

LIANG CHEN, JENS-UWE REPKE, GÜNTER WOZNY*

LARGE-SCALE PARAMETER ESTIMATION
FOR THREE-PHASE DISTILLATION
BASED ON A NONEQUILIBRIUM MODEL

OGÓLNE OSZACOWANIE PARAMETRÓW
DLA DESTYLACJI TRÓJFAZOWEJ NA PODSTAWIE
MODELU NIERÓWNOWAGOWEGO

Abstract

Mathematical NEQ modeling of three-phase distillation is a fundamental and difficult task, since no reliable correlations available to calculate the inter-phase mass and heat transfer coefficients. Parameter estimation based on a rigorous NEQ model rather than short-cut methods is needed to generate a robust and accurate model. In this paper, parameter estimation is carried out based on a large-scale optimization strategy. The transfer parameters are estimated based on multi-set experiment data. The NEQ model is updated with substantial improvement on the model predictive capability.

Keywords: nonequilibrium model, three-phase distillation, parameter estimation

Streszczenie

Zasadniczym celem artykułu jest matematyczne modelowanie NEQ destylacji trójfazowej. Podstawową trudnością jest brak dostępnych korelacji służących do obliczania współczynników przenikania masy i ciepła w fazie przejściowej. Stworzenie stabilnego i dokładnego modelu wymaga szacowania parametru opartego na rygorystycznym modelu NEQ, a nie na metodach uproszczonych. W niniejszej pracy zaprezentowano szacowanie parametru na podstawie ogólnej strategii optymalizacji. Parametry transportowe zostały oszacowane na bazie multizbioru danych eksperymentalnych. Model NEQ został uaktualniony z uwzględnieniem istotnej poprawy zdolności prognozowania modelu.

Słowa kluczowe: model nierównowagowy, destylacja trójfazowa, szacowanie parametrów

* MSc. Liang Chen, Dr.-Ing. Jens-Uwe Repke, Prof. Dr.-Ing. habil. Prof. h.c. Dr. h.c. Günter Wozny, Institut für Prozess- und Verfahrenstechnik, Technische Universität Berlin.

1. Introduction

Rate-based (or nonequilibrium, NEQ) model is an excellent way for real-world modeling of distillation [1]. In contrast with equilibrium models, rate-based models are governed by the fundamental mass and energy transfer equations, and the computation of Murphree efficiency or HETP is completely avoided. However, the main difficulty for rate-based modeling is the lack of reliable and consistent mass and heat transfer coefficients at vapor–liquid and liquid–liquid interfaces. Especially, for vapor–liquid–liquid (three-phase) distillation, no empirical correlations for mass and heat transfer can be found in the literature. Hence, parameter estimation for inter-phase transfer coefficients is an essential task in developing a NEQ distillation model with comprehensive predictive capability.

Most of the existing approaches for transfer parameter estimation are shortcut methods, either using a simplified mechanical model [2] or using heuristic method like neural network [3]. Parameters obtained with those shortcut models are lack of robustness and may not be directly used for modeling industrial distillation processes, since they are related to specific process setup and laboratory condition. The best approach to tackle this problem is to estimate the parameter using the rigorous distillation model to match sets of given experimental data.

In this paper, a rate-based three-phase distillation model is adopted to identify the mass and heat transfer parameters between all vapor–liquid and liquid–liquid phases. The approach turns out to be a large-scale optimization problem, since it involves more than a large number of model variables and decision variables. As a result, using the optimized rate-based model, substantial improvement on the NEQ model prediction has been obtained.

2. Nonequilibrium modeling

The physical representation of a three-phase packing section is shown in Fig. 1. In contrast to equilibrium model, the material and energy balance in rate-based model are written for all three phases (vapor phase V, first liquid phase L', second liquid phase L'') rather than the whole section. The single phase is linked and influenced by the mass transfer rates $N_{i,j}$ and energy transfer rates En_j . Here, $N_{i,j}''$ is, for example, the molar flux of component i in section j from vapor phase to the first liquid phase.

