

# STATE ESTIMATION IN CHROMATOGRAPHIC SMB PROCESSES WITH LINEAR ADSORPTION ISOTHERMS

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Abstract: Chromatographic separations are widespread in the fine chemicals and pharmaceutical industries for the downstream processing of complex mixtures with similar thermodynamic properties. In recent years, continuous chromatographic processes, in particular Simulated Moving Bed (SMB) processes are increasingly applied in industry. SMB processes are characterized by a counter-current movement of the liquid and the solid phase which is achieved by switching the inlet and the outlet ports of a closed loop of chromatographic columns periodically. Recent research has focused on the model-based optimization and control of SMB processes. In a model-based approach, plant/model mismatch has to be taken into account. In this paper, the issue of state estimation in SMB processes under plant/model mismatch is addressed. A Kalman filter is designed and successfully applied. The estimation is based on only one measurement device located in the recycle stream of the process. *Copyright*© 2005 *IFAC*

Keywords: Preparative Chromatography; Simulated Moving Bed Process; State Estimation; Kalman Filter

## 1. INTRODUCTION

Preparative chromatographic separation processes play an important role in the downstream processing in the pharmaceutical and fine chemicals industries. Until now, most industrial separations are performed discontinuously, leading to low productivity and high solvent consumption. In recent years, continuous Simulated Moving Bed SMB processes are increasingly applied due to their advantages with respect to the utilization of the adsorbent and solvent consumption.

SMB processes are characterized by mixed discrete and continuous dynamics, spatially distributed state variables with steep slopes, and slow and strongly nonlinear reactions of the product concentrations to changes of the operating parameters, and therefore difficult to control. An overview of recent achievements in the optimization and control of chromatographic separations can be found in (Engell and Toumi, 2004). A two-layer control strategy for SMB processes was proposed in (Klatt *et al.*, 2000). On the upper layer, the operating parameters are optimized using a rigorous plant model. The low level control task is to keep the process on the optimal trajectory despite disturbances and plant/model mismatch by controlling the positions of the concentrations

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fronts (Wang *et al.*, 2003). On the upper layer the model parameters are adapted based on online measurements. However, the stabilization of the front positions does not guarantee the product purities in the presence of plant/model mismatch and in case of structural plant/model mismatch an additional purity control layer must be introduced (Hanisch, 2002). In (Toumi and Engell, 2004a) and (Toumi and Engell, 2004b), a nonlinear optimizing control scheme was proposed and successfully applied to a 3-zone reactive SMB process for glucose isomerization. In each switching period, the operating parameters are optimized to minimize a cost function. The product purities appear as constraints in the optimization problem. In the optimization, a rigorous model of the general rate type is used. Plant/model mismatch is taken into account by assuming an additive constant disturbance derived from the difference of the predicted and the measured purities. In addition, the model parameters are regularly updated. The internal (distributed) states of the plant are computed from the available concentration measurements by forward simulation. The prediction is based upon the assumption that the columns are uniform and that modeling errors are small. However, the columns may have different effective lengths, and the adsorbent and the catalyst (for the case of reactive chromatography) may be packed differently. In addition, the column temperatures can exhibit some variation. Therefore, as observed in the experimental validation of the controller, the product purities of a SMB plant may oscillate over a cycle of operation when the operation parameters obtained from the simulation of a plant with uniform columns are applied.

In this paper, the issue of the estimation of the internal concentration profiles in SMB processes from available measurements at the column outputs is addressed. A particular challenge results from the fact that the measurement information which is available at a real plant is rather scarce. Even when both concentrations in the product streams are measured continuously (which is not the standard in production processes), the dynamic errors of these measurements usually are too large to use them for the computation of the internal concentration profiles. We therefore estimate the concentration profiles inside all columns from only one continuous concentration measurement in the recycle loop. The goal is to apply the state estimation scheme for the computation of the initial state and for the adaptation of crucial model parameters in the online optimizing control concept.

The remainder of this paper is structured as follows: in the next section, SMB processes are introduced. Section 3 is devoted to the observation of the states in the SMB plant based upon only one

measurement device, the situation encountered at our pilot plant. Simulation results are presented in section 4. Finally, a summary and outlook for future research are given.

