

Fluid flow-mass transfer interaction in perforated plate extractive reactors

A. O. Ghallab, A. K. Abo El Ata, R. S. Ettouney, and M. A. El-Rifai

Chemical Engineering Department, Faculty of Engineering, Cairo University,
Giza, Egypt

Abstract

A steady state model is developed for un-agitated perforated plate liquid-liquid counter current extraction columns conducting a very fast chemical reaction taking place at the interface between a solute in the raffinate phase and an excess of reactant in the extract phase. The model parameters reflect the interaction between mass transfer and hydrodynamic flow pattern and depend on the transport properties of the species involved, the relative phase mass velocities, and the available interfacial area. Transport property and fluid mechanics correlations are integrated into the mass transfer performance computations. Results of application of the developed models demonstrate vividly the effect of phase flow rates on the hydrodynamic and mass transfer parameters and on the separation obtainable in perforated plate extractive reactors.

1. Introduction

Extraction intensified by a chemical reaction is resorted to in applications covering the petroleum, petrochemicals, nuclear, and pharmaceutical industries [Ramkrishna, 2000; Bart et al., 2008]. It is used not only for facilitating the removal of an unwanted component from a liquid but also for achieving better selectivity or synthesis of a desired product [Bart, 2001]. Rigorous modeling of the performance of such columns has to consider the rate and stoichiometry of the enhancing reaction and the physical properties of the handled systems which affect the flow conditions and interfacial area of the contacted phases and those affecting the internal transport

within the phases. The performance of heterogeneous fluid-fluid reactors depends on the relative values of the mass transfer and reaction resistances [Danckwerts, 1970]. When both the Hatta modulus [van Krevelen and Hoftyzer, 1953] and rate constant are sufficiently high, the reaction takes place at the interface between the two phases. For small values of the Hatta modulus, the reaction is slow with respect to mass transfer and the reaction takes place in the bulk of the extract phase.

A wide variety of contacting equipment which differ considerably in the obtained fluid-fluid interaction flow pattern are in commercial use. Previous modeling studies considered non-ideal flow in discrete contacting and separation in mixer-settlers [El-Rifai, 1975], and plug flow [El-Rifai et al., 1977] and axial dispersion [El-Nashaie et al., 1978] models in continuous differential contact in spray and packed columns. More elaborate models have been developed for pulsed, agitated, and rotating disc columns which feature strong turbulence and droplets breakage and coalescence. These include mixing cells with recycle [Mijalli, et al., 2005] forward mixing led by particle size distribution [Tang et al., 2005] and CFD models based on population balance [Attarakih et al., 2013].

Un-agitated perforated plate liquid-liquid extraction columns [Garner et al., 1953] are particularly efficient for their high liquid handling capacity and for providing good separation efficiency especially for systems with low interfacial tension which are easily dispersed and do not require mechanical agitation. Owing to the coalescence and re-dispersion taking place on each plate, they may be modeled as a hybrid between discrete and continuous contact equipment [Ettouney et al., 2007].

The purpose of the present paper is to present a design and performance analysis of perforated plate extractive reactors conducting an infinitely fast chemical reaction. A model is developed to study the effect of changing phase flow rates on the obtained extract and raffinate compositions. Reliable empirical correlations [Treybal, 1981] are used to calculate the relevant model parameters which are functions of the hydrodynamic characteristics of perforated plate columns.

2. The Model System

2.1. Model Assumptions

Fig. (1) depicts a multi-stage plate extractive reactor. The organic phase containing solute A at concentration y_o is the dispersed phase entering the bottom of the column, it is extracted down to a raffinate composition y_N . The inlet concentration of reactant B , present in excess in the aqueous solvent continuous phase and insoluble in the dispersed phase, drops from x_{N+1} down to a bottoms composition x_1 . The very fast chemical reaction $A + bB \rightarrow C$ is assumed to take place at the interface. The column hydrodynamics will be modeled based on the assumption of plug flow of the dispersed phase along the active mass transfer height h on each plate, and complete back mixing of the continuous phase on the active area of a plate. The concentration profiles of A and B in the dispersed and continuous phase on each plate corresponding to the above model are depicted in Fig. (1).

