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"*A simultaneous approach for calibrating Rate Based Models of packed distillation columns based on multiple experiments*"

by *J. Bonilla, F. Logist, B. Huyck, J. Degrève, B. De Moor, J. Van Impe*

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A simultaneous approach for calibrating Rate Based Models of packed distillation columns based on multiple experiments

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Abstract

This paper presents the calibration of a steady state Rate Based Model (RBM) for distillation based on multiple experiments. A packed column is considered, using non-equilibrium stages to represent the packing segments. For an efficient and accurate calibration, the number of model equations and parameters is first reduced via analytical manipulations and a sensitivity analysis. Second, the model is formulated such that a sparse and banded Jacobian can be exploited. Finally, novel constraints on the physical parameters are derived such that the parameter estimation yields consistent results. The model prediction capabilities are successfully validated with experimental data.

Keywords: Distillation, packed columns, parameter estimation, nonlinear programming, structure exploitation, simultaneous estimation.

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1. Introduction

Distillation is not only the most common but also one of the most energy intensive unit operations in chemical industry. An excellent way to reduce the operating costs and energy consumption is by an appropriate (model-based) control of, e.g., applied heat, temperatures, product compositions and flows. However, this requires models which describe accurately the plant behavior. Developing accurate first-principle models is a challenge due to the nonlinear and complex nature of the equations. Moreover, this complexity is even increased when model calibration is aimed at, which simultaneously accounts for experiments under different operating conditions. Although distillation is in industry often performed using tray columns, packed columns are in general more efficient since the vapor and liquid are continuously in contact through the packing surface, enhancing the mass and energy transfer. Packed columns can be modeled using concepts from tray columns such as stage efficiency and height equivalent of a theoretical plate (Seader and Henley, 2006). However, these concepts do not account for deviations from equilibrium properly, in contrast to Rate Based Models, which divide the packing in segments and model them as a mass and energy transfer unit (Khrishnamurthy and Taylor, 1985; Taylor and Krishna, 1993). The aim is to propose a *simultaneous method* for calibrating a steady-state Rate Based Model using multiple experiments and to validate it on a binary pilot-scale distillation column. A similar plant was recently studied in Barroso et al. (2009); Chambel et al. (2011). To ensure an accurate and computationally efficient result, (*i*) model and parameter reduction, (*ii*) problem structure

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exploitation and (*iii*) additional constraints have been used.

2. Experimental context

The experimental setup involves a computer controlled packed distillation column (Figure 1). The column has an internal diameter of 7 cm containing three sections of 960 mm Sulzer CY packing. The feed stream containing methanol and isopropanol can be introduced into the column between the packed sections S_1 and S_2 or S_2 and S_3. The feed temperature can be adjusted by an electric heater of maximum 250 W. In the reboiler located at the bottom, two electric heaters of maximum 3000 W vaporize part of the liquid. The rest is extracted as bottom stream. A total condenser at the top allows condensing the entire overhead vapor stream, which is collected in a reflux drum. Part of the condensed liquid is fed back to the column as reflux, while the remainder leaves the column as distillate. Four variables can be manipulated: the reboiler duty Q_R (W), the feed rate F (g/min), the duty of the feed heater Q_F (W) and the reflux flow rate L_N (g/min). The distillate flow D (g/min) is adjusted to maintain a constant reflux drum level. Measurements are available for the reflux flow rate L_N, the distillate flow rate D, the feed flow rate F and twelve temperatures. These temperatures are the reflux temperature $T_{s_{12}}$, the temperature at the top of the condenser $T_{s_{11}}$, the temperatures in the center and extremes of each packing section (T_{s_2} to $T_{s_{10}}$), the temperature of the feed point T_F, and the temperature in the reboiler T_{s_1}. The concentrations in the distillate and bottom streams are measured offline by sampling.

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3. Simultaneous formulation for model calibration