## 2. SMB PROCESSES

### 2.1 Process description

The SMB process is a practical implementation of a counter-current movement between the liquid and the solid phase in chromatographic separations. The process consists of chromatographic columns which are connected in series to form a closed-loop system. The counter-current motion of the solid phase with respect to the liquid phase is achieved by synchronously advancing the inlet and outlet ports of the system in the direction of the liquid flow. The columns can be divided into four different zones according to their relative position with respect to the inlet and the outlet ports as depicted in Figure 1:

- (i) Zone I between eluent and extract port: desorption of the more strongly retained component
- (ii) Zone II between extract and feed port: desorption of the less retained component
- (iii) Zone III between feed and raffinate port: adsorption of the more strongly retained component
- (iv) Zone IV between raffinate and eluent port: adsorption of the less retained component.

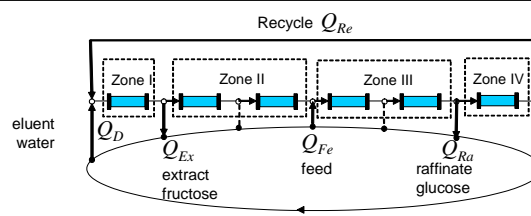


Fig. 1. Schematic diagram of a 4-zones SMB process

In this paper, the separation of fructose (A) from glucose (B) is considered (Jupke, 2004). The advantage of pure fructose is its higher sweetness and better solubility in water at room temperature. The columns are packed with an ion exchange resin adsorbent (Amberlite CR-13NA). An optimized SMB configuration with a 1/2/2/1 column distribution among the zones is considered. The concentration profiles in the plant move in the direction of the liquid flow and are then shifted by one column against the liquid flow at the end of each period  $T$ . When the concentration profiles after the switch are identical to those at the beginning of the period, stationary operation at a *cyclic* steady state is reached.

## 2.2 Modeling

Modeling of chromatographic processes has been the focus of many publications in recent years. Rigorous models of SMB plants consist of dynamic models of each column and periodic shifting of the ports. From mass balances around the inlet and outlet nodes the following expressions for the internal flow rates  $Q_I, Q_{II}, Q_{III}, Q_{IV}$  and the inlet concentrations  $c_{i,I}^{in}, c_{i,III}^{in}$  after the mixing nodes can be derived:

$$\begin{aligned} \text{Desorbent node:} \quad & Q_{IV} + Q_{De} = Q_I \\ & c_{i,IV}^{out} Q_{IV} = c_{i,I}^{in} Q_I \\ \text{Extract node:} \quad & Q_I - Q_{Ex} = Q_{II} \\ \text{Feed node:} \quad & Q_{II} + Q_{Fe} = Q_{III} \\ & c_{i,II}^{out} Q_{II} + c_{i,Fe} Q_{Fe} = c_{i,III}^{in} Q_{III} \\ \text{Raffinate node:} \quad & Q_{Ra} + Q_{IV} = Q_{III}. \\ & i = A, B \end{aligned}$$

$Q_{De}, Q_{Ex}, Q_{Fe}, Q_{Ra}$  denote the external flow rates while  $c_i^{out}$  denotes the concentration of component  $i$  leaving the respective zone.

The chromatographic columns are described accurately by the *general rate model* which accounts for all important effects of the column, i.e. mass transfer between the liquid and solid phase, pore diffusion, and axial dispersion (Gu, 1995), (Guichon *et al.*, 1994). It is assumed that the particles of the solid phase are uniform, spherical, porous (with a constant void fraction  $\epsilon_p$ ), and that the mass transfer between the particle and the surrounding layer of the bulk is in a local equilibrium. The concentration of component  $i$  is given by  $c_i$  in the liquid phase and  $q_i$  in the solid phase.  $D_{ax}$  is the axial dispersion coefficient,  $u$  the interstitial velocity,  $\epsilon_b$  the void fraction of the bulk phase,  $c_i^{eq}$  the equilibrium concentration,  $k_{l,i}$  the film mass transfer resistance, and  $D_{p,i}$  the diffusion coefficient within the particle pores. The concentration within the pores is denoted by  $c_{p,i}$ . Assuming that  $u$  and  $c_i$  are uniformly distributed over the radius, the following set of partial differential equations can be obtained from a mass balance around an infinitely small cross-section of the column:

$$\begin{aligned} \frac{\delta c_i}{\delta t} + \left( \frac{1 - \epsilon_b}{\epsilon_b} \right) \frac{3k_{l,i}}{r_p} (c_i - c_{p,i}|_{r=r_p}) \\ = D_{ax,i} \frac{\delta^2 c_i}{\delta z^2} - u \frac{\delta c_i}{\delta z} \end{aligned} \quad (1)$$

$$\begin{aligned} (1 - \epsilon_b) \frac{\delta q_i}{\delta t} + \epsilon_p \frac{\delta c_{p,i}}{\delta t} - \epsilon_p D_{p,i} \frac{1}{r^2} \frac{\delta}{\delta r} \left( r^2 \frac{\delta c_{p,i}}{\delta r} \right) \\ = 0 \end{aligned} \quad (2)$$

