Modelling of Mean Drop Size in a Extraction Spray Column and Developing a New Model

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ABSTRACT: *Drop size distribution plays a key role in the liquid extraction systems and related hydrodynamic and mass transfer parameters. In current research work, the size of drops in an extraction spray column has been measured by direct photography method, and the Sauter mean diameter has been obtained accordingly. Then, two types of models are assessed based on using dimensionless analysis and appropriate software. The interfacial tension in used chemical system is 1.8-32 dyne/cm. In first type of models, the correlation is established based on dimensionless parameters similar to approaches defined by other researchers. In second types, dimensional parameters have been used and therefore a new model is introduced for multi-drop extraction columns. The results show that the drop size diameter has the most affect on Eötvös number (Eo) in the first type of model and the correlation based on Eo number predicts drop size diameter with 8.1% error. In the second ones, four dimensional parameters (d_N, $\Delta\rho$, σ, u) have been selected as the most effective variables on drop size diameters and modelling results show prediction of drop size diameters with 5.82% error. In other cases, without using "u" as a variable, the amount of error has been decreased to 5.73% which shows better fitting.*

KEY WORDS: *Liquid extraction, Spray column, Drop diameter, Modeling.*

INTRODUCTION

In extraction processes, different methods and equipments are used such as mixer-settlers, packed columns, pulsed columns and spray columns to achieve appropriate separation performance. In all of these equipments, mass transfer is occurred by dispersing one phase into another one. Generally, among many types of extractors, extraction columns are widely used in industries, because of stable operation and high efficiency per stage. On the other hand, the extractor equipments without mechanical agitation, such as spray columns or packed columns, have high level performance despite small interfacial area between two phases because of large drop size. Extractor equipments with mechanical agitation, such as RDC columns, have high stage efficiency due to the large interfacial area, despite low level performance.

The stage efficiency depends on the interfacial area of dispersed phase, the mass transfer coefficients in the continuous phase and other properties of dispersed phase. The interfacial area increases with drop size decreasing and hold-up increasing related to dispersed phase.

Drop size has a key role on the design of liquid-liquid

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extraction columns. It affects the dispersed-phase hold-up, the residence time of the dispersed phase, and performance. Furthermore it affects on the mass transfer interfacial area and mass transfer coefficients of continuous and dispersed phases. It is therefore important to predict the drop diameter as a function of the column geometry, agitation conditions, physical properties of the liquid-liquid system, and direction of mass transfer.

Some investigators have developed mathematical relationships to describe the effects of column operation conditions on the drop size distribution. *Hayworth & Treybal* (1950) and *Null & Johnson* (1958) have presented semi-theoretical methods for estimating drop sizes in liquid-liquid systems. The *Hayworth & Treybal* correlation is derived from data with high interfacial tension and a maximum nozzle velocity of 0.33 ft/s. The *Null & Johnson* data includes relatively low interfacial tension systems and cover a range of nozzle flow rates in which uniform drop sizes are obtained. *Keith & Hixon* (1955) and *Christiansen & Hixson* (1957) have investigated the disintegration of organic liquid jets in water for the condition in which the interfacial area produced by the drops is at a maximum [2].

In recent years, several investigators have studied drop size diameter in extraction equipments. Some investigators have studied drop size in single nozzle distributors [3-6]. Other investigators have studied drop size in multi-nozzle distributors [7-12]. Some investigators have reviewed the three available methods for drop size distribution modeling including: the maximum entropy method, the discrete probability function method and the empirical method. The study of drop formation deals with the fundamental understanding of the behavior of liquid drops under the influence of various external body as well as surface forces [13-16].

In this paper, proposed models to predict drop size distribution have been considered basically with a focus on used variables in models. Then two types of models are established based on dimensionless and dimensional parameters and therefore two models are developed for multi-nozzle spray columns.

A fair comparison of present correlations with previously published equations is not always possible, since many of them only apply over limited ranges of variables. Furthermore, some of the earlier equations are implicit, and a solution for d_{32} is not always possible. However results show that models, developed in this paper, predict the mean drop size distribution with appropriate deviation of experimental data. Also present modelling has been verified with ten statistic tests and therefore it is more adaptable, compatible and reliable rather than other models obtained with other researchers.

