

Online Optimization Based Feedback Control of Simulated Moving Bed Chromatographic Units

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The Simulated Moving Bed (SMB) is a continuous multi-column chromatographic process that has become an attractive technology for complex separation tasks that are regularly encountered in the areas of pharmaceuticals, fine chemicals and biotechnology. Even though economic optimality of the SMB process has attracted significant amount of attention, operation of the SMB units at their optimal operating conditions is still an open issue due to the absence of proper feedback control schemes. Recently, we have developed an online optimization based feedback control scheme that can deliver the full economic power of the SMB technology despite disturbances and uncertainties in the system.^{1,2,3} This paper focuses on the implementation of the control concept to SMBs operating under overloaded chromatographic conditions that are characterized by nonlinear competitive adsorption isotherms. It is shown that despite the overloaded chromatographic conditions, the process can be controlled and optimized based on the linear adsorption isotherm information only. The performance and robustness of the control scheme is assessed under simulated challenging operating conditions.

Key words:

Simulated moving bed, nonlinear chromatography, online optimization, automatic control, repetitive model predictive control

Introduction

True moving bed (TMB) is a continuous countercurrent chromatographic process that can be used to separate chemical mixtures into two fractions. The separation principle is the different affinity of the components in the mixture to the solid phase in the solid/liquid countercurrent contact.

Simulated moving bed chromatography is a technical approximation of the TMB process that overcomes the problems connected to countercurrent continuous flow of the solid phase. The SMB plants comprise a series of looped fixed chromatographic columns where solvent is circulating in one direction. The continuous countercurrent solid flow is approximated by moving the solid phase over a finite length at discrete times, e.g. synchronous switches of the positions of inlet (feed, desorbent) and outlet (raffinate, extract) streams one column position in the direction of the liquid flow (see figure 1). Inlet and outlet streams divide the SMB units into four sections having different

functionalities. The mixture to be separated enters the unit at the feed-port between sections II and III. Given that switching interval and the liquid flow

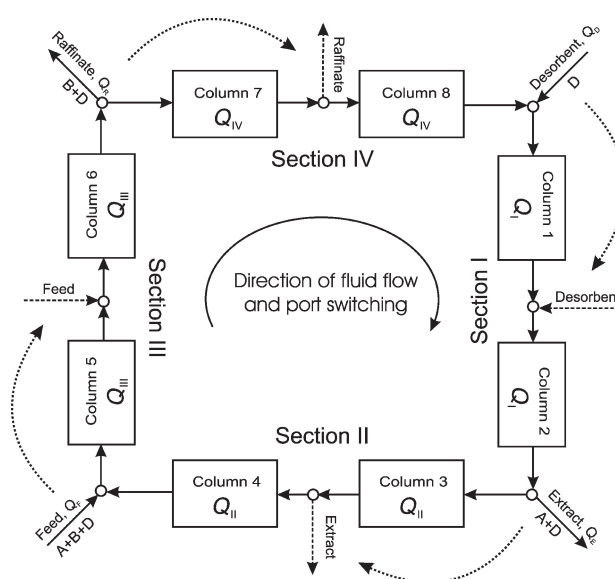


Fig. 1 – Scheme of a Simulated Moving Bed (SMB) unit. indicates the inlet/outlet positions after the first switch

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rates in each section are chosen properly, the separation takes place in two central sections, where the least retained component B is eluted from section III before the port switching and collected at raffinate outlet, i.e., located between sections III and IV, whereas the more retained component A remains adsorbed in the solid phase until it is carried to the section I by the port switch, where it is eluted with fresh solvent and collected in the extract outlet, i.e., located between sections I and II. The liquid and solid phases are regenerated at section IV and I, respectively. The stationary regime of the SMB process is a cyclic steady state, in which the process variables undergo a time varying transient state. The transient dynamics repeat themselves over the period of the process. The process period can be defined as the time between two successive port switches, i.e. t^* , with the assumption that all the columns constituting the unit are identical. On the other hand, because it is practically difficult to have identical columns, the global period of the process is defined as a complete cycle, i.e. $n_{\text{col}} \times t^*$ where n_{col} is the number of columns in the SMB loop. We refer to the available literature for more detailed description of the SMB process.⁴