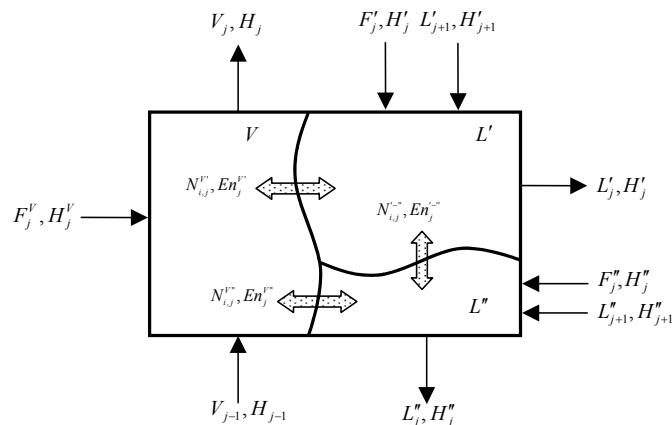


Fig. 1. Schematic diagram of a three-phase packing section

Rys. 1. Schemat przepływu trójfazowego

Component material balance for each phase

$$F_j^V z_{i,j}^V + V_{j-1} y_{i,j-1} - V_j y_{i,j} - (N_{i,j}^{V''}) a_j^{V''} - (N_{i,j}^{V''}) a_j^{V''} = 0 \quad (1)$$

$$F_j^L z_{i,j}^L + L_{j+1}^L x_{i,j+1}^L - L_j x_{i,j}^L + (N_{i,j}^{L''}) a_j^{L''} - (N_{i,j}^{L''}) a_j^{L''} = 0 \quad (2)$$

$$F_j^L z_{i,j}^L + L_{j+1}^L x_{i,j+1}^L - L_j x_{i,j}^L + (N_{i,j}^{L''}) a_j^{L''} + (N_{i,j}^{L''}) a_j^{L''} = 0 \quad (3)$$

Energy balances for each phase

$$F_j^V H_j^V - V_j H_j^V + V_{j-1} H_{j-1}^V - En_j^{V''} - En_j^{V''} + Q_j^V = 0 \quad (4)$$

$$F_j^L H_j^L - L_j H_j^L + L_{j+1}^L H_{j+1}^L + En_j^{L''} - En_j^{L''} + Q_j^L = 0 \quad (5)$$

$$F_j^L H_j^L - L_j H_j^L + L_{j+1}^L H_{j+1}^L + En_j^{L''} + En_j^{L''} + Q_j^L = 0 \quad (6)$$

The basic idea of the NEQ model is the explicit consideration of mass and heat transfer from one phase to another. Mass transfer is driven by gradients in concentration between the bulk phase and the interface. The term $K_{i,m,j}$ represents the matrix of multicomponent mass transfer coefficients, which can be calculated from binary mass transfer coefficients using the Maxwell-Stefan approach. To calculate the energy transfer coefficients, the well-known Chilton-Colburn analogy is used [4].

Mass transfer equations

$$N_{i,j}^{V'} = \bar{C}_j^{V'} \sum_{m=1}^{n-1} \left(k_{i,m,j}^{V'} \left(\bar{y}_{m,j}^{V'} - y_{m,j}^{V'} \right) \right) + \bar{y}_{i,j}^{V'} \sum_{i=1}^n N_{i,j}^{V'} \quad (7)$$

Vapor–Liquid I
Interface

$$N_{i,j}^{V''} = \bar{C}_j^{V''} \sum_{m=1}^{n-1} \left(k_{i,m,j}^{V''} \left(x_{m,j}^{V''} - \bar{x}_{m,j} \right) \right) + \bar{x}_{i,j} \sum_{i=1}^n N_{i,j}^{V''} \quad (8)$$

Vapor–Liquid II
Interface

$$N_{i,j}^{V'''} = \bar{C}_j^{V'''} \sum_{m=1}^{n-1} \left(k_{i,m,j}^{V'''} \left(\bar{y}_{m,j}^{V'''} - y_{m,j}^{V'''} \right) \right) + \bar{y}_{i,j} \sum_{i=1}^n N_{i,j}^{V'''} \quad (9)$$

$$N_{i,j}^{V''''} = \bar{C}_j^{V''''} \sum_{m=1}^{n-1} \left(k_{i,m,j}^{V''''} \left(x_{m,j}^{V''''} - \bar{x}_{m,j} \right) \right) + \bar{x}_{i,j} \sum_{i=1}^n N_{i,j}^{V''''} \quad (10)$$