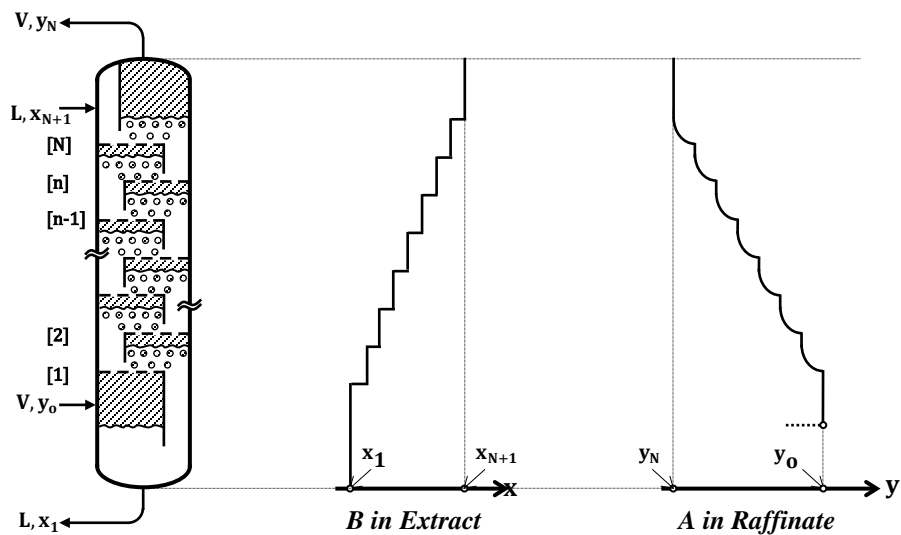


Fig. (1) Nomenclature and composition profiles along a multi-plate extractive reactor

2.2. Model Equation

Since an infinitely fast reaction is assumed, the solute is immediately consumed on reaching the interface [Levenspiel, 1999]. In this case the rate of mass transfer of the solute from the raffinate phase is proportional to its concentration as shown by the two film representation shown in Fig. (2-a).

A steady state differential mole balance on reactant A in the dispersed phase at a level enclosed between plates $n-1$ and n gives

$$\frac{dy}{y} = \frac{-k_{Ad}a'\Phi_d A_a}{V} dz \quad (1)$$

which integrates to give the ratio of the dispersed phase reactant concentrations entering and leaving the effective contact height h as

$$\frac{y_n}{y_{n-1}} = e^{-\frac{\tau_d}{\tau_D}} \quad (2)$$

Where

$$\tau_d = \frac{\Phi_d A_a h}{V} \quad (3)$$

$$\tau_D = \frac{1}{k_{Ad}a'} \quad (4)$$

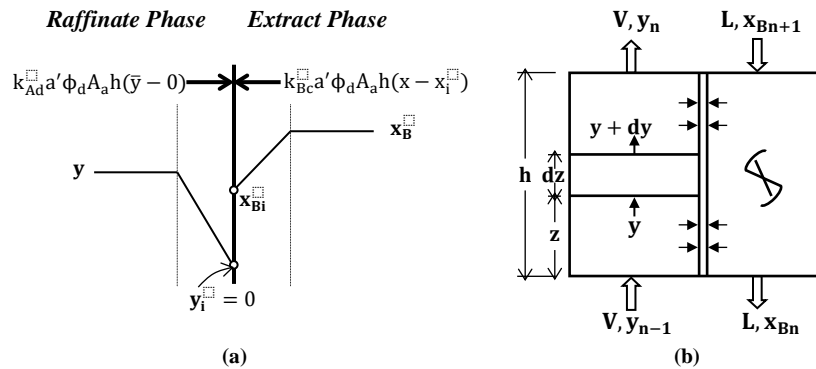


Fig. (2) (a) two film representation of concentration profiles

(b) Mass balance on plate (n)

The average dispersed phase concentration within a plate \bar{y} may be calculated as the mean value between the input and output dispersed phase concentrations

$$\bar{y} = \frac{1}{h} \int_0^h y dz \quad (5)$$

The combination of Eqs. (1) and (5) integrates to

$$\bar{y} = \frac{V}{k_{Ad}a'\Phi_d A_a h} \left(e^{\frac{\tau_d}{\tau_D}} - 1 \right) y_n \quad (6)$$

Mole balance on reactant B in the continuous phase gives

$$Lx_{B_{n+1}} = Lx_{B_n} + k_{Bc} a' \Phi_d A_a h (x_{B_n} - x_{B_{in}}) \quad (7)$$

Where $x_{B_{in}}$ is the average concentration of B at the interface

Referring to Fig. (2), the rates of mass transfer of solute A and reactant B to the interface are related to the reaction stoichiometry by

$$k_{Bc}(x_{B_n} - x_{B_{in}})a' \Phi_d A_a h = bk_{Ad} \bar{y} a' \Phi_d A_a h \quad (8)$$

Substitution of \bar{y} from Eq. (5) into Eq. (8)

$$k_{Bc}(x_{B_n} - x_{B_{in}})a' \Phi_d A_a h = bV \left(e^{\frac{\tau_d}{\tau_D}} - 1 \right) y_{A_n} \quad (9)$$

Substitution of Eq. (9) into Eq. (7)

$$Lx_{B_{n+1}} = Lx_{B_n} + bV \left(e^{\frac{\tau_d}{\tau_D}} - 1 \right) y_{A_n} \quad (10)$$