Generally, the drops are generated in elliptical or spherical form in spray columns. Also, usually the form of small drops is spherical and with increasing the drop size, probably the form of drops will change to elliptical. In addition, drop inertial with drop size increasing will increased. The area of elliptical drops could be calculated with Eq. (1) [3].

$$A = \frac{\pi}{2}\left[d_H{}^2 + \frac{d_V\, d_H}{E^2 - 1}\ln\left(E + \sqrt{E^2 - 1}\right)\right] \quad (1)$$

Modified correlation for equal spherical drop area calculation (or equal diameter) could be calculated via Eq. (2) [3].

$$\frac{A}{A_e} = \frac{1}{2}\left[E^{2/3} + \frac{1}{E^{1/3}\sqrt{E^2 - 1}}\ln\left(E + \sqrt{E^2 - 1}\right)\right] \quad (2)$$

Current research work has been planned and implemented to develop models to predict mean drop size in extraction spray columns to achieve more accuracy.

Apparatus and experimental method

The spray column used in these experiments is made of glass in order to observe phases. The diameter and height of column is 30 cm and 200 cm respectively. A moving camera with macro lens has been adjusted through the column for photography of drops.

The volumetric flow rate of phases has been controlled with two digital measurement pumps. A drawing of spray column used in these experiments is shown in Fig. 2.

In order to start up the operations, at the beginning of each experiment, the column fills with continuous phase and then the pump turn on for phases. The drops have been produced in a perforated plate type distributor into continuous phase and collected at top of the column. Four different diameters of distributors (0.4, 0.8, 1, 1.3 mm) have been used in these experiments. Also three volumetric flow rates of dispersed phase have been used for each distributor. In these experiments, five chemical systems with different interfacial tensions have been used. The properties of chemical systems are shown

Table 1: Properties of used chemical systems.

	ρ_d (g/cm^3)	ρ_c (g/cm^3)	μ_d (cp)	μ_c (cp)	σ (dyne/cm)
Butanol-Water	0.848	0.988	1.020	1.378	1.80
Benzene-Water	0.873	0.994	0.591	0.916	25.7
Toluene-Water	0.864	0.997	0.612	0.986	30.0
Cumene-Water	0.863	0.992	0.772	0.928	32.5
Heptane-Water	0.681	0.994	0.387	0.921	36.2

Table 2: Sauter mean diameter for chemical systems.

Butanol-Water											
d_{32} (mm)	1.555	1.562	1.572	1.818	1.842	1.855	2.389	2.453	2.466	3.945	4.448	4.718
Benzene-Water												
d_{32} (mm)	3.011	3.546	3.671	3.951	4.111	4.156	4.251	5.231	5.554	6.376	6.877	7.003
Toluene-Water												
d_{32} (mm)	3.311	3.645	3.739	3.741	3.872	3.945	3.946	4.016	4.065	6.283	6.803	7.010
Cumene-Water												
d_{32} (mm)	3.764	3.785	3.808	4.484	4.605	4.971	5.264	5.448	5.626	8.773	8.905	8.948
Heptane-Water												
d_{32} (mm)	5.851	5.873	5.882	5.894	5.944	6.032	6.051	6.281	6.821	6.544	8.359	6.953

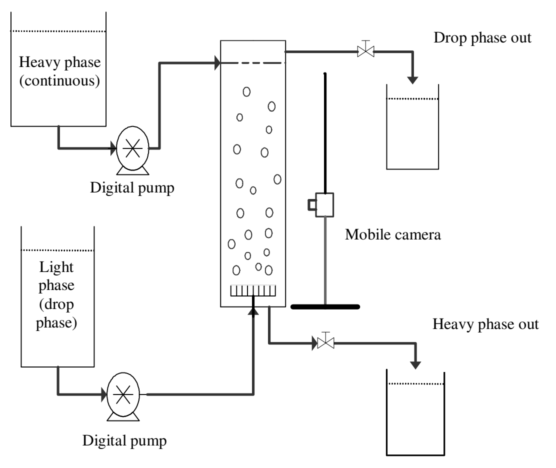

Fig. 2: Schematic of used spray column.

in Table 1. The temperature in all experiments is constant and equal to room temperature (25 °C).

In each experiment, the photography of drops is done through the column height. An 8 mega pixel camera (Sony-DSC-F828) has been used and numerous photos are taken from experimental system. Then photos are analyzed with AutoCAD software and Sauter mean diameters have been calculated in each series of photos.

The size of drops is analyzed with appropriate software and then the sauter mean diameter is measured with Eq. (3):

$$d_{32} = \frac{\sum_{i=1}^{N} n_i\, d_i^3}{\sum_{i=1}^{N} n_i\, d_i^2} \tag{3}$$

Totally, 60 experiments were done and almost over than 1000 drops were analyzed. The obtained results for d_{32} have been shown in Table 2.