SMB technology has been regarded as an attractive technology not only because of its advantages, e.g., high productivity per unit mass of stationary phase and low solvent consumption, over conventional separation techniques, e.g., batch chromatography, but also because of its ability to deliver these advantages, both, in the early development and the production stages. Chiral separations, e.g., single enantiomer drug development, constitute the favorite application group for the SMB chromatography, because they are characterized by low selectivities and the cost in chiral separations is dominated by the solvent consumption and expensive chiral stationary phases. This is reflected by the similar trends in the popularity of the SMB technology and the significance of the chiral drugs in the global drug market, e.g., single enantiomer drugs took up forty percent of the global drug sales in 2 000.⁵

SMB technology enjoys the gained experiences and well established modelling concepts in the area of conventional chromatographic techniques that eased the development of dynamic SMB modelling concepts. Optimization tools with different level of complexity^{6,7} resulted in new SMB schemes and operating modes such as VARICOL,⁸ PowerFeed⁹ and ModiCon,¹⁰ which allow for further improvements in the efficiency of the SMB units.

There are two critical issues concerning the optimal operation of SMB units. First of all, regardless of their complexity, the performance of the optimization algorithms is limited by the accuracy of

the available physical data on the system. SMB units are operating under overloaded conditions, which is the main reason for their high productivity, and the description of the separation process requires the knowledge of the multicomponent adsorption isotherm characterizing the separation. Unfortunately, precise measurement of the competitive isotherm parameters is a rather difficult task. Especially in the case of enantiomer separations it may not be possible at all, because of a lack of pure enantiomers or the shortage of racemate, i.e., a common problem during the development stage of chiral drugs.

Even, if precise physical data of the system are available, the optimal operating conditions depend on the operating parameters such as feed concentration, and the physical parameters of the system, e.g., adsorption isotherm and column properties, which are subject to change. Therefore, re-characterization and re-optimization of the system are necessary.

The second critical issue is the sensitivity of the SMB units to disturbances, e.g., aging of the stationary phase, feed composition or temperature changes, and to uncertainties in the system, e.g., column characteristics or the adsorption isotherms, which may lead to either sub-optimal operation conditions or off-spec production.⁴ Therefore, it is common practice to operate the SMB units far from the optimal operating conditions to guarantee a certain level of robustness during the operation.

Automatic control of the SMB units has the potential to deliver the full economic potential of the technology. On the other hand automatic control of SMB process has its own challenges. SMB involves complex hybrid dynamics with nonlinearities and delays. Several SMB control approaches have been proposed.^{11–16} The main drawback of these approaches is the need for accurate physical data about the system. This work proposes a feedback control strategy that is based on the linear adsorption isotherm information only regardless of the type of isotherm characterizing the mixture to be separated. This is a key improvement because contrary to competitive adsorption isotherm parameters, the linear adsorption isotherm parameters, i.e. Henry's constants, can be determined easily and precisely. The control problem is constructed along the lines of repetitive model predictive control (RMPC), i.e., a new model periodic control (MPC) method for periodic control problems,^{17,18} and it integrates the online optimization and control of the SMB process to address specific difficulties mentioned above.

Modelling and simulation of SMB process

The modelling and simulation techniques for the SMB process have been well established thanks to a significant level of experience gained in the area of batch chromatography. Here, we give a short description of the modelling concept and refer to widely available literature for detailed modelling principles and their experimental verifications.^{19–22}

There are a number of available models that describe the single columns dynamics and their level of complexity varies depending on the physical phenomena they include.²² The mathematical model of the SMB process is obtained by interconnecting the single column models with the proper initial and boundary conditions. In this work, we make use of the equilibrium dispersive model (EDM) which has been widely exploited for the design and optimization of chromatographic separations, and regarded as a good compromise between model accuracy and computational efficiency.^{22,23} EDM enforces the local equilibrium of the mobile and the stationary phases and lumps the effects of finite mass transfer rate and the axial dispersion in an apparent axial dispersion coefficient, i.e., denoted by $D_{ap,i}$ below.