Successive application of Eqs. (2) and (10) enables to determine the composition evolution of both reactants in their respective phases. This is graphically displayed in Fig. (3). Alternatively, the number of plates required to achieve a given purification ratio $\frac{y_{AN}}{y_{Ao}}$ may be obtained as

$$N = -\frac{\tau_D}{\tau_d} \ln \left(\frac{y_{AN}}{y_{Ao}} \right) \quad (11)$$

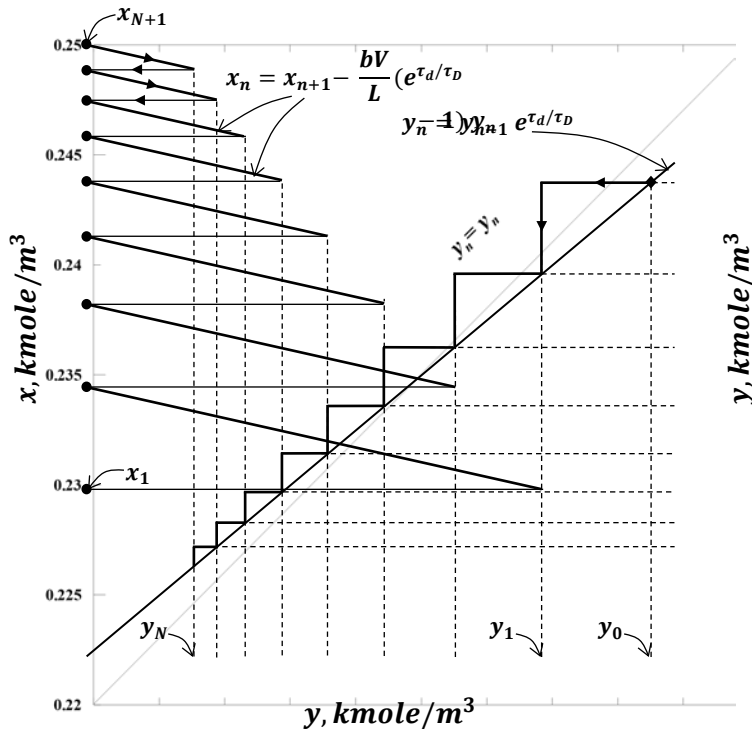
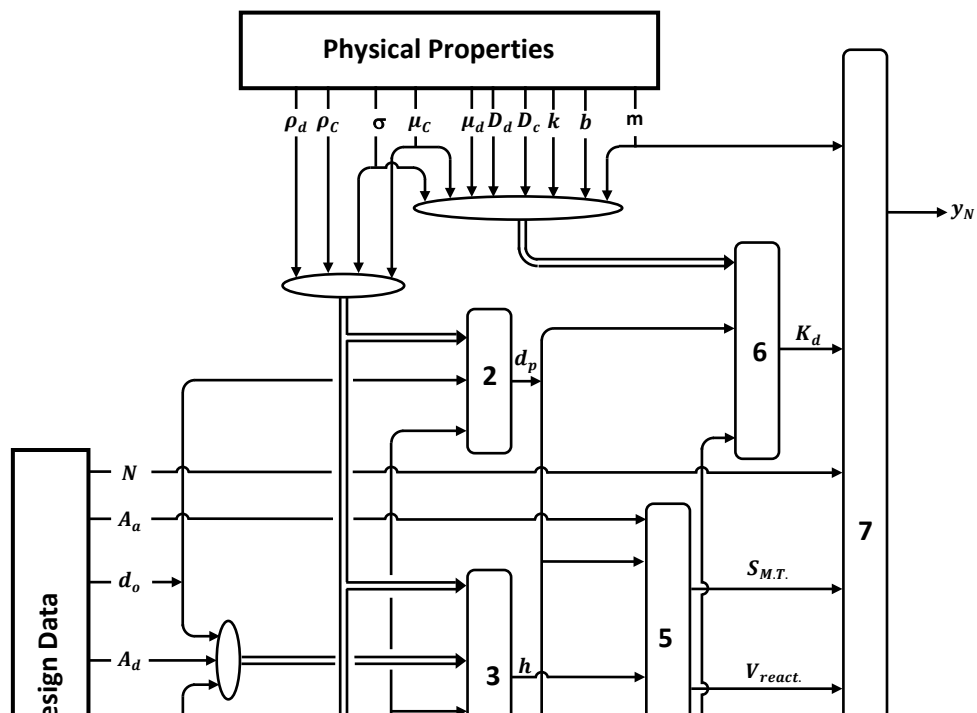


Fig. (3) Graphical Generation of the composition profiles

3. Estimation of Model Parameters

It is clear from the above that the two time constants τ_d and τ_D determine the static characteristics of the class of plate columns conducting an infinitely fast reaction. For given system physical properties and column design dimensions τ_d and τ_D are functions of the phase flow rates. MatLab programs have been developed, on the basis of published correlations, to calculate the relevant variables in terms of the flow rates. Fig. (4) outlines the block diagram of the calculations.