The initial and boundary conditions are

$$\begin{aligned} c_{i,t=0} = c_i^{in}; \quad c_{p,i,t=0} = c_{p,i}(0, r, x), \\ \left. \frac{\delta c_i}{\delta z} \right|_{z=0} = \frac{u}{D_{ax,i}} (c_i - c_i^{in}); \quad \left. \frac{\delta c_i}{\delta x} \right|_{z=L} = 0 \quad (3) \\ \left. \frac{\delta c}{\delta r} \right|_{r=0} = 0; \quad \left. \frac{\delta c_p}{\delta r} \right|_{r=r_p} = \frac{k_i}{\epsilon_p D_{p,i}} (c_i - c_{p,i,r=r_p}). \end{aligned}$$

For sugar separations at low and medium concentrations, the adsorption can be described by a linear (Henry) adsorption isotherm:

$$q_i = H_i c_{p,i}. \quad (4)$$

The extract purity  $Ex$  and raffinate purity  $Ra$  are calculated according to:

$$Ex = \frac{\int_0^T C_{Ex,A} dt}{\int_0^T (C_{Ex,A} dt + C_{Ex,B} dt)} \quad (5)$$

$$Ra = \frac{\int_0^T C_{Ra,B} dt}{\int_0^T (C_{Ra,A} dt + C_{Ra,B} dt)}. \quad (6)$$

The resulting coupled partial differential equations can be solved efficiently using the numerical approach proposed by (Gu, 1995), where the solid phase is discretized by applying orthogonal collocation and the bulk phase is discretized using the Galerkin finite element method. The finite element method is especially suitable for stiff systems as chromatographic processes which are characterized by steep gradients in the fluid phase. If the concentration of the feed, the switching period, the input and the output flows are constant and the absorption isotherm is linear, the resulting ordinary differential equation system is linear. For the observer application presented in this paper the classical SMB operation with constant flows is considered. With the chosen number of finite elements and collocation points, the six column SMB process in Figure 1 is described by 312 states. In the sequel, the states of the discretized model are denoted by  $x$  and the set of linear ODEs is referred to as  $\dot{x} = f(x)$ . In the simulations, the set of ordinary differential equations is solved using the ODE solver "dvoid".

## 3. OBSERVER DESIGN

The development of the state estimator is related to the pilot SMB-plant which is operated in our department. The plant is equipped with three concentration measurement devices which are located at the outlet ports of the extract and the raffinate flows and in the recycle stream. The concentration measurements in the product streams are not useful for dynamic state estimation because of considerable hold-ups that cause delays and back mixing. They are only used to determine the average product purities over one period (Hanisch, 2002). The measurement device in the recycle stream has small dead volumes and provides precise dynamic concentration profiles. Hence only two measurements (two concentrations at one location) are available to estimate the remaining 310 states. The recycle measurement is stationary within the physical plant. Therefore, the measurement position is shifted one column in the opposite direction to the liquid flow when the

ports are moved. The available measurement thus provides different state variables during each of the 6 switching periods over one cycle. The output matrix  $C$  is adjusted after each period accordingly.

### 3.1 Kalman Filter design

The purpose of the state estimator is to provide corrected predictions of the model states. It can be shown that the system is observable from the available measurements. A linear Kalman Filter (KF) is applied here. It is characterized by a prediction step and a correction step:

(1) Prediction:

$$\hat{x}_{k+1,k} = \hat{x}_{k,k} + \int_{t_k}^{t_{k+1}} \hat{f}(\hat{x}) dt \quad (7)$$

$$P_{k+1,k} = AP_{k,k}A^T + Q \quad (8)$$

(2) Correction:

$$K_k = P_{k,k-1}C^T(CP_{k,k-1}C^T + R)^{-1} \quad (9)$$

$$\hat{x}_{k,k} = \hat{x}_{k,k-1} + K_k(y_k - \hat{y}_{k,k-1}) \quad (10)$$

$$P_{k,k} = (I - K_kH_{k,k-1})P_{k,k-1}, \quad (11)$$

where  $P$  is the error covariance,  $K$  the Kalman gain,  $A$  the system matrix,  $Q$  the model error covariance matrix,  $R$  the measurement error covariance matrix,  $\hat{f}$  is the right hand side of the ODE system describing the process model, and  $y$  are the measurements.  $\hat{x}$  represents the estimated states. The first index denotes the actual time instance, while the second index is the time instance when the respective matrix is calculated. The error matrix  $P_{smb}$  of the SMB plant can be represented as:

$$P_{smb} = \begin{pmatrix} P_{1,1} & P_{1,2} & P_{1,3} & P_{1,4} & P_{1,5} & P_{1,6} \\ P_{2,1} & P_{2,2} & P_{2,3} & P_{2,4} & P_{2,5} & P_{2,6} \\ P_{3,1} & P_{3,2} & P_{3,3} & P_{3,4} & P_{3,5} & P_{3,6} \\ P_{4,1} & P_{4,2} & P_{4,3} & P_{4,4} & P_{4,5} & P_{4,6} \\ P_{5,1} & P_{5,2} & P_{5,3} & P_{5,4} & P_{5,5} & P_{5,6} \\ P_{6,1} & P_{6,2} & P_{6,3} & P_{6,4} & P_{6,5} & P_{6,6} \end{pmatrix}, \quad (12)$$

where for example  $P_{1,2}$  is the error covariance of the states of column 1 and the states of column 2. The evolution of the error covariance matrices  $P_{i,j}$  over time depends on the position of the respective column as well as potentially on different model parameters for each column.

As the state variables are defined relative to the port locations and not along the physical columns, the states and the state error covariances must be shifted when the ports are switched. To account for the shift, the error matrix of the SMB-process is transformed after each period  $T$  according to:

$$P_{i,j}(T^+) = P_{i+1,j+1}(T^-), \quad i, j = 1, \dots, 5 \quad (13)$$

$$P_{6,i}(T^+) = P_{1,i+1}(T^-), \quad i = 1, \dots, 5 \quad (14)$$

$$P_{i,6}(T^+) = P_{i,1}(T^-), \quad i = 1, \dots, 5 \quad (15)$$

$$P_{6,6}(T^+) = P_{1,1}(T^-). \quad (16)$$

The transformation of  $P_{smb}$  reflects that the error covariances of the physical columns remain unchanged as the ports are shifted.

The SMB-process considered here is simulated with the set of plant parameters and operating parameters given in Table 2. In the investigation of the performance of the estimator, it is assumed that the measurements are corrupted with additive white noise with a variance of 2.5% of the largest concentration value. The measurement error covariances are assumed to be diagonal with variances  $\sigma_i^2$  on the diagonal. The state noise covariance matrix  $Q$  is calculated at each time step using the so-called Linearization approach introduced by (Valapil and Georgakis, 1999). The mean values of the parameters,  $p_{nom}$ , and the covariances  $C_p$  of the model parameters are utilized to determine  $Q(t)$  according to:

$$Q(t) = J_{p,nom}(t)C_pJ_{p,nom}^T(t), \quad (17)$$

where  $J_{p,nom} = \left. \frac{\delta \hat{f}}{\delta p} \right|_{nom}$ .

In this approach it is assumed that  $\hat{f}$  is linear in its parameters  $p$ . The SMB example process considered here is only slightly nonlinear in the parameters so this assumption is justified. (Valapil and Georgakis, 1999) demonstrated that this tuning approach is efficient. In the simulation model errors introduced by perturbed Henry coefficients are investigated. The covariance matrix  $C_P$  of the Henry coefficients is calculated by:

$$C_P = \begin{pmatrix} \sigma_{H_A}^2 & \sigma_{H_A}\sigma_{H_B} \\ \sigma_{H_A}\sigma_{H_B} & \sigma_{H_B}^2 \end{pmatrix}. \quad (18)$$

For the simulations the standard deviations  $\sigma_{H_A}$  and  $\sigma_{H_B}$  are assumed to be 0.1126 and 0.0837, respectively. The initial error covariance matrix  $P_0$  is considered as a tuning parameter of the estimator.  $P_0 = 100 \cdot I$  was found to give good results.