$$\begin{aligned} \varepsilon_h \frac{\partial c_{i,h}}{\partial t} + (1 - \varepsilon_h) \frac{\partial q_{i,h}^*}{\partial t} + \frac{Q_h}{A_{cr}} \frac{\partial c_{i,h}}{\partial z} = \\ = \varepsilon_h D_{ap,i} \frac{\partial^2 c_{i,h}}{\partial z^2} \end{aligned} \quad (1)$$

In the equation above, Q_h and ε_h are the volumetric flow rate and the total packing porosity in the h -th column, respectively. The adsorption isotherm relates the liquid and solid phase concentrations of component i denoted by c_i and q_i^* in the equation above, respectively.

$$q_{i,h}^* = f(c_{i,h}) \quad (i = A, B) \quad (2)$$

A number of isotherm models for liquid-solid equilibrium are available in the literature.^{22,24} Linear and nonlinear competitive Langmuir isotherms are of main interest in this work.

Linear isotherm:

$$q_{i,h}^* = H_i c_{i,h} \quad (i = A, B) \quad (3)$$

where H_i is the Henry's constant of component i .

Non-linear isotherm:

Competitive Langmuir isotherm

$$q_{i,h}^* = \frac{H_i c_{i,h}}{1 + K_A c_{A,h} + K_B c_{B,h}} \quad (i = A, B) \quad (4)$$

where K_i is the equilibrium constant of the i -th component. Note that the linear isotherm can be considered as a special case of the Langmuir isotherm, because the Langmuir isotherm converges to the linear isotherm when the concentration of species in the liquid phase is very small, i.e., when $c_T^F = c_A^F + c_B^F$ approaches zero.

In this work, the binary mixture of the enantiomers of the antitussive agent guaifenesin is separated, i.e., the more retained enantiomer (*S*)-(+)-guaifenesin and the less retained enantiomer (*R*)-(–)-guaifenesin. The equilibrium behavior of the mixture is characterized by the binary Langmuir adsorption isotherm (given by Eq. 4). The isotherm parameters are taken from the available literature²⁵ and are given in Table 1. We consider an eight column ($h = 1, \dots, 8$), four section ($j = I, \dots, IV$) closed-loop SMB unit to separate the racemate. The columns are distributed to the sections as 2-2-2-2. The geometrical and physical specifications of the plant are given in Table 1. The columns constituting the SMB unit are assumed to be initially saturated with mobile phase in equilibrium with the stationary phase, and therefore zero concentrations of both components along all the columns are enforced with the following initial conditions:

$$c_{i,h} = 0 \quad \forall z, \quad t = 0 \quad \text{for } h = 1, \dots, 8 \quad (5)$$

The boundary conditions at the inlet and outlet of the columns are as follows:

$$c_{i,h}^{\text{in}} = c_{i,h} \Big|_{z=0^+} \quad \forall t \quad \text{for } h = 1, \dots, 8 \quad (6)$$

$$\frac{\partial c_{i,h}}{\partial z} \Big|_{z=L} = 0 \quad \forall t \quad \text{for } h = 1, \dots, 8 \quad (7)$$

Table 1 – Physical parameters for the SMB unit and the separation system

Quantities	Value
number of columns, N_c	8
column distribution	2/2/2/2
column diameter, d_c/cm	1
column length L/cm	10
nominal porosity, ε_h	0.7
switch time, t^*/s	400
number of theoretical plates per column, $D_{ap,i}$	100
Henry's constants	$H_A = 3.5, H_B = 1.4$
equilibrium constants	$K_A = 0.0550, K_B = 0.0135$

where $c_{i,h}^{\text{in}}$ is the mass concentration of component i fed into the column h . The material balances at each inlet/outlet position complete the mathematical description of the process.

