An Inverse Problem Method for RDC Simulation

Hanin B. Jildeh[#1], Menwer Attarakih[*2], Hans-Jörg Bart[#3]

[#] *Chair of Separation Science and Technology, TU Kaiserslautern, Postfach 3049 - 67653 Kaiserslautern, Germany*

[*] *Faculty of Engineering Technology, Chemical Engineering Department, Al-Balqa Applied University, Postfach 15008 - 11134 Amman, Jordan*

[1]hanin.jildeh@mv.uni-kl.de

[2]attarakih@yahoo.com

[3]bart@mv.uni-kl.de

***Abstract*— The hydrodynamic and mass transfer behavior of a Rotating Disk Contactor (RDC) extraction column is investigated for two different liquid-liquid systems recommended by the European Federation of Chemical Engineering (EFCE). An inverse problems method is applied to estimate the coalescence parameters in a RDC extraction column. Single-droplet studies in a small lab scale RDC is used to evaluate the coalescence parameters necessary for column simulations, which were obtained by an inverse solution of the population balance model using the generalized fixed-pivot technique for the discretization of the droplet internal coordinate. The coalescence parameters resulted from solving the inverse problem are dependent on the chemical test system used for the simulation. Then the resulted coalescence parameters values were used in LLECMOD to study the hydrodynamic and mass transfer behavior of pilot plant of RDC extraction column. The simulated Sauter mean droplet diameter, hold-up values and concentration profiles for organic and aqueous phase were found to be well predicted compared to the experimental data.**

I. Introduction

Liquid-liquid extraction is a powerful separation technique after distillation and it is widely used in many industrial fields such as petrochemical and biochemical industries. Liquid-liquid extraction columns are classified into three main categories: stirred columns (Rotating Disc Contactor (RDC) and Kühni columns), non-agitated columns and pulsed columns (sieve plate and packed columns).

Liquid–liquid extraction column (LLEC) simulation based on population balance modeling has difficulties due to contiguous change in the droplet properties such as Sauter mean droplet diameter, hold-up values and concentration profiles for organic and aqueous phase. The most common mechanisms effects on the population balance model are breakage, coalescence and mass transfer [1].

Inverse problem is an ill-posed in general and need some stabilization techniques to get reliable optimized parameters. Solving it is a necessary part for reliable modelling strategy. However, this is complicated due to the increase in the size of the differential-algebraic system when dealing with inverse problems using population balance models.

The objective of this paper is to evaluate the coalescence parameter in a small lab scale RDC for two EFCE systems as is butylacetate-water and toluene-water. We then use these estimated constants to predict the Sauter mean droplet diameter, holdup and concentration profiles for a pilot plant scale column.

II. The Mathematical Model

The general spatially distributed population balance equation (SDPBE) for describing the coupled hydrodynamics and mass transfer in liquid-liquid extraction columns (LLECs) in a one spatial domain could be written as [2]:

$$\frac{\partial f_{d,c_y}(\psi)}{\partial t}+\frac{\partial\left[u_y f_{d,c_y}(\psi)\right]}{\partial z}+\frac{\partial\left[\dot{c}_y f_{d,c_y}(\psi)\right]}{\partial c_y}=$$
$$\frac{\partial}{\partial z}\left[D_y\frac{f_{d,c_y}(\psi)}{\partial z}\right]+\frac{Q_y^{in}}{A_c}f_y^{in}(d,c_y;t)\delta(z-z_y)+\Upsilon\{\psi\}$$

In this equation the components of the vector $\psi = [d\ c_y\ z\ t]$ are those for the droplet internal coordinates (diameter and solute concentration), the external coordinate is (z) and for time is (t). The velocity vector along the internal coordinates is given by (c_y) is ($\dot{c_y}$) and (Υ) represent the source term. (ζ) represent the net number of droplets produced by breakage and coalescence per unit volume and unit time in the coordinates range [ζ, ζ+$\partial\zeta$]. The dispersed phase velocity (u_y) relative to the walls of the column is determined in terms of the slip velocity (u_s) with respect to the continuous phase velocity (u_x) with respect to the walls of the column [3], [4].