The dispersed phase rate and plate perforations area are used to calculate the hole velocity and check that it lies within an appropriate range [Hayworth and Treybal, 1950; Scheele and Meister, 1968]. The drop diameter is estimated in step (2) from the hole velocity, orifice diameter, and system physical properties [Hayworth and Treybal, 1950]. The active mass transfer height on a plate computed in step (3) is obtained as the difference between plate spacing and the height of the coalesced layer. This corresponds to the sum of the dispersed and continuous phase pressure drops [Bussolari et al., 1953] and may be calculated from the system physical properties, design dimensions, and the flow rate of both phases. The drop diameter, phase flow rate, relevant physical properties, and net area are used in the iterative calculation of the fractional dispersed phase holdup and slip velocity in step (4) [Beyaert et al., 1961; Weaver et al., 1959]. Individual and overall mass transfer coefficients are calculated from appropriate correlations in step (5) [Newman, 1931; Kronig and Brink, 1950; Handlos and Baron, 1957; Garner and Tayeban, 1960; Treybal, 1963; Slater, 1994].

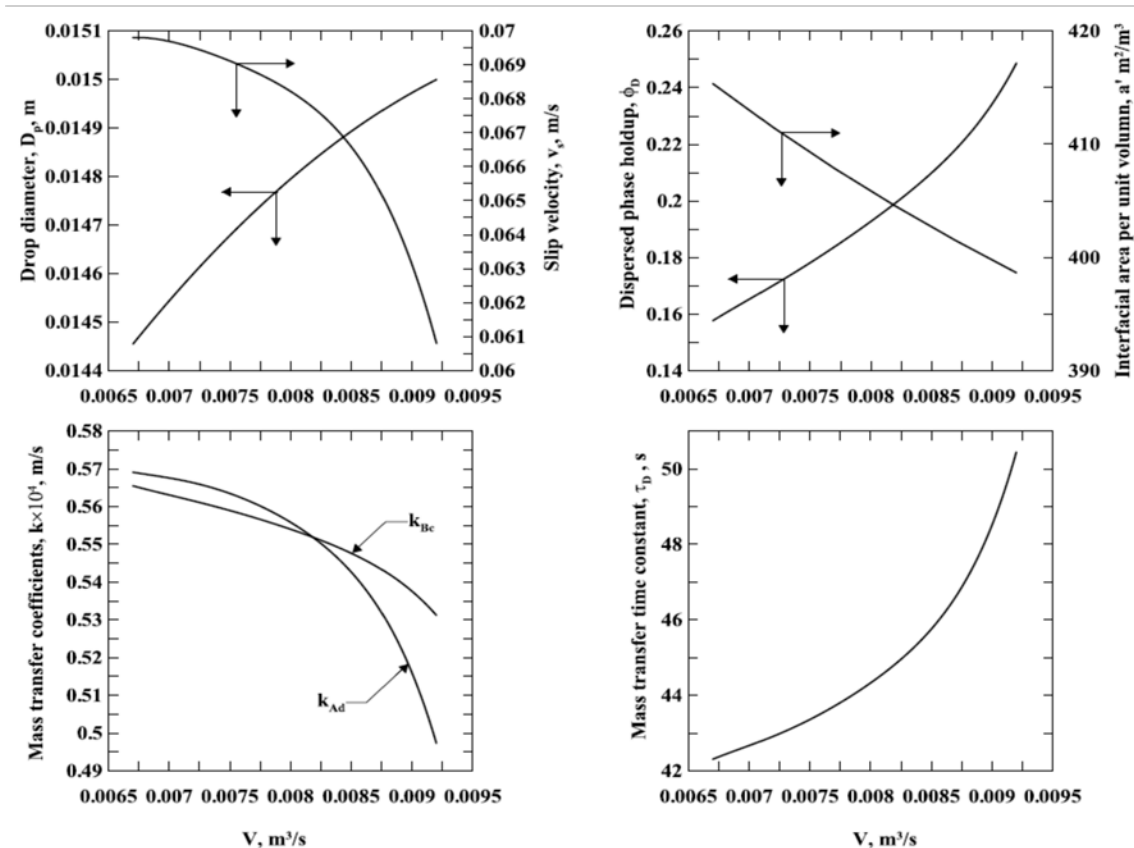


Fig. (5) Effect of dispersed phase flow rate on relevant hydrodynamic and mass transfer parameters

Based on the drop diameter, physical properties, plate area, active mass transfer height, and fractional holdup, the mass transfer surface area and the volume of both the continuous and dispersed phases on a plate are computed in step (6). These are used in step (7) along with the number of plates and overall mass transfer coefficient to calculate the relevant time constants and then the terminal extract and raffinate product compositions depending on the reaction kinetic regime and the phase in which the chemical reaction takes place [Ettouney et al., 2007].