$$Q_1 = Q_8 + Q_1^{\text{I/O}} \quad (8)$$

$$Q_1 c_{i,1}^{\text{in}} = Q_8 c_{i,8}^{\text{out}} + Q_1^{\text{I/O}} c_{i,1}^{\text{I/O}} \quad (9)$$

$$Q_{h+1} = Q_h + Q_{h+1}^{\text{I/O}} \quad \text{for } h=1, \dots, 7 \quad (10)$$

$$Q_{h+1} c_{i,h+1}^{\text{in}} = Q_h c_{i,h}^{\text{out}} + Q_{h+1}^{\text{I/O}} c_{i,h+1}^{\text{I/O}} \quad \text{for } h=1, \dots, 7 \quad (11)$$

where $c_{i,h}^{\text{out}}$ is the mass concentration of component i at the outlet of the column h . $Q_h^{\text{I/O}}$ identifies the flow rate of the inlet or the outlet stream entering or leaving the SMB loop just before column h and $c_{i,h}^{\text{I/O}}$ is the mass concentration of component i in the corresponding stream. Note that the volumetric flow rates in the columns belonging to the same section, are same, e.g., $Q_1 = Q_2 = Q_{\text{I}}$, $Q_3 = Q_4 = Q_{\text{II}}$, $Q_5 = Q_6 = Q_{\text{III}}$, $Q_7 = Q_8 = Q_{\text{IV}}$ for the SMB configuration given in Fig.1.

The SMB process has a hybrid nature and the modelling concept should describe, both, continuous and discrete (switching mechanism of the inlet outlet positions) parts of its dynamics. Therefore the switching operation is considered explicitly by shifting the node balances. For instance the node balances applicable to the SMB configuration given in Fig. 1 during the first and the second switch intervals are defined in Table 2. Here we make use of a finite difference approximation to transform the PDEs into a system of ODEs. The boundary conditions and the node balances are substituted into the resultant ODE systems.

Table 2 – Flow rates of the inlet/outlet streams entering/leaving the SMB loop and the mass concentration of component i in the corresponding stream at successive switch periods

h	$0 : t^*$		$t^* : 2t^*$	
	$Q_h^{\text{I/O}}$	$c_{i,h}^{\text{I/O}}$	$Q_h^{\text{I/O}}$	$c_{i,h}^{\text{I/O}}$
1	Q_D	c_i^D	0	0
2	0	0	Q_D	c_i^D
3	$-Q_E$	$c_{i,2}^{\text{out}}$	0	0
4	0	0	$-Q_E$	$c_{i,3}^{\text{out}}$
5	Q_F	c_i^F	0	0
6	0	0	Q_F	c_i^F
7	$-Q_R$	$c_{i,6}^{\text{out}}$	0	0
8	0	0	$-Q_R$	$c_{i,7}^{\text{out}}$

Replacement of the constant apparent dispersion term by numerical dispersion is a widely used technique to obtain a computationally efficient solution of the equilibrium dispersive model.²² Concerning the SMB units, the equivalence of numerical dispersion and apparent axial dispersion term is only an approximation, because SMB units are operating under overloaded conditions that require nonlinear adsorption isotherms to describe the phase equilibrium, and the apparent dispersion coefficient is concentration dependent for nonlinear chromatography. On the other hand, it has been shown that this approximation leads to satisfactory results also in nonlinear chromatographic applications even with a small number of stages, i.e., $N_{p,i} < 100$.^{22,23,26,27}

The process is simulated by integrating the system of ODEs of the SMB model in time by a commercial stiff integrator. Note that each inlet/outlet port configuration is described with a different set of ODEs, because the input/output switching mechanism is explicitly imposed on the node balances substituted into the ODEs. For the sake of clarity, the simulated SMB plant will be referred to as SMB plant throughout the work.