The source term is given by the following equation:

$$\underbrace{\Upsilon\{\psi\}}_{Source\ Term}=\underbrace{B^b(d,c_y;t,z)}_{Birth\ by\ Breakage}-\underbrace{D^b(d,c_y;t,z)}_{Death\ by\ Breakage}$$
$$+\underbrace{B^c(d,c_y;t,z)}_{Birth\ by\ Coalescence}-\underbrace{D^c(d,c_y;t,z)}_{Death\ by\ Coalescence}$$

The bivariate source term for the four rates of droplets birth and death due to droplet breakage and coalescence are quite complicated they are expressed in details [2].

Simulating liquid-liquid extraction columns is a challenging task due to the discrete character of the dispersed phase. In technical geometries the population balance model has no general analytical solution and hence a special numerical technique is required to solve the PBE [4]. Several numerical approach are proposed to solve the PBE which are

classified into two categories namely classes method Kumar and Ramkrishna [5] and method of moments [6]. Attarakih has invented a new technique for the solution of the population balance equation using the sectional quadrature method of moments (SQMOM) which combines the advantages of classes method and method of moments [7].

III. Inverse Problem Approach

Inverse problems for population balances have attracted many researchers in the recent years [3], [8]–[10]. Inverse problem is considered an ill-posed in general and need some stabilization techniques to get reliable optimized parameters. In more general cases a mathematical programming procedure has to be applied in order to find the best fit of the experimental data. Since the inverse problems are highly sensitive to errors in the experimental data, the experimental data has been fitted to a normal distribution to give a smother curve and remove some noise, by regularization.

By solving the population balance equations the optimization algorithm derives the unknown coalescence parameters by fitting the simulated outlet distribution to the experimental one using an objective function.

In this work an example of the inverse problem program has been used to obtain the coalescence parameters of a lab scale RDC for two cases of two EFCE systems as is butylacetate-water and toluene-water.

IV. Estimation Parameters

Despite the rich literature on population balance equation modeling, there are few studies on optimization packages for coalescence models that helps to choose the best coalescence model for the experimental data. These optimization packages must fully describe the system by solving the population balance problem inversely to get the best values of constants in the coalescence model. In addition the objective function should be wisely chosen to minimize the error and to have a unique solution for a given data. Since droplet coalescence is highly sensitive to the hydrodynamics and physic-chemical properties, interfacial dynamics and mass transfer [3].

Coulaloglou & Tavlarides (1977) developed a model for stirred vessel, which is based on the kinetic theory of gases and drainage film theory; and calculate the coalescence frequency from the collision rate (frequency) (h) and the coalescence efficiency (λ) [11].

$$\omega(d,d',\phi_d) = h(d,d',\phi_d)\ \ \lambda(d,d',\phi_d)$$

The Coulaloglou & Tavlarides coalescence model is given by the following equation [12]:

$$\omega(d,d',\phi_d) = \left[c_1 \frac{\varepsilon^{1/3}}{1+\phi} (d+d')^2 (d^{2/3}+d'^{2/3})^{1/2} \right] \times \left[\exp\left(-\frac{c_2 \eta_x \rho_x \varepsilon}{\sigma^2 (1+\phi)^3} \right) \left(\frac{(dd')}{(d+d')} \right)^4 \right]$$

An alternative to this model is developed by Sovova (1981) introducing a different formula for the efficiency of collisions, the mechanism of coalescence of drops by film drainage is replaced by a mechanism based on the effect of collision impact [13]:

$$\omega(d,d',\phi_d) = \left[c_3 \frac{\varepsilon^{1/3}}{1+\phi} (d+d')^2 (d^{2/3}+d'^{2/3})^{1/2} \right] \times \left[\exp\left(-\frac{c_4 \eta_x \rho_x \varepsilon}{\sigma^2 (1+\phi)^3} \right) \left(\frac{(d^2+d'^2)\times(d^3+d'^3)}{(dd')\times(d^{2/3}+d'^{2/3})} \right) \right]$$

The coalescence frequency model proposed by Laso (1986) is expressed by the following equation [14]:

$$\omega(d,d',\phi_d) = c_7 \left(\frac{\pi d^3}{6 D_R^3} \right)^{-0.49} We^{-0.51} \phi_d^{\,0.9} Oh^{-0.05}$$

A semi-empirical model proposed by Casamatta and Vogelpohl in (1985) in which the first term at the right hand of the equation is the coalescence frequency caused by random motion, which is assumed to be proportional to the drop volumes. The second term represents the coalescence induced by different rise velocities are expressed by the following equation [11], [15]:

$$\omega(d,d',\phi_d) = \left[c_5 \frac{\pi^2}{36} (d \times d')^3 \right] + \left[c_6 \frac{\pi^2}{6} \left(\frac{d+d'}{2} \right)^2 \left| d^3 - d'^3 \right| \right]$$

The inverse problem program can estimate the constants $c_1 \ldots\ldots c_7$ of the previous coalescence models along with the ability to choose one of the breakage models available to run the simulation or with no breakage mode. Additional results will be discussed in further publications.