The steady-state model can be described by a set of nonlinear equations in the form $F(\mathrm{x}, p) = 0$ with x the model states and p the model parameters to be calibrated. Typically, the parameter values have to be optimised such that model predictions are as close as possible to the measured outputs y. In a *sequential approach* only the parameters are degrees of freedom in the optimization problem while the model equations are solved each time as an inner simulation problem. Alternatively, in a *simultaneous approach*, both the parameter and state variables are degrees of freedom in the optimization problem, while the model equations are introduced as additional equality constraints. This is a more efficient approach that preserves sparsity in the optimization, at the cost of increasing the number of optimization variables. Hence, the optimization problem is cast as:

$$\min_{\mathrm{x},p} \ \|\bar{\mathrm{y}} - \mathrm{Cx}\|^2_{Q_\mathrm{x}} \text{ subject to } \begin{cases} F(\mathrm{x}, p) &= 0 \\ \mathrm{x}_{\min} \leq \mathrm{x} &\leq \mathrm{x}_{\max} \\ p_{\min} \leq p &\leq p_{\max} \end{cases} \tag{1}$$

where the vector $\bar{\mathrm{y}}$ represents the measurement data, C is a positive semidefinite diagonal matrix with zero entries in the diagonal corresponding to the states that are not measured and Q_x is a weight matrix. Note that this formulation accounts only for one experiment. In order to use multiple experiments, the optimization vector, the residual vector and the constraints are augmented such that:

$$\begin{aligned} \tilde{\mathrm{y}}^T &= [\bar{\mathrm{y}}_1^T, \ldots, \bar{\mathrm{y}}_M^T], \quad \mathrm{w}^T = [\mathrm{x}_1^T, \ldots, \mathrm{x}_M^T, p], \\ \mathrm{F(w)}^T &= [F(\mathrm{x}_1, p)^T, \ldots, F(\mathrm{x}_M, p)^T] \end{aligned} \tag{2}$$

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and the problem is formulated as:

$$\min_{\mathrm{w}} \ \|\tilde{\mathrm{y}} - \tilde{\mathrm{C}}\mathrm{w}\|^2_{Q_\mathrm{w}} \text{ subject to } \begin{cases} \mathrm{F}(\mathrm{w}) & = & 0 \\ \mathrm{w}_{\min} & \leq \mathrm{w} \leq & \mathrm{w}_{\max} \end{cases} \tag{3}$$

with appropriate matrices Q_w, $\tilde{\mathrm{C}}$, and appropriate bounds $\mathrm{w}_{\min}$, $\mathrm{w}_{\max}$. As the size of the optimization problems grows with the number of experiments, model reduction and structure exploitation are of major importance.

4. Numerical approach

This section details the steps taken to accurately and efficiently solve the calibration problem.

4.1. Model reduction

Starting from a general Rate Based Model (Khrishnamurthy and Taylor, 1985), a reduced order model is derived based on the following assumptions.

- A binary mixture is considered.
- Bulk phases are perfectly mixed.
- Vapor-liquid equilibrium is only valid at the vapor-liquid interface.
- The reboiler and condenser are in thermodynamical equilibrium.
- The liquid volumes of reboiler and reflux drum, $\bar{v}_R$ and $\bar{v}_D$, are perfectly controlled.
- Each stage is in mechanical equilibrium.

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- Equilibrium holds in the condenser. Subcooling in the condenser is attributed to the heat loss in the reflux drum.

The reduction is performed by inclusion of these assumptions as well as differentiation of enthalpy correlations and algebraic manipulation of energy and mass balance equations. As a result the number of variables and equations in the Reduced Order Rate Based Model (RORBM) is now $14N - 12$ instead of $20N - 17$ in the Full Rate Based Model (FRBM), with N the number of stages (Bonilla, 2011; Bonilla et al., 2012). This means a reduction of $6N - 5$ equations, or approximately 30%.

4.2. Parameter reduction

To reduce the number of parameters to be estimated, a parameter sensitivity analysis is performed, i.e., an analysis of the effect of the parameters on the model states. This analysis indicates which parameters can be estimated from the steady state measurements.

4.3. Structure exploitation

The steady state constraints give rise to a root finding problem where a nonlinear system of equations of the form $F(\mathrm{x}, p) = 0$ has to be solved. Although its structure is obtained directly from the formulation of balances and equilibrium relations separately, the variables and equations can be grouped per stage in order to exploit sparsity. This leads to a sparse and reduced band pattern in the model Jacobian (see Bonilla et al. (2012) for the actual re-arranged banded structure). This banded structure is known in distillation models and *equation-tearing* methods have been proposed to solve the nonlinear system of equations (Seader and Henley, 2006; Taylor and Krishna,

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1993). However, a Newton based approach yields a more general procedure and provides more flexibility in the problem specification (Seader and Henley, 2006). Hence, the sparsity pattern is exploited (Dongarra et al., 1988) at each Newton iteration of the preconditioned conjugate gradient algorithm used. Proper scaling of the equations accelerates the convergence (Nocedal and Wright, 2006).