## 4. RESULTS

Figures 2 and 3 demonstrate the convergence of the estimator described above for a perfectly known model but an initial error of -100% from the actual plant operated at CSS which is equivalent to assuming an empty plant while the real plant is already at the cyclic steady state. The observed state reaches the real state within 6 switching periods. The assumption that the concentration profile is completely unknown initially is overly pessimistic. Instead, the measured concentrations over one full cycle (the so-called Assembled Elution Profile, AEP) provide a good approximation of the internal profiles at the beginning of a cycle and can be taken as the initial state for the estimator. In the scheme used in

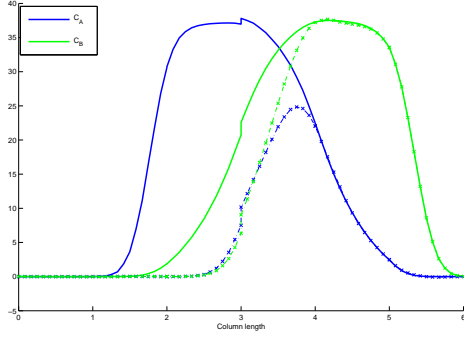


Fig. 2. True and estimated elution profiles at  $t=3T$ , concentration measurement at the outlet of column 4; initial error -100%; thick lines: reference plant, marked dashed lines: estimation

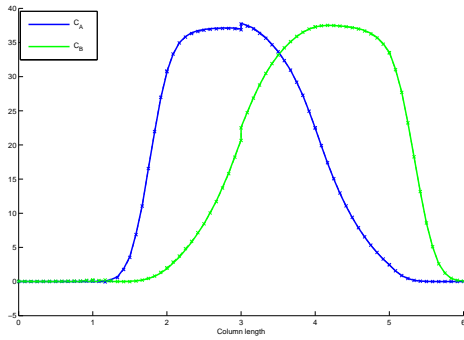


Fig. 3. True and estimated elution profiles at  $t=6T$ , concentration measurement at the outlet of column 1; initial error -100%; thick lines: reference plant, marked dashed lines: estimation

(Toumi and Engell, 2004a,b), the internal states are estimated by simulating the plant model starting from the AEP as the initial state. This scheme is thus equivalent to the Kalman filter without the correction step.

In a second scenario, a virtual plant with parameters that are different from the model used in the estimator operated at the CSS with the simulation initialized by the AEP is considered. The evolution of a corrected and an uncorrected model is considered over 6 periods since a new AEP is available after each cycle and can then be used for reinitializing the model states. A model error is introduced by increasing the value of both Henry coefficients in the model by 30% ( $H_{i,model} = 1.3 \cdot H_{i,ref}$ ), hence the adsorption is overestimated in the model. An error of the Henry coefficients was chosen because the adsorption isotherm is of crucial importance for the behaviour of the process. The measurement device is positioned at the outlet of column 6 for the first period and then moves one column to the left for each period.

The initial profile and the profiles at the end of periods 4 and 6 are shown in figures 4, 5, 6 and the achieved purities for each period and average purities are given in Table 1. During the first period, the observer does not correct the model states since the outlet concentrations of the reference plant and the model are both close to zero. The superior performance of the observer is demonstrated from the second period on. The corrected states remain close to the profile of the actual plant but exhibit a cyclic behaviour depending on the position of the recycle measurement, see Table 1. Especially for periods 4 - 6, the product purity forecast of the observer is much closer to the value of the "real" plant than the forecast of the uncorrected model. The average product purity computed by the observer is within 1.4% of the true value while the use of an uncorrected model leads to a bias of almost 4% for the extract purity.

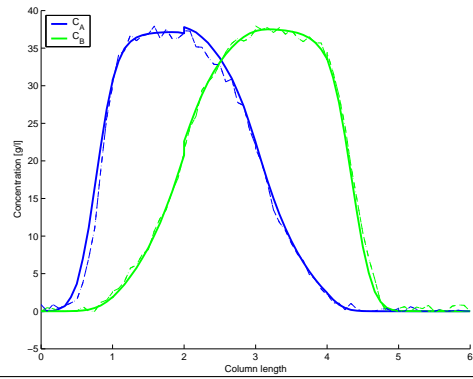


Fig. 4. Elution profiles at  $t=0$ , concentration measurement at the outlet of column 6; model error:  $H_i = 1.3 \cdot H_{i,ref}$ ; thick lines: reference plant, dashed lines: estimation, dash-dots: open-loop observer