V. LLECMOD Program

The mathematical model described above was programmed using Visual Digital FORTRAN and to facilitate the data input and output a LLECMOD program was designed. A graphical user interface of LLECMOD program (Liquid-Liquid Extraction Column MODule) simulates liquid-liquid extraction columns based on the population balance model to predict the column performance at steady state and transient conditions. Furthermore the design of LLECMOD is flexible in such a way it enables the user to define the input operating condition, the coalescence and breakage parameters and other different options according to the simulated case studied. Also LLECMOD enables to input the droplet terminal velocity, energy dissipation, axial dispersion, breakage and coalescence frequencies and the other internal geometrical details of the column.

LLECMOD is considered a user-friendly and flexible windows-based program that contains the main input window and other sub-windows for parameters and correlations input. Using LLECMOD simulations can now be carried out successfully for different types of extraction columns

including agitated (Rotating Disc Contactor (RDC) and Kühni columns) and non-agitated or pulsed columns (sieve plate and packed columns) with different internal geometry.

VI. Results and Discussion

In this study we simulate two cases of two phase liquid-liquid system for butylacetate-water (B-W) and toluene-water (T-W) in a lab scale RDC [16]. The RDC extraction column has five compartments, its internal diameter is 150 mm, column height 150 mm, compartment height is 30 mm, internal stator diameter is 105 mm, rotator diameter 90 mm and diameter of the rotating shaft is 54 mm. The angular velocity of the shafts is constant 300 rpm and volumetric flow rate for continuous and dispersed phase is $Q_c = Q_d = 100$ l/hr for both systems.

The values of coalescence parameters for Coulaloglou and Tavlarides coalescence model (c_1 and c_2) were estimated using the inverse problem for both systems shown in Table 1.

TABLE I
Coalescence Parameters for Coulaloglou and Tavlarides

Chemical System	c_1 (-)	c_2 (m^{-2})	Objective Function
B-W	8.19×10^{-02}	$2.37\times10^{+11}$	1.76×10^{-04}
T-W	2.17×10^{-02}	$1.19\times10^{+11}$	1.69×10^{-04}

The inlet and outlet cumulative volume distribution are used from the work of Simon [16]. The unknown coalescence parameters values were obtained by minimizing the square sum of errors according to the following equation:

$$Objective\ Function = \sum_{k=1}^{N} (Q_{3,k}^{sim} - Q_{3,k}^{exp})$$

where; Q3 is the cumulative volume distribution.

$$Q_3^{sim} = \int_0^{d_{max}} q_3(d)\, \partial d$$

where; d: droplet diameter [mm]. Fig. 1 shows the resulted curve form simulating the cumulative volume distribution at the outlet to the experimental data, where a very good agreement is achieved.

Then the estimated constants were used to simulate a pilot plant RDC extraction column at steady state using the LLECMOD program; for the column height 4.4 m, column diameter 0.08 m, inlet of the dispersed and continuous phase are 0.85 m and 3.8 m, respectively. The two EFCE test systems butylacetate-acetone-water (B-A-W) and toluene-acetone-water (T-A-W) are used with inlet feed that is normally distributed with mean value equal to 2.1 mm and standard deviation of 0.6 mm.

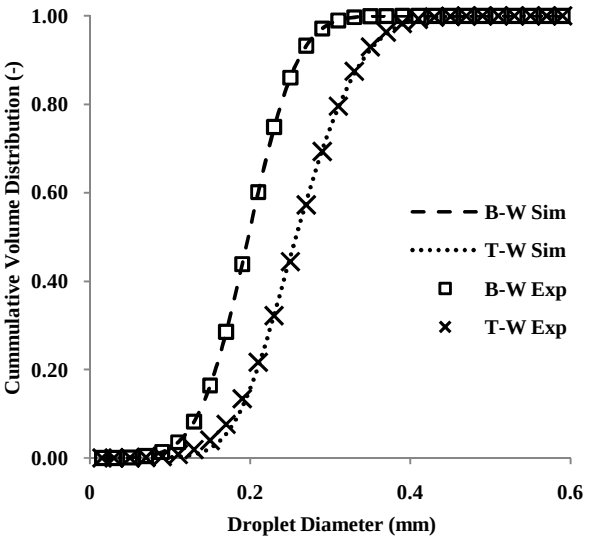

Fig. 1 Simulated and experimental cumulative volume distribution for butylacetate-water (B-W) and toluene-water (T-W) in a lab scale RDC