A complete model not only must be able to predict drop sizes accurately, but also must cover all governing assumptions on modeling basis after passing appropriate statistical tests. To achieve above targets, "Eviews" software version 3.1 [17] has been used.

Generally, we proposed two types of models. First types of models are established based on dimensionless parameters such as We, Eo and Re. But in second types of models, we used the results of first types of models and proposed a new model based on the most effective variables. Of course, in respect of statistics, all models are optimized and several tests used for statistics accuracy such as R^2, R^2_{adj}, t-statistics, F-statistics and D.W.T(1).

Generally the drop diameter depends on interfacial tension, nozzle velocity, density and viscosity of two phases, nozzle diameter and gravity acceleration. Therefore, we can write a general correlation as:

$$f\,(d_{32,}\,\sigma, u, \rho_c, \rho_d, \Delta\rho, \mu_c, \mu_d, d_N, g) = 0 \quad (4)$$

Based on correlation 2 and pai theorem, we can defined seven dimensionless groups and we can write a correlation as below based on dimensional analysis:

$$\frac{d_{32}}{d_N} = k\,(Eo)^A\,(We)^B\,(Re)^C\,\left(\frac{\Delta\rho}{\rho_d}\right)^D \left(\frac{\Delta\rho}{\rho_c}\right)^E \left(\frac{\mu_d}{\mu_c}\right)^F \quad (5)$$

The constants k, A, B, C, D, E and F can be evaluated by several different methods. In general, this model has strong statistics basis and it can predict of d_{32} well [9,10]. Usually, most of models consist of Eo and We. They can predict drop diameter with a low-error [8-12].

The constants are measured with regression analysis by use of Ordinary Least Square (OLS) method. In OLS method, the actual purpose is minimizing the sum of square made by differences between model and actual values. Whereas the "Eviews" software has high ability in OLS method, the analysis of models are done by this software.

In according to the method and software mentioned above, it is necessary to define some parameters, which help in result analysis, including R^2, R^2_{adj}, D.W.T, Prob(t-statistic) and Prob (F-statistic). Not only these parameters are important and effective statistic parameters in modeling, but also are defined well in "Eviews" software.

R^2 and R^2_{adj} are the most important parameters to determine the model ability in fitting the variations of the models provided on the basis of experimental data. In general, the amount of these parameters is between 0.0 and 1.0 and the level of fitting increases with growing the parameters to 1.0. It must be considered that increasing the number of points will hold the R^2 untruly but R^2_{adj} solves this problem. Thus, in the cases with enormous experimental data, R^2_{adj} is considered in analysis.

(1) Durbin-Watson Test

Table 3: Experimental ranges studied

	From	To
Eo	0.07	1.0
We	0.08	1.5
Re	2.0	35.0

D.W.T is another parameter considering the difference between real and model amount in every point knowing as residual. In fact, this parameter assesses the relation among residual data. Relation between real and model amounts obviously decreases the validity of prepared equation in modeling. In a good model, the amount of D.W.T must be 1.0 at least, but more than 1.7 will made an appropriate model.

Prob(t-statistic) and Prob(F-statistic) are the statistical parameters to confirm the model of used independent variables and the whole format of equation respectively. In "Eviews" software, the amounts more than 0.05 for these parameters could be the results of using unnecessary variables in modelling or inaccurate format of the equation.

MODELLING RESULTS

In this paper, different types of models have been developed on the basis of related independent variables and five mentioned statistical tests, have been implemented in each ones. The experimental ranges studied are presented in Table 3.

The results of the first type models have been shown in table 4 and their assessment is as following:

- In according to the amounts of R^2, R^2_{adj} and D.W.T, it could be observed that the We and Re numbers aren't appropriate variable in this modeling. In addition, experimental results show an error more than 30% in these models. Thus, the models in the rows 2, 3 and 5 could not be proposed for modeling.
- In general, the amounts of Prob(t-statistic) show the importance level of the variables in modeling. As shown in table 4, the amounts of this parameter about We and Re for the models in the rows 2, 3 and 5 show that these

Table 4: Prepared models on the basis of dimensionless variable and the results of statistical tests.