Online optimization based control scheme

The proposed control concept is based on online optimization of the process. It makes use of an explicit model of the system in order to predict and optimize the future evolution of the process along the lines of the desired economic objective (see Fig. 2 for the scheme of the control concept). The internal flow rates in the four sections, i.e., $Q_1, \dots, Q_{\text{IV}}$, and the concentration levels in the two outlet streams, i.e., $c_A^R, c_B^R, c_A^E, c_B^E$, are used as the manipulated and the measured variables, respectively. It is worth noting at this point that it is hard to access the internal flow rates directly, therefore it is com-

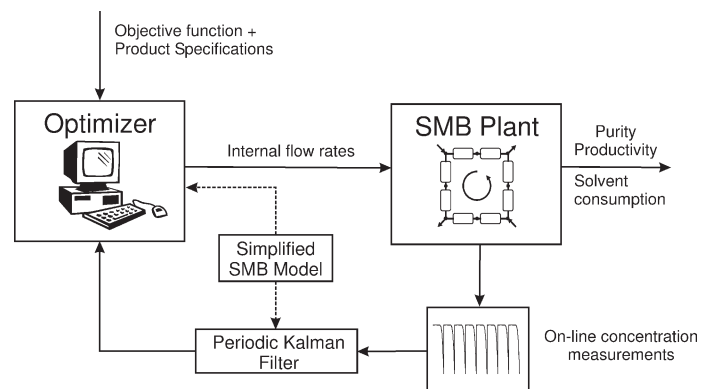


Fig. 2 – Scheme of the online optimization based automatic control concept

mon practice to control the sectional flow rates by acting on one internal and three external flow rates, e.g. by acting on Q_I , Q_E , Q_F and Q_R . Because the RMPC formulation requires a predefined period of the process, the cycle time, i.e., $n_{\text{col}} \times t^*$, is fixed a priori and the switch time, i.e. t^* , is not used as a manipulated variable. A periodic Kalman filter provides a recursive correction for the model errors by combining the model estimation and the available measurements in an optimal sense.³

The SMB model is one of the most critical component of the control scheme. It should capture the most significant dynamics of the process, but at the same time it should be simple enough to allow for online computations. Note that detailed dynamic models of SMB would lead to nonlinear programming problems which are computationally demanding. Therefore, a time-varying linear model of the SMB process has been obtained based on linearization of the dynamic model introduced in the section of Modelling and simulation of SMB process. We give a brief explanation of the modelling procedure and refer to the available literature for the detailed description.^{1,3}

The SMB process does not have a steady state in which all the process variables are constant in time. For instance, the concentration profiles in the SMB unit undergo a transient within a cycle. On the other hand, the concentration profiles are periodinvariant, i.e., the transient dynamics are the same cycle to cycle. Therefore, we have defined N time instances ($N = 64$ in this case), i.e., sample points, within the duration of a cycle and extracted N different internal profiles corresponding to the defined sample points. The cyclic steady state profiles are obtained by using the dynamic modelling concept with EDM described in the previous section of SMB process. It is important to note that the whole control concept is based on the idea that the nonlinear adsorption isotherm parameters of the system under consideration are not known, and the only information available to the controller are the linear adsorption isotherm parameters, i.e., the only physical data entering the system of ODEs of SMB are Henry's constants ($H_A = 3.5$, $H_B = 1.4$) and the nominal column porosity ($\varepsilon_h = 0.7$). Therefore, the reference steady state profiles are generated for an operation under linear chromatographic conditions. The nonlinear terms of the ODE systems, i.e., the convective terms containing the product of concentrations and flow rates, are linearized at each sample point using the corresponding cyclic steady state concentration values and the flow rates in the four sections. This procedure leads to the linear time-varying state-space SMB model to be used in the control algorithm.

$$x_k(n+1) = A(n)x_k(n) + B(n)u_k(n)$$

$$y_k(n) = C(n)x_k(n) \text{ for } n = 0, \dots, N-1 \quad (12)$$

The transition from one cycle to the next one is written as:

$$x_{k+1}(0) = x_k(N) \quad (13)$$

where, k is the cycle index and n is the time index running within the cycle index. x and u are the state and input vectors, respectively. The output vector y comprises the concentration levels at both outlets, i.e., $c_A^R, c_B^R, c_A^E, c_B^E$. All variables are defined in terms of deviation variables. For instance, the state vector comprising the internal concentration values along the unit is defined as $x(n) = c(n) - c^{\text{ref}}(n)$ and similarly the manipulated variable vector is defined as $u(n) = Q(n) - Q^{\text{ref}}(n)$. Eq. (13) implies that the space composition profiles at the end of the previous cycle, i.e., k , are used as initial conditions for the next cycle, i.e., $k+1$.