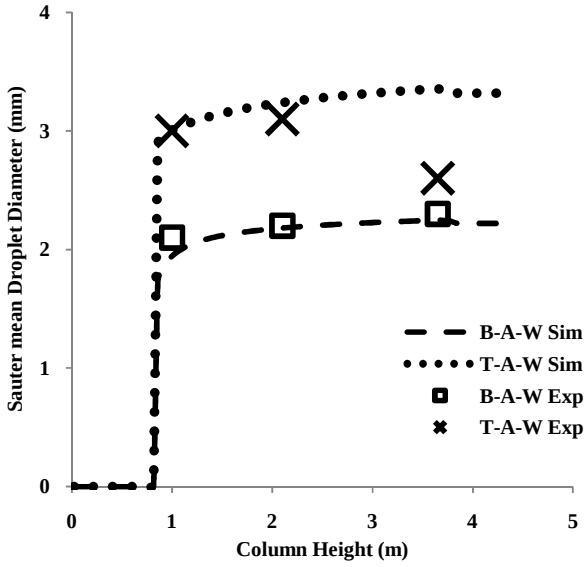

Fig. 2 Simulated Sauter mean droplet diameter along the column height compared to the experimental data [17] for two EFCE test system B-A-W & T-A-W

The pilot plant RDC extraction column geometric internal data: its compartment height is 50 mm, internal stator diameter is 50 mm, rotator diameter 45 mm and diameter of the rotating shaft is 10 mm. The angular velocity of the shafts is constant 200 rpm and volumetric flow rate for continuous and dispersed phase is 40 l/hr and 48 l/hr, respectively for both systems. The direction of mass transfer is from the continuous to the dispersed phase.

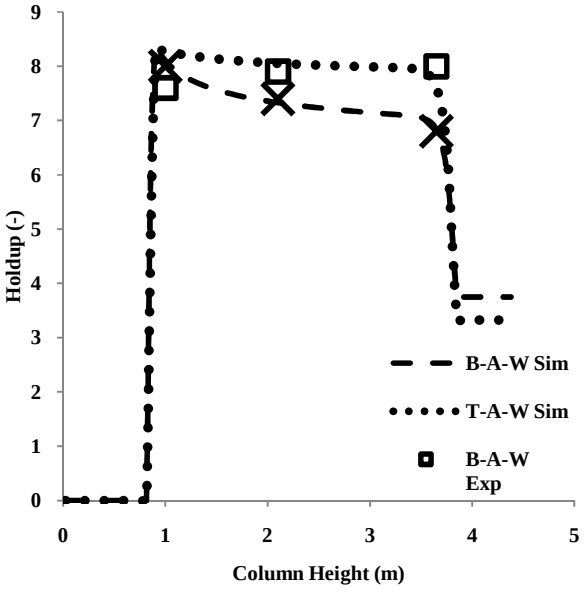

Fig. 3 Simulated holdup along the column height compared to the experimental data [17] for two EFCE test system B-A-W & T-A-W

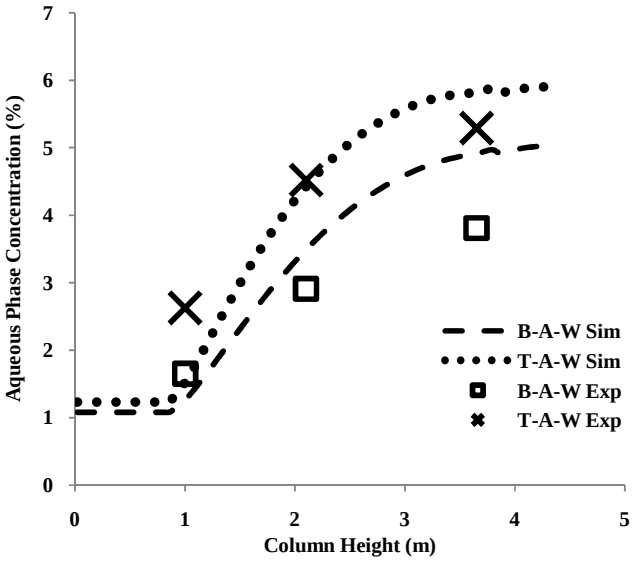

Fig. 4 Simulated aqueous solute concentration profile along the column height compared to the experimental data [17] for two EFCE test system B-A-W & T-A-W