4.4. Physically inspired constraints

Although the parameters have a physical meaning, some combinations of their positive values have been observed to yield state profiles that are physically impossible (Bonilla, 2011). This issue can be solved by adding proper constraints. As the current RBM does not incorporate any constraint by itself that restrict pairs (y^V, T^V) or (x^L, T^L)[1] to superheated vapor or subcooled liquid regions, respectively. This translates into additional inequality constraints, which require the computation of the dew and bubble point curves and, hence, pose an embedded root finding problem. However, as a simultaneous approach is exploited the relations for dew and bubble points can be solved as a part of the model equations.

5. Results and discussion

This section presents and discusses the obtained results.

[1] y^V represents the composition in the bulk vapor phase, x^L the composition in the bulk liquid phase, T^V the temperature of the bulk vapor phase and T^L the temperature of the bulk liquid phase.

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5.1. Computational speedup

To illustrate the necessity of the model reduction and sparsity exploitation, a column with N equal to 20 is simulated as a test case. The computational statistics can be found in Table 2. Clearly, when going from the full to the reduced order rate based model, the number of function evaluations and computation time is roughly halved. However, when exploiting sparsity, it is seen that a 90% reduction is possible. Hence, the combined effect yields a 20 times speedup. Moreover, this effect will only grow when the size is increases, e.g., by simultaneously considering multiple experiments.

5.2. Experimental data

Five experiments are used for identification and two for validation. The values for the manipulated variables are presented in Table 3 while the measured steady state profiles for eleven temperature sensors are illustrated in Figure 7. Note that the measurement coming from the sensor in the condenser $T_{s_{11}}$ corresponds to a temperature below the boiling point of pure methanol at atmospheric pressure, i.e., $T_{s_{11}} < 338$ K. Hence, assuming that the methanol composition at the top is close to one, it would be difficult for an equilibrium condenser or for the condenser proposed here, without subcooled product, to fit this temperature. Consequently, in the parameter estimation, the measurement coming from the condenser is weighted in a smaller proportion with respect to the rest of measurements. On the other hand, the parameter estimation is formulated such that the temperature of the liquid bulk phase of the model fits the measurement data. The vapor phase is not used here since measured profiles seems to adjust better to a subcooled liquid phase than to a vapor phase. The physical explanation is

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related to the fact that condensed liquid does not directly detaches from the spiral condenser after condensation, but runs down the spiral before falling down and wetting the temperature sensor.

5.3. Model calibration and validation

Figure 3 depicts the normalized sensitivity of the vapor and liquid temperatures with respect to model parameters along with sensitivities for the liquid composition at top and bottom stages. On the one hand, the heat loss coefficients (ψ_L, ψ_V, ψ_R and ψ_C) and feed composition (x_F) exhibit a considerable effect over the steady state profiles while the mass transfer coefficients (C_{k^L} and C_{k^V}) have less influence in the steady state temperature profiles. On the other hand, it is clear that volumes in the reboiler and condenser ($\bar{v}_R$ and $\bar{v}_C$) along with pressure drop ($C_{\Delta p}$) and liquid holdup coefficients (C_h) cannot be estimated from the steady state temperature measurements. As this study only considers a pilot-scale column that is made out of glass and is not insulated for educational reasons, sensitivity results can be different for industrial columns. Nevertheless a sensitivity analysis will in general always reveal the most informative parameters.

Hence, only parameters $\phi_L, \phi_V, \phi_R, \phi_D, C_{k^L}, C_{k^V}$ and x_F have to be tuned. To this end the deviations between the predicted temperatures for the liquid bulk phase and the measured temperatures is minimised. Initial values, bounds and obtained estimates are presented in Table 1. The experiments are performed under different conditions, allowing for different feed compositions, x_F. Consequently, a different value of x_F is estimated for each experiment. This increases the number of parameters again to $N_p = 6 + M$

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where M is the number of experiments used for identification. Due to the structure of the setup, N is selected equal to 20. Five experiments are used for the estimation task ($M = 5$), leading to $N_p = 11$ and an optimization problem of size 1351 ($M(14N - 12) + N_p$).