The program has been applied to a 25 plate column extracting HCl (reactant A) from an organic aromatic phase by the infinitely fast reaction with aqueous NaOH (reactant B). The effect of changing the phase flow rates within the stable column operating range on the relevant hydrodynamic parameters, mass transfer coefficients, and the time constants τ_d and τ_D are shown in Figs. (5) and (6).

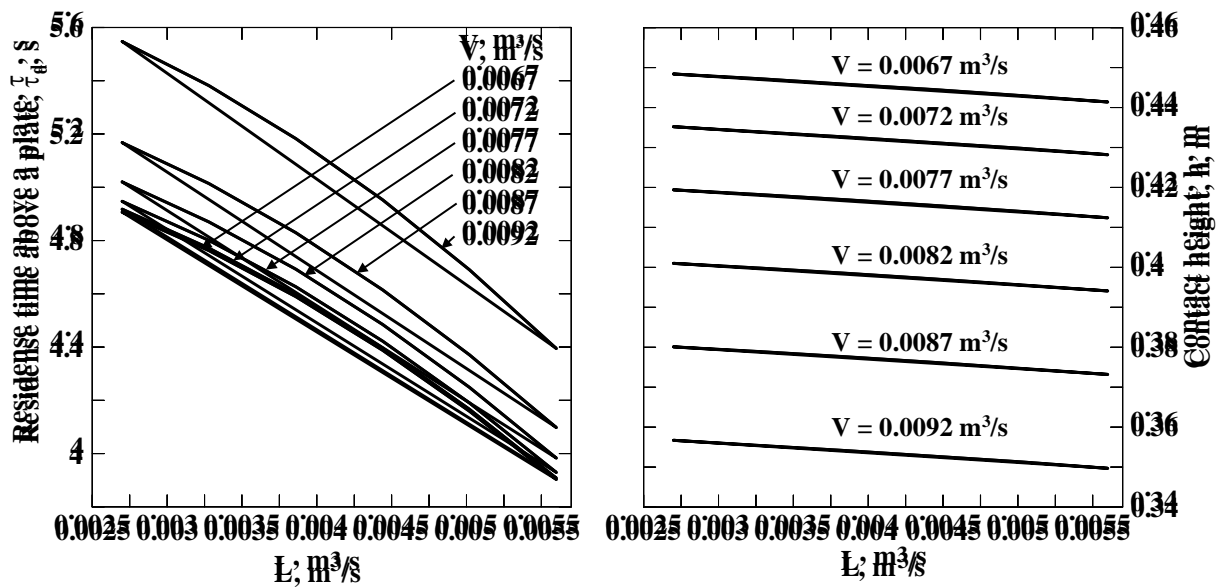


Fig. (6) Effect of both phase flow rates on effective plate contact height and dispersed phase residence time

4. Steady State Response

The steady state concentration profiles along the 25 plate neutralization column are obtainable by successive application of Eqs. (1) and (10) as shown in Fig. (3). Fig. (7) shows typical profiles under flow conditions corresponding to about 94% conversion.

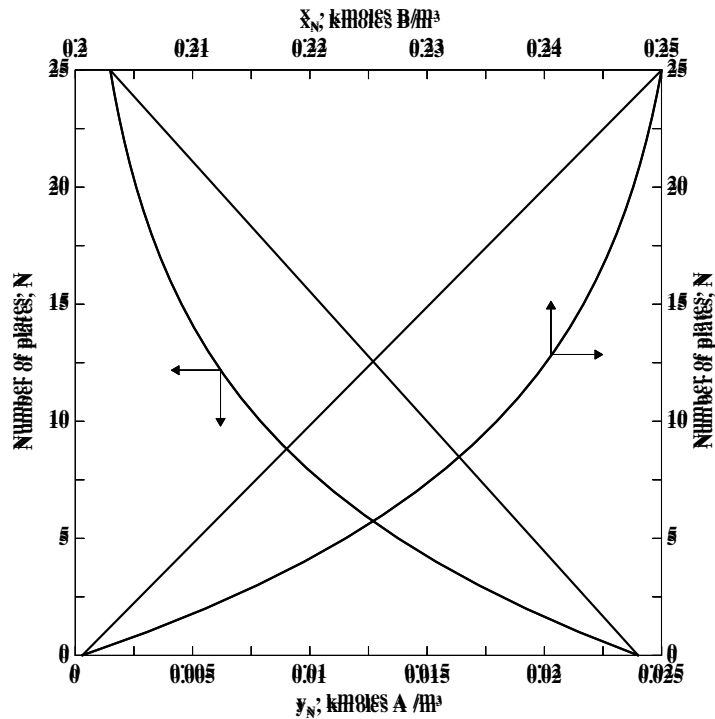


Fig. (7) Typical continuous and dispersed phase concentration profiles in a 25-plate column