A comparison between the simulated Sauter mean droplet diameter along the column height and the experimental data taken from the work of Garthe (2006) [17] is shown in Fig. 2 where a good agreement is achieved for both tested chemical systems.

Fig. 3 shows the simulated holdup along the column height compared to the experimental data for both chemical systems; where a very good agreement is achieved for both tested systems.

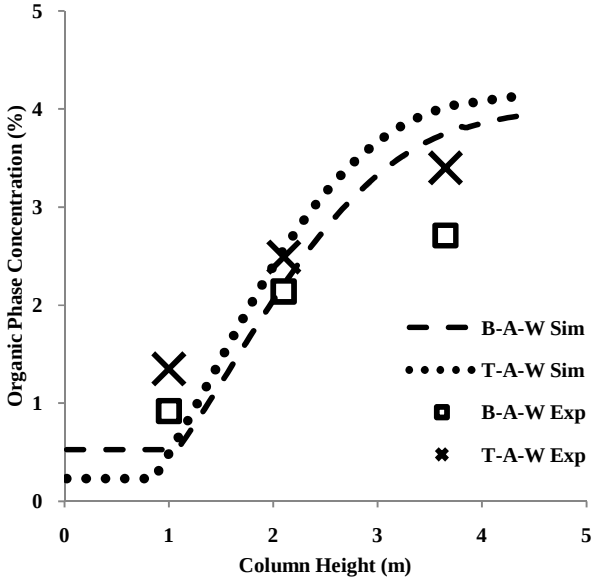

Fig. 5 Simulated organic solute concentration profile along the column height compared to the experimental data [17] for two EFCE test system B-A-W & T-A-W

Fig. 4 & 5 shows the simulated curve and experimental data for solute concentration profiles for aqueous and organic phase respectively as a function of column height, the agreement between the simulation and experiment is good for both test systems.

Results shown in Fig. 2 to 4; prove that the obtained values from the solution of the inverse problem gave a very good agreement to express the experimental data used from a pilot RDC plant in a simulation when inserting the values of Table 1 into the LLECMOD program.

VII. Conclusion

In this paper the inverse problem method was used for parameter estimation for different droplet coalescence models for lab scale RDC extraction column. The resulted parameters derived are valid for pilot plant extraction column but it is dependent on the chemical system used. The user friendly windows-based program LLECMOD was used for the hydrodynamics and mass transfer simulation of RDC extraction columns for steady state simulation of pilot plant RDC using the derived values of coalescence parameters resulted from the inverse problem program. The resulted simulation for the Sauter mean droplet diameter, hold-up and concentration organic and aqueous phase profiles showed a good agreement to the experimental data.

Acknowledgment

The authors wish to thank the State Research Centre of Mathematical and Computational Modelling.

NOMENCLATURE

A_c: column cross-sectional area [m^2]
C: solute concentration [kg/m^3]
B, D: birth and death source terms [$1/m^3/s$]
c_1……c_7: coalescence parameters
d: droplet diameter [m]
d′: mother droplet diameter [m]
D_R: column diameter[m]
f: Flux vector
Oh: Ohnesorge no. =$Oh = \frac{\eta_d}{(\sigma d \rho_d)^{0.5}}$
Q: volumetric flow rate [m^3/s]
Q_3: cumulative volume distribution
u: velocity [m/s]
t: time [s]
We: Weber no. =we $= \frac{\rho \varepsilon^{2/3} d^{5/3}}{\sigma}$
z: space coordinate [m]

GREEK SYMBOLS

ε: energy dissipation [m^2/sec^3]
η: dynamic viscosity [kg/m sec]
λ: coalescence efficiency
ρ: Density [kg/m^3]
σ: surface tension [N/m]
ϒ: source term that represents the net number of droplet produced by breakage and coalescence [1/s]
φ: volume fraction
φ_d: hold up
ψ: internal and external coordinates vector ([d c_y z t])
ω: coalescence rate [m^3/sec]