Figure 4 (top) illustrates the fit results for the identification set, while Figure 4 (bottom) displays the results for the validation set. Although the estimation problem is nonlinear, a simultaneous approach estimating both the states and parameters at the same time allows to initialise also the states based on the available measurements and, hence, reduces the chances of getting stuck in a local minimum. Note that there is a group of points that lies outside de ±3 K band around 335 K. These are measurements obtained from the condenser which cannot be totally explained by the model due to the inability to model subcooled liquid at this place. In the real setup, the liquid stream leaving the condenser is subcooled when falling along the spiral condenser. This liquids falls into the reflux drum decreasing its temperature. Since the model assumes that the liquid coming out from the condenser is at equilibrium, it cannot reach temperatures that the modified Rault's law does not predict. Hence, the only form of obtaining a subcooled liquid in the drum is increasing the heat loss coefficient in the reflux drum ϕ_C. Consequently, the heat that is removed by the subcooling of the liquid falling in the real condenser is compensated by the reflux drum losses in the model. A better representation of what is happening in the condenser can be achieved by using a non-equilibrium stage. This implies, however, increasing the complexity of the condenser model, since holdups for the non-equilibrium condenser have

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to be estimated from its geometry. This estimation is in general not trivial and the current model already provides an acceptable prediction.

Figure 5 displays the boiling point diagrams for the validation set with experiments 6 and 7 from Table 3. This figure illustrates the consistency of the results, i.e., pairs composition-temperature for vapor (y^V, T^V) and liquid (x^L, T^L) bulk phases lie either in the superheated region or the subcooled region, respectively.

6. Conclusions

In this paper a rigorous rate based model for separation in packed columns has been calibrated based on multiple experiments. As a simultaneous approach has been used, an efficient computational procedure to deal with the large number of degrees of freedom is required. Model reduction and structure exploitation have been employed, yielding a speedup by a factor 20. In addition, the number of parameters has been reduced via a sensitivity analysis. Also additional constraints to ensure feasibility of the solution have been included in the simultaneous approach in a natural way. As a result, the calibrated model obtained with the presented procedure based on five experiments, has been successfully validated on two additional experiments.

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Figures

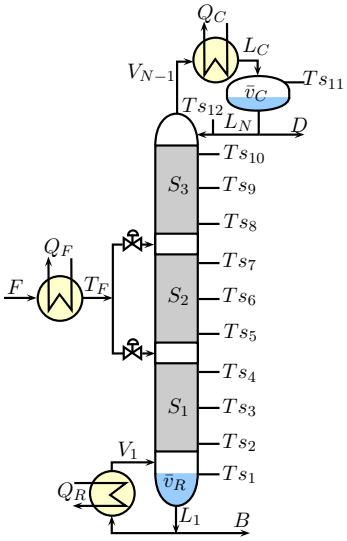

Figure 1:

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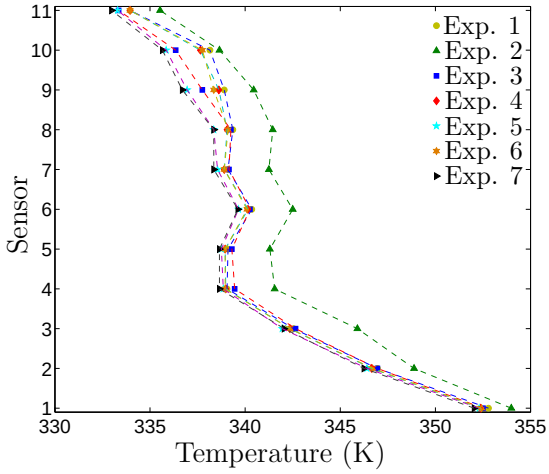

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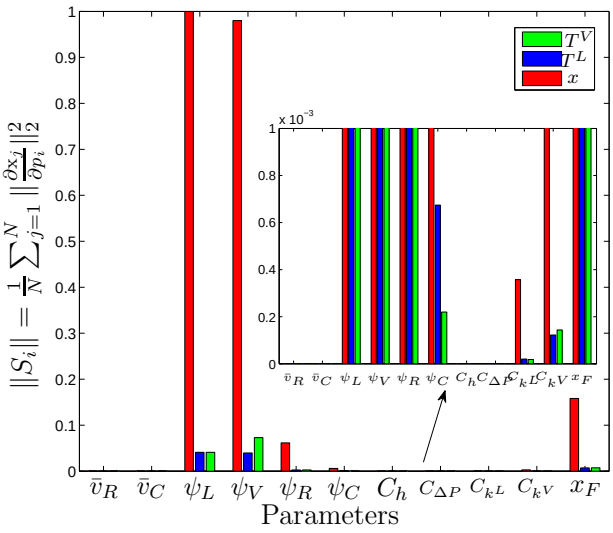

Figure 3:

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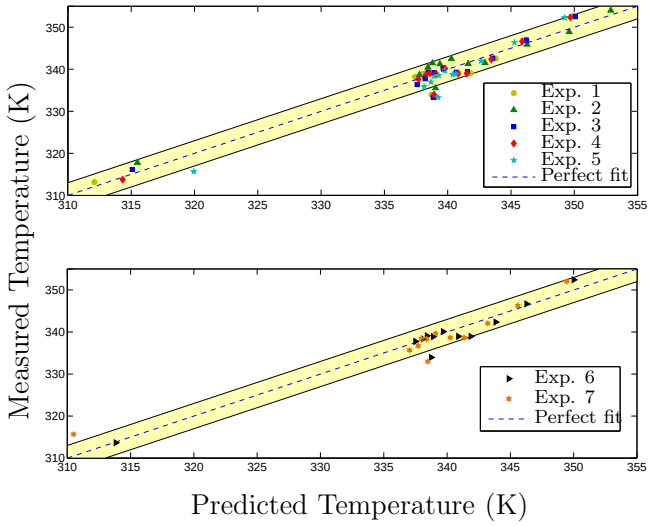

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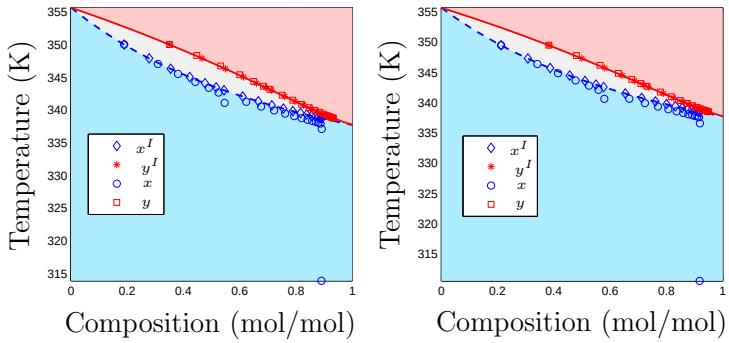

Figure 5:

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Figure captions

Figure 1: General layout of the distillation setup.

Figure 2: Experimental data from the 11 temperature sensors along the column. Temperature of the reflux, around 315 K, has not been included in the plot but is used in the estimation.

Figure 3: Norm on the normalized sensitivity of the states with respect to the parameters.

Figure 4: Results for five calibration experiments (top) and two validation experiments (bottom).

Figure 5: Boiling point diagram for the mixture of Methanol-Isopropanol with states given by the steady state solution of the RBM for experiment 6 (left) and 7 (right).

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Table 1: Parameter vector: initial (p_0) and optimised (p^*)

Parameter	Initial value	Bounds	Optimum[a]	Units
$\bar{v}_R$	5	[3 6]	-	l
$\bar{v}_C$	2	[1 3]	-	l
ϕ_L	1	[0 10]	2.3	W/K
ϕ_V	0.5	[0 10]	0.0	W/K
ϕ_R	8	[0 100]	0.0	W/K
ϕ_C	5	[0 100]	8.3	W/K
C_h	1	[0 2]	-	
C_p	1	[0 2]	-	
C_{k^L}	1	[0 5]	0.22	
C_{k^V}	1	[0 5]	2.12	
x_F	0.67	[0.4 0.7]	0.4670	mol/mol

[a]Only identifiable parameters are optimized.

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Table 2: Performance comparison between the full rate based model (FRBM) and the reduced rate based model (RORBM) when sparsity is exploited (SP) or not (NSP).

	FRBM-NSP	FRBM-SP	RORBM-NSP	RORBM-SP
Number of variables	383	383	272	272
Function evaluations	2688	161	1638	108
Execution time (s)	**17.6**	1.89	11.96	**0.883**
Memory used[a] (kB)	**1146**	4.36	578	**2.73**

[a]Only the number of kilobytes to store the Jacobian in double precision format are considered as indication of the memory usage.

Table 3: Steady state experiments used for identification and validation.

Exp	Q_R	L_N	F_{in}	T_F	T_{amb}	D	B
1	4.0	60.0	150	313.15	292.90	70	80
2	4.5	86.0	110	313.15	292.95	70	40
3	4.5	80.0	150	318.15	292.45	70	80
4	4.0	59.0	150	313.15	295.20	70	80
5	4.5	76.5	150	313.15	298.85	70	80
6	4.0	65.0	150	313.15	294.95	70	80
7	4.5	77.2	150	313.15	287.45	70	80

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