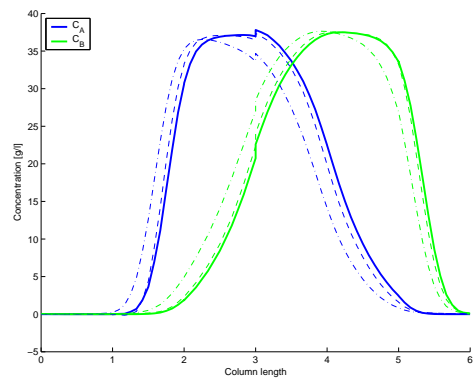


Fig. 5. Elution profiles at  $t=3T$ , concentration measurement at the outlet of column 4; model error:  $H_i = 1.3 \cdot H_{i,ref}$ ; thick lines: reference plant, dashed lines: estimation, dash-dots: open-loop observer

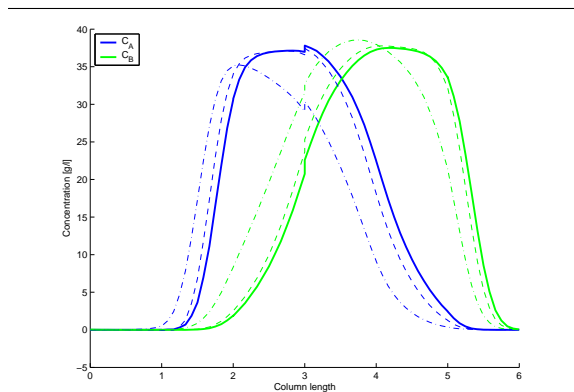


Fig. 6. Elution profiles at  $t=6T$ , concentration measurement at the outlet of column 1; model error:  $H_i = 1.3 \cdot H_{i,ref}$ ; thick lines: reference plant, dashed lines: estimation, dash-dots: open-loop observer

Table 1. Comparison of the predicted purities for simulation and state estimation at the cyclic steady state; reference purities: 96.8% (extract) and 97.0% (raffinate)

Period	$Ex_{est}$	$Ra_{est}$	$Ex_{sim}$	$Ra_{sim}$
1	94.10%	96.81%	94.10%	96.81%
2	93.90%	96.92%	94.10%	97.09%
3	95.25%	97.20%	93.66%	97.43%
4	96.20%	97.52%	93.03%	97.83%
5	96.59%	97.83%	92.21%	98.23%
6	96.55%	98.04%	91.21%	98.59%
average	95.43%	97.39%	93.05%	97.60%

## 5. CONCLUSION

The performance of the Kalman Filter developed for Simulated Moving Bed processes is satisfactory, given the difficult task to observe the plant based on only one measurement device. In combination with the AEP, the forecast of the product purities is improved compared to a pure simulation. The application of the observer for SMB processes presented in this paper is especially suitable for processes with high purity requirements, which require a precise purity forecast for optimal operation. It is expected that the performance of the model predictive control approach to SMB processes as described in (Toumi and Engell, 2004a,b) can be improved by using a state estimation scheme rather than uncorrected simulations. In future research, the observation of SMB processes with nonlinear adsorption isotherms will be studied.

## 6. APPENDIX

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Table 2. Plant and operating parameters

$H_A = 0.375$	$Q_{De} = 8.85 \frac{ml}{min}$
$H_B = 0.279$	$Q_{Ex} = 6.60 \frac{ml}{min}$
$L = 57.8cm$	$Q_{Fe} = 0.92 \frac{ml}{min}$
$D = 2.6cm$	$Q_{Ra} = 3.17 \frac{ml}{min}$
$\epsilon_p = 0.01$	$Q_{Re} = 11.63 \frac{ml}{min}$
$\epsilon_b = 0.376$	$c_{A,Fe} = 50 \frac{g}{l}$
$r_p = 1.3cm$	$c_{B,Fe} = 50 \frac{g}{l}$
$k_{l,A} = 8.21 \cdot 10^{-5} \frac{cm}{s}$	$T = 12.5min$
$k_{l,B} = 5.08 \cdot 10^{-5} \frac{cm}{s}$	$\eta = 5.8 \cdot 10^{-3} \frac{g}{cm \cdot s}$
$D_p = 0.001 \frac{cm^2}{s}$	$\rho = 1 \frac{g}{cm^3}$

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