When the phase flow rates are varied, the concentration profiles along the column undergo a shift depending on the values of the phase throughputs. This can be illustrated by considering the composition changes at one and the same plate for the spectrum of flow rates lying within the stable operating range. Fig. (8) presents the steady state composition changes at plate number 13 in the 25 plate column as a function of both flow rates. The obtained trends may be elucidated by considering Eq. (2) which shows that for a given plate, as the ratio of the time constants (τ_c/τ_D) increases, the depletion of the solute in the dispersed phase increases. However, (τ_c/τ_D) decreases with the increase in either of the flow rates because this has the effect of decreasing the contact height. Also, the increase in the dispersed phase flow rate results in a decrease in the interfacial area per unit volume and the mass transfer coefficient.

The effect of phase flow rates on the terminal product compositions is shown in Fig. (9) which indicates that the solute concentration in the raffinate increases with the increase of both L and V . The concentration of reactant B in the final extract increases correspondingly.

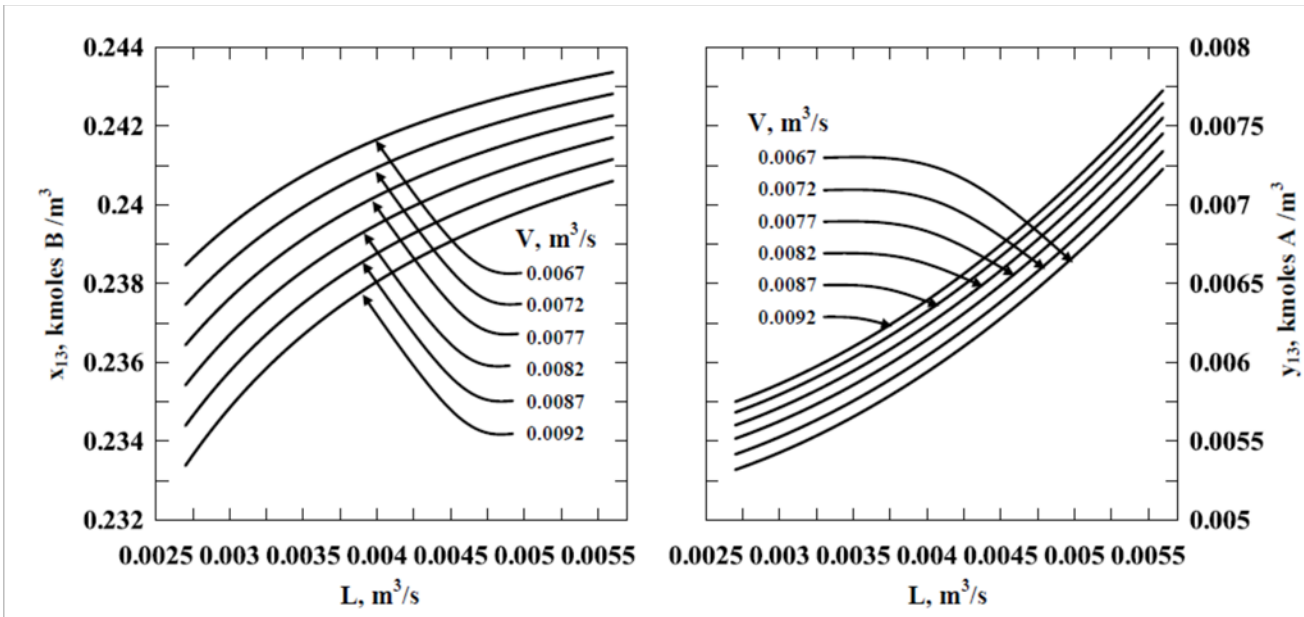


Fig. (8) Effect of phase flow rates on: (a) the concentration of B in extract x_{13} and (b) the concentration of A in raffinate y_{13} in a column conducting a very fast interfacial reaction