The obtained time-varying linear model of the SMB process constitutes the basis for the formulation of the control problem along the lines of RMPC. The RMPC formulation is based on the assumption that possible model prediction errors and/or effect of disturbances on the plant output are likely to repeat due to the periodic nature of the process, and therefore the information from the past cycles can be used to correct for the model errors in the future cycles. We refer to the available literature for the RMPC formulation and implementation details.^{3,18}

Optimal operation of the SMB plants can be formulated as maximizing the throughput, i.e., feed input, and minimizing the solvent consumption given that the plant layout and the switch time, i.e. t^* , are predefined. The economical objective should include constraints on the product specifications as well as on the operating conditions.

Note that raffinate and extract purities can be calculated directly from the model outputs (Eqs. 12–13) and the corresponding outlet flow rates, i.e. $Q_E = Q_I - Q_{II}$ and $Q_R = Q_{III} - Q_{IV}$. In this work, the required product specifications are enforced by constraining the average purities over the prediction horizon, i.e. 2 cycles in our case, with a lower bound.

$$P_E^{\text{ave}} \geq P_E^{\text{min}} - s_1 \quad (14)$$

$$P_R^{\text{ave}} \geq P_R^{\text{min}} - s_2 \quad (15)$$

$$s_i > 0 \quad (16)$$

The average purity expressions are nonlinear in terms of flow rates and output concentrations.

Therefore, they are successively linearized and included in the optimization problem as linear inequality constraints. The slack variables, i.e., s_1 and s_2 , are introduced to avoid infeasibility problems during the online application. Excessive usage of slack variables is avoided by penalizing them in the cost function (Eq. 18).

The operating constraints due to hardware limitations such as maximum allowable flow rate, e.g., related to pressure drop limitations for the columns or pumps, are also considered explicitly in the optimization problem.

$$0 \leq Q_j \leq Q_{\max} \quad \text{for } j = \text{I}, \dots, \text{IV} \quad (17)$$

Here, the objective is defined as minimizing the cumulative solvent consumption and maximizing the cumulative throughput over the prediction horizon, i.e., $n_c = 1$ cycle.

$$\min_{Q^{n_c, s}} [\lambda_D Q_D^{n_c} - \lambda_F Q_F^{n_c} + \lambda_1 s_1 + \lambda_2 s_2] \quad (18)$$

In the equation above, $Q_D^{n_c}$ and $Q_F^{n_c}$ are the cumulative solvent consumption and throughput over the control horizon, respectively; λ_D and λ_F are the weights of the corresponding terms, e.g., $\lambda_D = 4$ and $\lambda_F = 20$ in our case. Q^{n_c} comprises the manipulated variables, i.e., internal flow rates, for the whole control horizon. λ_1, λ_2 are the weights of the corresponding slack variables in the cost function which are kept large in order to punish the excessive use of the slack variables, e.g., $\lambda_1 = \lambda_2 = 100$ in our case.

The linear cost function (Eq. 18) together with the linear constraints (Eqs. 14–17) constitutes a Linear Program (LP). The online solution of the constructed optimization problem provides the optimal flow rate sequence which is implemented according to a receding horizon strategy, i.e., the element corresponding to the current time n is implemented on the plant and the remaining elements are discarded. As new information becomes available with the new measurements collected from the plant, a new optimization problem is solved at time $n+1$ based on the updated state of the plant. Formulation and implementation details of the optimization problem are given elsewhere.³

Case study: controller performance

As mentioned in the previous sections, the main challenges concerning the optimal operation of SMB units arise from the difficulties to characterize accurately the system involving nonlinear adsorption isotherm parameters and single column packing characteristics as well as from the mechani-

cal and chemical changes that the system undergoes during the operation. This necessitates a control scheme that is robust under extreme system uncertainties. In this case study, it is assumed that the column to column variations are negligible, therefore identical porosity is assigned for all the columns in the SMB loop, i.e., $\varepsilon_h = 0.7$ for $h = 1, \dots, 8$. The performance of the controller for a system with varying packing characteristics is given elsewhere.²⁸ Here, we show the performance and the robustness of the controller under extreme plant/model mismatch.

