eta_c^2 a_p}{\sigma \rho_c} \right)^{0.50} \left( \frac{\sigma a_p^2}{\Delta \rho g} \right)^{0.23} \quad (3)$$

In practice, the dispersed phase holdup is not known and should be calculated from another correlation. If predicted values of the holdup are used for prediction of drop size, an additional error will be introduced. For this reason, it is necessary to develop a correlation for Sauter mean diameter in pulsed packed columns in terms of operational variables and physical properties of liquid systems, which will certainly be known in a practical case.

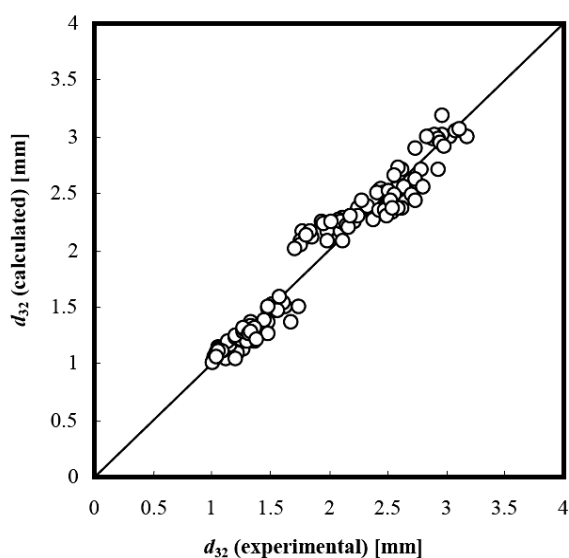
On this basis, correlation (4) is derived in terms of operating variables and physical properties of the liquid systems with varying constants for different mass transfer directions as follows.

$$d_{32} = c_1 \left( \frac{\sigma}{\Delta \rho g} \right)^{0.5} \left( \frac{\sigma}{Af \eta_c} \right)^{c_2} \left( 1 + \frac{V_d}{V_c} \right)^{c_3} \quad (4)$$

where

for no mass transfer	$c_1 = 52.45$	$c_2 = 0.30$	$c_3 = 0.07$
for d→c transfer	$c_1 = 55.19$	$c_2 = 0.33$	$c_3 = 0.10$
for c→d transfer	$c_1 = 51.04$	$c_2 = 0.32$	$c_3 = 0.02$

The constants of the correlation are obtained by the least squares method using the "EViews software" version 3.1. The comparison of experimental results with those calculated by the presented correlation is shown in Fig. 6. This figure indicates that the suggested correlation can estimate the dispersed phase Sauter mean drop size with very high accuracy. The Sauter mean drop size calculated with this correlation reproduces the experimental data with an average error of 6.16%.



**Figure 6.** Comparison of experimental data with calculated values

## 5. Conclusions

Design procedures for pulsed packed extraction column still depend on having large amounts of experimental data which are difficult and expensive to obtain. The aim

here was to try to reduce the experimental effort by proposing experimental correlation for the prediction of Sauter mean drop size. The effects of operational variables on mean drop size were investigated. The results showed that mean drop size was drastically affected by pulsation intensity as well as interfacial tension, while only slightly dependent on phase flow rates and mass transfer direction. An empirical correlation is derived to predict the Sauter mean drop diameter from a measurement of more than 20000 drops. The predictive correlation can reproduce the experimental data with very high accuracy.

## 6. Nomenclature

$A$	pulse amplitude [m]
$a_p$	packing surface area per unit volume of the column [ $\text{m}^2/\text{m}^3$ ]
$c_1, c_2, c_3$	constants of empirical correlation [-]
$d$	drop diameter [m]
$d_{32}$	Sauter mean drop diameter [m]
$f$	frequency [ $\text{s}^{-1}$ ]
$g$	acceleration due to gravity [ $\text{m}/\text{s}^2$ ]
$Q$	flow rate of the continuous or dispersed phase [ $\text{m}^3/\text{s}$ ]
$V$	superficial velocity [m/s]
$\sigma$	interfacial tension [N/m]
$\Delta\rho$	density difference between phases [ $\text{kg}/\text{m}^3$ ]
$\eta$	viscosity [Pa.s]
$\rho$	density [ $\text{kg}/\text{m}^3$ ]
$\phi$	holdup of dispersed phase [-]

## Subscripts

- c continuous phase  
d dispersed phase

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