Liquid I–Liquid
II Interface

$$N_{i,j}^{L'} = \bar{C}_j^{L'} \sum_{m=1}^{n-1} \left(k_{i,m,j}^{L'} \left(\bar{x}_{m,j} - x_{m,j}^{L'} \right) \right) + \bar{x}_{i,j} \sum_{i=1}^n N_{i,j}^{L'} \quad (11)$$

$$N_{i,j}^{L''} = \bar{C}_j^{L''} \sum_{m=1}^{n-1} \left(k_{i,m,j}^{L''} \left(x_{m,j}^{L''} - \bar{x}_{m,j} \right) \right) + \bar{x}_{i,j} \sum_{i=1}^n N_{i,j}^{L''} \quad (12)$$

Energy transfer equations

$$En_j^{V'} = \left(\alpha_j^{V'} \left(\bar{T}_j^{V'} - T_j^{V'} \right) + \sum_{i=1}^n \left(N_{i,j}^{V'} \bar{h}_{i,j}^{V'} \right) \right) a_j^{V'} \quad (7)$$

Vapor–Liquid I
Interface

$$En_j^{V''} = \left(\alpha_j^{V''} \left(T_j^{V''} - \bar{T}_j \right) + \sum_{i=1}^n \left(N_{i,j}^{V''} \bar{H}_{i,j} \right) \right) a_j^{V''} \quad (8)$$

Vapor–Liquid II
Interface

$$En_j^{V'''} = \left(\alpha_j^{V'''} \left(\bar{T}_j^{V'''} - T_j^{V'''} \right) + \sum_{i=1}^n \left(N_{i,j}^{V'''} \bar{H}_{i,j}^{V'''} \right) \right) a_j^{V'''} \quad (9)$$

$$En_j^{V''''} = \left(\alpha_j^{V''''} \left(T_j^{V''''} - \bar{T}_j \right) + \sum_{i=1}^n \left(N_{i,j}^{V''''} \bar{H}_{i,j} \right) \right) a_j^{V''''} \quad (10)$$

Liquid I–Liquid
II Interface

$$En_j^{L'} = \left(\alpha_j^{L'} \left(T_j^{L'} - \bar{T}_j \right) + \sum_{i=1}^n \left(N_{i,j}^{L'} \bar{H}_{i,j} \right) \right) a_j^{L'} \quad (11)$$

$$En_j^{L''} = \left(\alpha_j^{L''} \left(T_j^{L''} - \bar{T}_j \right) + \sum_{i=1}^n \left(N_{i,j}^{L''} \bar{H}_{i,j} \right) \right) a_j^{L''} \quad (12)$$

In Table 1, we list detailed empirical correlations to calculate the binary mass transfer coefficient [2] (for 2-phase distillation), heat transfer coefficient (Chilton-Colburn analogy),

and effective interfacial area [2] (for 2-phase distillation). Here, binary diffusion coefficient D_V and D_L are calculated by Fuller Eqs and Tyn-Calus Eqs, Vignes Eqs respectively.

Table 1

Correlations for mass and heat transfer calculation	
Category	Equation
Binary mass transfer coefficient	$\beta_{VL} = C_V \frac{1}{(\varepsilon - h_L)^{1/2}} \frac{a_p^{3/2}}{d_h^{1/2}} D_V \left(\frac{u_V}{a_p v_V} \right)^{4/3} \left(\frac{v_V}{D_V} \right)^{1/3} \left(\frac{1}{a_p} \right)$ $\beta_{LV} = 12^{1/6} C_L \left(\frac{u_L}{H_L} \right)^{1/2} \left(\frac{D_L}{d_L} \right)^{1/2}$ $\beta_{L'L''} = \frac{D_L}{\delta_L}, \quad \delta_L = \left(\frac{3v_L L}{A \rho_L a_p g (\sin \kappa)^2} \right)^{1/3}$
Heat transfer coefficient	$\alpha_{LR}^V = \bar{\beta}^V \rho^V c_p^V \left(\frac{\lambda^L}{c_p^V \rho^V D^V} \right)^{2/3}, \quad \alpha_{LR}^L = \bar{\beta}^L \rho^L c_p^L \left(\frac{\lambda^L}{c_p^L \rho^L D^L} \right)^{1/2}$ $\alpha = \alpha_{LR} \frac{\Phi_H}{\exp \Phi_H - 1} \quad \text{with} \quad \Phi_H = \frac{\sum_{i=1}^n N_i c_{p,i}}{\alpha_{LR}}$
Effective interfacial area	$\frac{a}{a_p} = 1.5 (a_p d_h)^{-0.5} \left(\frac{u_L d_h}{v_L} \right)^{-0.2} \left(\frac{u_L \rho_L d_h}{\sigma_L} \right)^{0.75} \left(\frac{u_L^2}{g d_h} \right)^{-0.45}$
Maxwell-stefan equation	$[k^V] = [B^V]^{-1} [\Xi^V], \quad [k^L] = [B^L]^{-1} [\Gamma] [\Xi^L]$ $B_{ii} = \frac{y_i}{\beta_{in}} + \sum_{\substack{k=1 \\ i \neq k}}^n \frac{y_k}{\beta_{ik}}, \quad \text{or} \quad B_{ij} = -y_i \left(\frac{1}{\beta_{ij}} - \frac{1}{\beta_{in}} \right)$