	Model	R^2	R^2_{adj}	D.W.T	Probability					%E.
	$\frac{d_{32}}{d_N} = C_1\,(Eo)^{C_2}\,(We)^{C_3}\,(Re)^{C_4}$				t- stat				F-Stat.	
					C_1	C_2	C_3	٤		
1	$\frac{d_{32}}{d_N} = 1.913Eo^{-0.2665}$	0.94	0.94	1.137	0.00	0.00	---	---	0.00	8.10
2	$\frac{d_{32}}{d_N} = 3.690\,We^{0.0922}$	0.06	0.04	0.090	0.00	---	0.07	---	0.07	36.21
3	$\frac{d_{32}}{d_N} = 2.586Re^{0.0572}$	0.04	0.02	0.066	0.00	---	---	0.14	0.14	32.09
4	$\frac{d_{32}}{d_N} = 1.911\,Eo^{-0.2665}\,Re^{0.0004}$	0.94	0.93	1.140	0.00	0.00	---	0.97	0.00	8.10
5	$\frac{d_{32}}{d_N} = 3.220\,We^{0.0737}\,Re^{0.0338}$	0.07	0.03	0.102	0.00	---	0.18	0.42	0.14	30.59
6	$\frac{d_{32}}{d_N} = 2.053\,Eo^{-0.2628}\,We^{0.0312}$	0.94	0.94	1.224	0.00	0.02	0.00	---	0.00	7.92
7	$\frac{d_{32}}{d_N} = 2.137\,Eo^{-0.2642}\,We^{0.0368}\,Re^{-0.0107}$	0.94	0.94	1.175	0.00	0.00	0.00	0.31	0.00	7.82

dimensionless parameter are not appropriate and using them in modeling causes problems in the fitting.

- As mentioned above, the amount of Prob(F-statistic) analyses the validity of the model including all of the variables. Thus the results observed in Table 4 show that the equations including Re number are not appropriate and these dimensionless number is not useful in modeling of mean drops diameter in spray columns. Also the amount of Prob(F-statistic) shows that We number is not useful lonely.

In concluding, the models in the row 2, 3, 4, 5 and 7 of Table 4 are inappropriate for mean drop size distribution modelling and the models in the rows 1 and 6 are proposed providing almost equal acceptable error. But, the complexity of a model is a criterion to select the model, thus the model in the row 6 is the best model in this field, providing required accuracy in mean drop size modelling in spray columns.

Another important result in table 4 is the negligible effect of adding dimensionless variables in modelling, decreasing error less than 0.5%.

Thus, the variable priority to use in modelling is as following:

1. Eo
2. We
3. Re

The research in this paper continued based on the results of table 4, distinguishing the effective parameters to make model. As mentioned above, Eo and We are more effective in modelling, including main variables $\sigma, \Delta\rho, d_N$ and u. Thus, a new model is defined as following:

$$d_{32} = C_1\, d_N^{C_2}\,\sigma^{C_3}\,(\Delta\rho)^{C_4}\,u^{C_5} \qquad (6)$$

In according to the results of statistical analysis, it is better to justify C_1 coefficient equal to one, to obtain appropriate accuracy in modelling.

In opposite of the first type models, the second type models are not including dimensionless variables. The output of the “Eviews” software based on new model (Eq. 6) is shown in Table 5. In this table, all of the main variables are used in modelling. Analysing the variable trends, leads to omit u from model and this is the cause of decreasing error level to 5.73, more acceptable and appropriate than similar models, provided with other researcher. The output of the “Eviews” software based on new model with omission of u is shown in Table 6. On the other hand, this form of models, decreases error strongly almost over 30%, in relation to previous models which were based on dimensionless variables. The results of this research paper and other researcher activities are compared in Table 7.

Table 5: The output of the "Eviews" software.

Model: $d_{32} = d_N^{0.496302}\, \sigma^{0.221950}\, (\Delta\rho)^{-0.227903}\, u^{0.064360}$				
%Error: 5.8161				
LOG(D32)=C(2)*LOG(DN)+C(3)*LOG(SIG)+C(4)*LOG(DRO)+C(5)				
	Coefficient	Std. Error	t-Statistic	Prob.
C(2)	0.496302	0.022138	22.41868	0.0000
C(3)	0.221950	0.018452	12.02867	0.0000
C(4)	-0.227903	0.028151	-8.095685	0.0000
C(5)	0.064360	0.024361	2.641928	0.0108
R-squared	0.959557	Mean dependent var		-5.656403
Adjusted R-squared	0.957310	S.D. dependent var		0.566970
S.E. of regression	0.117145	Akaile info critenrion		-1.384344
Sum squared resid	0.741034	Schwarz criterion		-1.242244
Log likelihood	44.14597	F-statistic		427.0714
Durbin-watson stat	1.410262	Prob(F-statistic)		0.000000

Table 6: The output of the "Eviews" software.