SUBSCRIPTS

x, y: continuous and dispersed phases respectively
s: slip velocity

SUPERSCRIPTS

●: derivative with respect to time
B: breakage
C: coalescence
exp: experimental
in: inlet
max: maximum
out: outlet
sim: simulated

REFERENCES

[1] J. Kumar, G. Warnecke, M. Peglow and S. Heinrich," Comparison of numerical methods for solving population balance equations incorporating aggregation and breakage," Powder Technology, vol. 189, issue 2, pp. 218-229, 2009.

[2] M. M. Attarakih, H.-J Bart, T. Steinmetz, M. Dietzen and N. M. Faqir, "LLECMOD: A Bivariate Population Balance Simulation Tool for Liquid- Liquid Extraction Columns," Open Chemical Engineering Journal, vol. 2, pp. 10-34, 2008.

[3] M. M. Attarakih H.-J Bart. L. Lagar G., and N. M. Faqir, "LLECMOD: A Window-based program for hydrodynamics simulation of liquid-liquid extraction columns," Chemical Engineering Processing, vol. 45, pp. 113-123, 2006.

[4] M. M. Attarakih H.-J Bart. and N. M. Faqir, "Numerical Solution of the Bivariate Population Balanced Equation for the Interacting Hydrodynamics and Mass Transfer in Liquid-Liquid Extraction Columns," Chemical Engineering Science, vol. 61, pp. 113-123, 2006.

[5] S. Kumar and D. Ramkrishna, "On the solution of population balance equations by discretization—II. A moving pivot technique," Chemical Engineering Science, vol. 51, issue 8, pp. 1333–1342, 1996.

[6] R. McGraw, "Description of aerosol dynamics by the quadrature method of moments," Aerosol Science and Technology, vol. 27, pp. 255–265, 1997.

[7] M. M. Attarakih H.-J Bart and N. M. Faqir, "Solution of the population balance equation using the sectional quadrature method of moments (SQMOM)," Computer Aided Chemical Engineering, vol. 21, pp. 209-214, 2009.

[8] A. M. O'Rourke and P.F. MacLoughlin, "A study of drop breakage in lean dispersions using the inverse-problem method," Chemical Engineering Science, vol. 65, issue 11, pp. 3681-3694, 2010

[9] A. Vikhansky, M. Kraft, M. Simon, S. Schmidt and H.-J. Bart, "Droplets Population Balance in a Rotating Disc Contactor: An Inverse Problem Approach," AIChE Journal, vol. 52, issue 4, pp. 1441- 1450, 2006.

[10] A. Braumann, P. L. W. Man and M. Kraft, "Statistical approximation of the inverse problem in multivariate population balance modeling," pp. 1473 – 4273, August 2009.

[11] Y. Liao and D. Lucas, "A literature review on mechanisms and models for the coalescence process of fluid particles," Chemical Engineering Science, vol. 65, issue 10, pp. 2851-2864, 2010.

[12] C. A. Coulaloglou and L. L. Tavlarides, "Description of interaction processes in agitated liquid–liquid dispersions," Chemical Engineering Science, vol. 32, pp. 1289–1297, 1977. H.

[13] Sovova, "Breakage and coalescence of drops in a batch stirred vessel-II Comparison of model and experiments," Chemical Engineering Science, vol. 36, pp. 1567–1573, 1981.

[14] T. Steinmetz, "Tropfenpopulationsbilanzgestütztes Auslegungs-verfahren zur Skalierung einer gerührten Miniplant Extraktions-kolonne," Forchr.-Ber. VDI, Reihe 3, Nr. 885, Düsseldorf: VDI Verlag 2007.

[15] G. Casamatta and A. Vogelpohl, "Modelling of fluid dynamics and mass transfer in extraction columns," German Chemical Engineering, vol. 8, pp. 96-103, 1985.

[16] M. Simon, "Koaleszenz von Tropfen und Tropfenschwärmen," Dissertation, Technischen Universität Kaiserslautern, Germany, April 2004.

[17] D. Garthe, "Fluid Dynamics and Mass Transfer of Single Particles and Swarms of Particles in Extraction Column," Dissertation, Technischen Universität München, Germany, April 2006.