3. Parameter estimation problem formulation

3.1. Parameter to be estimated

The accuracy of the above NEQ model depends strongly on the appropriate parameters. To select the parameters that have to be estimated, detailed analysis is carried out.

Effective interfacial area. The effective interfacial area is crucial to determine the mass and heat transfer in both vapor and liquid interfaces as shown in Eqs (1)–(3) and (7)–(12). Our former contribution has demonstrated that it is the most sensitive parameter in a three-phase NEQ model [6]. Simulation results indicate that the overall effect on the column performance by step response of effective interfacial area a is much greater than those of other parameters (e.g. β_{VL} , β_{LV}). Hence, Billet Eqs is modified to Eqs (13) to estimate the effective interfacial area for both vapor–liquid and liquid–liquid interfaces.

$$\frac{a}{a_p} = \theta_1 (a_p d_h)^{-0.5} \left(\frac{u_L d_h}{v_L} \right)^{-0.2} \left(\frac{u_L \rho_L d_h}{\sigma_L} \right)^{0.75} \left(\frac{u_L^2}{g d_h} \right)^{-0.45} \quad (13)$$

Binary mass transfer coefficients. Since the Maxwell-Stefan equations are established based on a solid theoretical foundation, binary mass transfer coefficients could be another main uncertainty contributing to model variation. Although a set of C_V , C_L are given in Billet's work [2], parameter re-estimation is still needed for unknown packing and material system. In this paper, we define $\theta_2 = C_V$, $\theta_3 = C_L$ to introduce another two degrees of freedom.

The most complicated issue lies in liquid-liquid interface, where no empirical mass transfer formulation can be found in the literature. Here, two more degrees of freedom are added in Eqs (14)

$$\beta_{LL'} = \theta_4 D_L \left(\frac{3v_L L}{A \rho_L a_p g (\sin \kappa)^2} \right)^{\theta_5} \quad (14)$$

Heat transfer coefficient. Mass transfer is an important aspect that motivates much research effort, however the estimation of heat transfer coefficients is normally ignored for simplification. In this paper heat transfer coefficients are estimated meanwhile to carry out a thorough analysis of the mass and heat transfer in three-phase distillation

$$\alpha_{LR}^V = \bar{\beta}^V \rho^V c_p^V \left(\frac{\lambda^L}{c_p^V \rho^V \bar{D}^V} \right)^{\theta_6}, \quad \alpha_{LR}^L = \bar{\beta}^L \rho^L c_p^L \left(\frac{\lambda^L}{c_p^L \rho^L \bar{D}^L} \right)^{\theta_7} \quad (15)$$