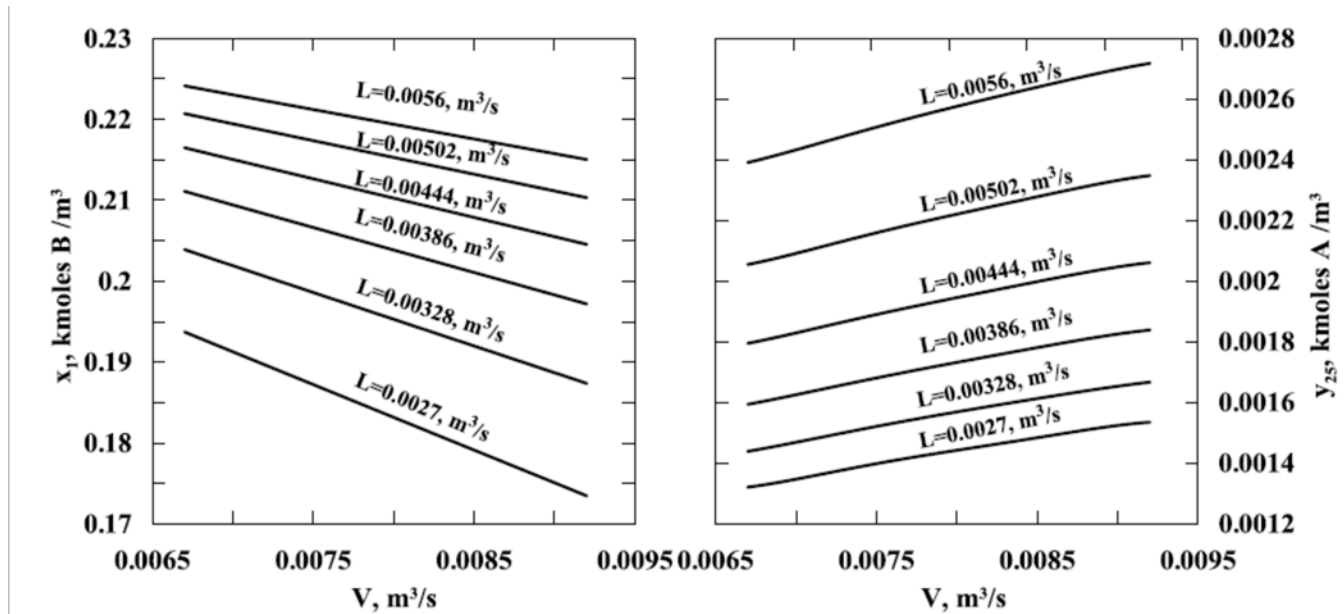


Fig. (9) Effect of phase flow rates on: (a) the concentration of B in extract x_1 and (b) the concentration of A in raffinate y_N in a column conducting a very fast interfacial reaction ($y_0 = 0.024$ kmoles/m³, $x_{N+1} = 0.25$ kmoles/m³)

5. Conclusions

1. A steady state model has been derived for perforated plate liquid-liquid extraction columns conducting a very fast chemical reaction taking place at the interface between the two phases. The steady state model is characterized by two time constants reflecting the interaction between the different physical, kinetic, hydrodynamic, and design parameters.
2. Existing design correlations have been programmed to generate the effect of phase flow rates on the obtained drop diameter, slip velocity, hold up, interfacial area per unit volume, active plate contact height, and mass transfer coefficients. These are integrated in the computations of the model time constants.
3. An analytical and a graphical design method have been developed to calculate the number of actual plates required to achieve a given purification ratio of the dispersed phase raffinate composition. Both methods enable to follow up the composition profiles of both phases along the column for different phase flow rates. Typical steady state composition profiles have been generated for a given column in which extraction of HCl from an organic phase consisting essentially of benzene takes place using an aqueous solution of NaOH.
4. Contrary to intuitive expectations, maintaining the required purification despite increases in either feed concentration or feed flow rate would necessitate reducing rather than increasing the continuous phase flow rate.

Nomenclature

a' : interfacial area per unit volume of dispersed phase (m^2/m^3)

A_a : active plate area (m^2)

b : reaction stoichiometric coefficient

h : active contact height above a plate (m)

k_{Ad} : mass transfer coefficient of A in raffinate phase (m/s)

k_{Bc} : mass transfer coefficient of B in extract phase (m/s)

L : volumetric flow rate of the continuous solvent phase (m^3/s)

n : plate number

N	:	total number of plates
V	:	dispersed phase feed flow rate (m^3/s)
x	:	reactant B concentration in the solvent phase (kmol/m^3)
y	:	reactant A concentration in the dispersed phase (kmol/m^3)
\bar{y}	:	mean dispersed phase reactant A concentration on a plate (kmol/m^3)
z	:	Elevation above tray level, m

Subscript:

A	:	pertaining to solute A
B	:	pertaining to solute B
n	:	leaving plate n
i	:	at interface

Greek Symbols

ϕ_D	:	Dispersed phase holdup volume fraction
τ_d	:	residence time of the dispersed phase on a plate, s
τ_D	:	mass transfer time constant on a plate, s