Model: $d_{32} = d_N^{0.462849}\, \sigma^{0.258836}\, (\Delta\rho)^{-0.270592}$				
%Error: 5.7290				
LOG(D32)=C(2)*LOG(DN)+C(3)*LOG(SIG)+C(4)*LOG(DRO)				
	Coefficient	Std. Error	t-Statistic	Prob.
C(2)	0.462849	0.019121	24.20644	0.0000
C(3)	0.258836	0.012703	20.37651	0.0000
C(4)	-0.270592	0.024273	-11.14788	0.0000
R-squared	0.954	Mean dependent var		-5.656403
Adjusted R-squared	0.952669	S.D. dependent var		0.566970
S.E. of regression	0.123348	Akaile info critenrion		-1.297268
Sum squared resid	0.83617	Schwarz criterion		-1.190694
Log likelihood	40.62078	F-statistic		574.6404
Durbin-watson stat	1.341188	Prob(F-statistic)		0.000000

CONCLUSIONS

In according to the obtained results, we can conclude that for first type models:

- The simple and appropriate model to predict the mean drops diameter in a spray column with dimensionless variables is based on the Eo number with 8.1% error. In addition, using this model in the high velocity systems could be strongly proposed.
- The priority of dimensionless parameters for modelling in this field is Eo, We and Re respectively.

For second type models:

- The best and appropriate model in this case is an equation including $\sigma, \Delta\rho, d_N$ and u as independent variables. However, if the data for u is not reliable, the model shown in table 6 (the model without u) could be proposed.
- The most effective variable for modelling the mean drops diameter in a spray column is d_N. Furthermore, the results show that the priority of dimensional parameters for modelling in this case is d_N, σ, $\Delta\rho$ and u respectively.

Table 7: Selected correlations for drop size distribution in the spray columns.

No.	Model		% Error
1	$\frac{d_{32}}{d_N} = 1.913037\,Eo^{-0.266549}$		8.10
2	$\frac{d_{32}}{d_N} = 2.05306336\,Eo^{-0.262783}\,We^{0.031179}$		7.92
3	$d_{32} = d_N^{0.496302}\,\sigma^{0.221950}\,(\Delta\rho)^{-0.227903}\,u^{0.064360}$		5.82
4	$d_{32} = d_N^{0.462849}\sigma^{0.258836}\,(\Delta\rho)^{-0.270592}$		5.73
5	$d_{32} = 1.15c_1\left[\frac{\sigma}{\Delta\rho g}\right]^{0.5}$	Seibert and Fair [12]	34.57
6	$d_{32} = 2.07\left[1-0.193Eo\right]$, $0.011 < Eo < 1.70$	Perrut and Loutaty [7]	Out of range (Refused)
7	$d_{32} = 1.592\left[\frac{u^2}{2gd_N}\right]^{-0.0665}\left[\frac{\Delta\rho d_N}{\sigma}\right]^{-0.5}$	Vedaiyan et al. [10]	Out of range (Refused)
8	$d_{32} = 1.59d_N\,We^{-0.068}\,Eo^{-0.278}$, $0 < We < 2$	Kumar and Hartland [8]	11.87
9	$d_{32} = 1.909d_N\,We^{-0.0028}\,Eo^{-0.1457}$, $0.005 < We < 2$	Chun and Wilkinson [9]	17.85

- In according to the error results, the best and appropriate model is an equation based on the d_N, σ and $\Delta\rho$ with 5.73% error (according to table 6).
- Based on the results of statistical analysis, it is proposed to justify C_1 coefficient in Eq. 4 equal to one, to obtain appropriate accuracy in modelling of these models.

Nomenclature

c_1	Drop size correction factor in Siebert's correlation (=1)
d_{32}	Sauter mean drop diameter, mm
d	Drop diameter, mm
d_i	i th drop, mm
E	Drop Inertia
g	Gravity, m/s^2
n	Number of drops in the sample
u	Nozzle velocity, mm/s

Greek Symbols

$\Delta\rho$	Density difference between phases, g/cm^3
μ_c	Continuous phase viscosity, cp
μ_d	Dispersed phase viscosity, cp
ρ_c	Continuous phase density, g/cm^3
ρ_d	Dispersed phase density, g/cm^3
σ	Interfacial tension, dyne/cm

Dimensionless Numbers

We	"Weber" dimensionless number, $\Delta\rho\, d_N\, u^2/\sigma$
Re	"Reynolds" dimensionless number, $\Delta\rho\, u\, d_N/\mu$
Eo	"Eotvos" dimensionless number, $\Delta\rho\, d_N^2\, g/\sigma$

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