3.2. Large-scale optimization

A general parameter estimation problem with multiple data sets can be formulated as follow [5]. Here, (decision variables) $\Theta = [\theta_1, \dots, \theta_7]$ are the parameters to be estimated, (dependent variables) \mathbf{y} consist of measured temperature and concentration ($\tilde{\mathbf{y}}$, experiment profiles), and \mathbf{x} are other unmeasured state variables in the model. In the objective function, \mathbf{W} is a given weight matrix, covariance matrices of the measurement errors. It is noted variables \mathbf{y} as well as \mathbf{x} vary with each individual data sets, while the parameters Θ are independent of data sets

$$\begin{aligned} \min \sum_{i=1}^{NDS} (\mathbf{y}_i - \tilde{\mathbf{y}}_i)^T \mathbf{W}^{-1} (\mathbf{y}_i - \tilde{\mathbf{y}}_i) \\ \text{st.} \\ \mathbf{g}_i(\mathbf{x}_i, \mathbf{y}_i, \Theta) = 0 \\ \mathbf{h}_i(\mathbf{x}_i, \mathbf{y}_i, \Theta) \geq 0 \\ \Theta^L \leq \Theta \leq \Theta^U \end{aligned} \quad (16)$$

Here, \mathbf{g} is the vector of model equations and \mathbf{h} the vector of inequality constraints. The three-phase NEQ model contains more than 2000 algebraic equations. Moreover, the complexity of the problem increases linearly with the number of data sets [5]. For instance, with 3 data sets, the optimization engages more than 6000 state and dependent variables to

estimate 21 decision variables. This is a highly nonlinear, stiff large-scale problem, which is computationally expensive [7].

4. Case study

Two examples are considered here. The first one utilizes the experiment data published by Pelkonen et al. for two-phase distillation equipped with Sulzer BX packing [8]. The parameter estimation is carried out based on a 2-phase NEQ model (a special case of 3-phase NEQ model). The second example uses measured data of our own experiment. Our investigations were carried out in a 0,07 m diameter column packed with Sulzer Optiflow C3.6. The concentration and temperature data were sampled along the column and used as the basic data sets.

Taking effective interfacial area for example, as the most sensitive parameter, the optimized value was found about 1,5 times bigger than that computed by Billet method. For the first example, $\theta_1 \approx 2,25$. Although it's not intuitive, the optimized value is reasonable. We re-calculate the effective interfacial area by Rocha method [9], $a_{Rocha} = 0,2998 (m^2 / m^3)$, $a_{Billet} = 0,1457 (m^2 / m^3)$. Therefore, the estimated value $a_{opt} = 0,2294 (m^2 / m^3)$ is a tradeoff between both methods. And the model prediction responds well with the experiment data, indicating a significant improvement in model accuracy.

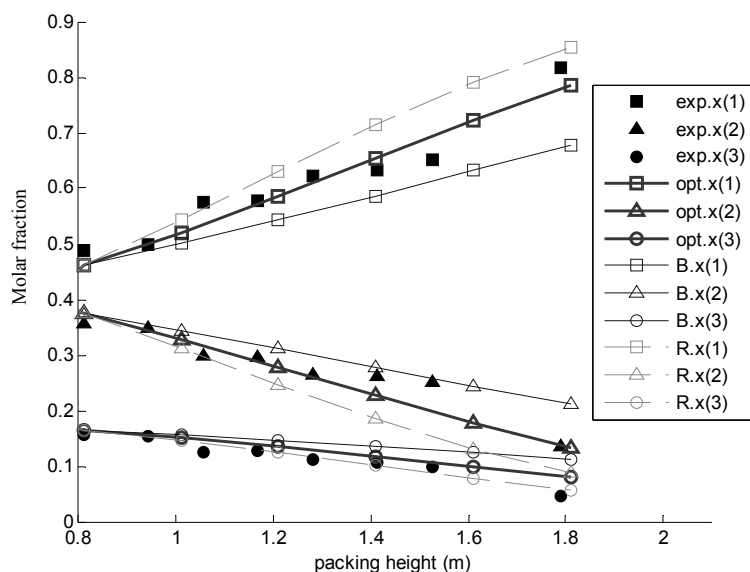


Fig. 2. Model prediction with and without optimization (system: ethanol (1), propanol (2), water (3)). (B: Billet method; R: Rocha method; opt: optimized interfacial area; exp: experiment data)

Rys. 2. Modele z optymalizacją i bez optymalizacji (układ: etanol (1), propanol (2), woda (3))

More detailed results will be given in our presentation.

5. Conclusion

Mass and heat transfer parameters are the key elements of NEQ distillation modeling. Parameter estimation based on a rigorous model is needed to generate a set of reliable and consistent transfer parameters. The estimated model parameters can be compared with an existing correlation or used to develop new correlations. The first results will be shown in the presentation.

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