References

- Attarakih M., Abu-Khader, M., Bart, H.J., 2013. Modeling and dynamic analysis of a rotating disc contactor (RDC) extraction column using one primary and one secondary particle method (OPOSPM). *Chem. Eng. Sci.*, 91, 180–196
- Bart, H.-J., 2001. *Reactive Extraction*. Springer, Berlin.
- Bart, H.-J., Drumm, C., Attarakih, M.M., 2008. Process intensification with reactive extraction columns. *Chem. Eng. Process.* 47, 745–754.
- Beyaert, B. O., Lapidus, L., Elgin, J. C., 1961. The mechanics of vertical moving liquid-liquid fluidized systems: II. Countercurrent flow. *AIChE Journal*, 7(1) 46–48.

Bussolari, R. J., Schiff, S., Treybal, R. E., 1953. [Flow of liquids through perforated-plate liquid extraction towers](#). *Industrial and Engineering Chemistry*, 45(11), 2413–2417.

Danckwerts, P.V., 1970. *Gas-Liquid Reactions*. McGraw-Hill Book Company, New York.

El Nashaie, S. S. E. H., El-Rifai, M. A., and Abd El-Hakim, M. N., 1978. Effect of kinetic regime and axial dispersion on the dynamic response of counter flow extractive reactors. *Chem. Eng. Sci.*, 33, 847-852.

El-Rifai, M. A., 1975. Composition dynamics in multi-mixer-settler extractive reaction batteries. *Chem. Eng. Sci.*, 30 (2), 79-87.

El-Rifai, M.A., El Nashaie, S.S.E.H., Kafafi, A.A., 1977. Analysis of a countercurrent tallow-splitting column. *Trans. Instn. Chem. Eng.*, 55, 59–63.

Ettouney, R. S., El-Rifai, M. A., Ghallab, A. O., 2007. Steady state modeling of perforated plate extraction columns. *Chem. Eng. and Proc.*, 46, 713-720.

Garner, F. H., Ellis, S. R. M., Fosbury, D. W., 1953. Perforated plates in liquid-liquid extraction: toluene-diethylamine-water system., *Chemical Engineering Research and Design*, 31a, 348-362.

Garner, F. H., Tayeban, M., 1960. The importance of the wake in mass transfer from both continuous and dispersed phase systems. *Anales Real Soc. Espan. Fis. Quim*, B56, 479-491.

Handlos, A. E., Baron, T., 1957. Mass and heat transfer from drops in liquid-liquid extraction. *AIChE Journal*, 3(1), 127-135.

Hayworth, C. B., Treybal, R. E., 1950. Drop formation in two-liquid-phase systems. *Industrial and engineering chemistry*, 42, 1174-1181.

Kronig, R., Brink, J., 1950. On the theory of extraction from falling droplets. *Appl.Sci.Res.*, A2, 142-154.

Levenspiel, O., 1999. *Chemical Reaction Engineering*, 3rd edition, John Wiley and Sons, New York, pp 523-539.

Meister, B. J., Scheele, G.F., 1969. Drop formation from cylindrical jets in immiscible liquid systems. *AIChE Journal*, 15(5), 700–706.

Mjalli, F.S., Abdel-Jabbar, N.M., Fletcher, J.P., 2005. Modeling, simulation, and control of a Scheibel liquid-liquid contractor: Part 1. Dynamic analysis and system identification. Chem. Eng. and Proc. 44, 541-553.

Newman, A.B., 1931. The drying of porous solids: diffusion and surface emission equations. AIChE Journal, 27, 203-220.

Ramkrishna, D., 2000. Population balances: theory and applications to particulate systems in engineering. Academic Press, San Diego.

Slater, J.M., 1994. Rate coefficients in liquid-liquid extraction studies. In: Godfrey, J.C., Slater, J.M. (Hrsg.), Liquid-Liquid Extraction Equipment, John Wiley & sons, Chichester.

Tang, X.J., Luo, G.S., Wang, J.D., 2005. An improved dynamic combined model for evaluating the mass transfer performances in extraction columns. Chem. Eng. Sci., 60, 4409–4421.

Treybal, R. E., 1963. Liquid Extraction, McGraw Hill International Editions.

Treybal, R. E., 1981. Mass Transfer Operations, 3rd edition, McGraw Hill International Editions, pp 530-541.

Van Krevelen, D.W., Hoftyzer, P. J., 1953. [Graphical design of gas-liquid reactors](#). Chemical Engineering Science, 2(4) 145-156.

Weaver, R.E.C., Lapidus, L., Elgin, J.C., 1959. The mechanics of vertical moving liquid-liquid fluidized systems: I. Interphase contacting of droplets passing through a second quiescent fluid. AIChE Journal, 5(4) 533–539.