The SMB unit is operated under overloaded chromatographic conditions and the mixture behavior is characterized by the nonlinear binary Langmuir adsorption isotherm (the isotherm parameters taken from literature are listed in Table 1). On the other hand the controller is synthesized based on only the linear adsorption parameters, i.e., $H_A = 3.5$, $H_B = 1.4$. This constitutes the major plant/model mismatch for the case study.

In addition it is assumed that the actual system parameters are different from the ones obtained from the literature, specifically, that the Henry's constants characterizing the behavior of the mixture at low concentrations are actually $H_A = 3.15$ and $H_B = 1.33$ instead of $H_A = 3.5$ and $H_B = 1.4$. This means that the nonlinear isotherm parameters used for simulating the SMB plant are $H_A = 3.15$, $H_B = 1.33$, $K_A = 0.0550$ and $K_B = 0.0135$, whereas the linear isotherm parameters used for synthesizing the controller are $H_A = 3.5$ and $H_B = 1.4$. This corresponds to an error of 10 and 5 % in the linear adsorption parameters used within the control scheme. Note also that this implies that the plant operates with a selectivity $S = H_A/H_B = 2.37$, i.e. 5 % less than the nominal one. This constitutes the second major plant/model mismatch within the considered scenario.

The mixture to be separated has a total concentration of 10 g l^{-1} , i.e., $c_A^F = 5 \text{ g l}^{-1}$ and $c_B^F = 5 \text{ g l}^{-1}$. The required product specifications are defined as 99 % for both outlets, i.e., $P_E^{\min} = P_R^{\min} = 99 \%$. The plant is operated open loop, i.e., without the controller, for a complete cycle in order to initialize the control algorithm. The startup operating conditions are the same as the ones used for the linearization of the system of ODEs of the SMB model in section 3, i.e., $Q_I^{\text{ref}} = 2.084 \text{ ml min}^{-1}$, $Q_{II}^{\text{ref}} = 1.284 \text{ ml min}^{-1}$, $Q_{III}^{\text{ref}} = 2.094 \text{ ml min}^{-1}$, $Q_{IV}^{\text{ref}} = 1.308 \text{ ml min}^{-1}$. It is worth noting that zero-mean white noise (with a standard deviation of 2 % of the measured concentration values in this case) is added to the output measurements in order to make the simulation more realistic.

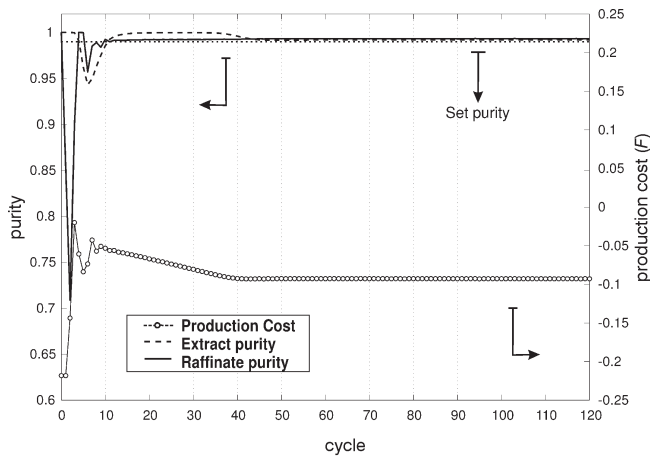


Fig. 3 – Average output purities for the controlled plant over cycles and the production cost (F)

Figure 3 gives the evolution of the average outlet purities over cycles for the controlled SMB plant. It can be observed that the purity specifications for extract and raffinate outlets are fulfilled within eleven and ten cycles, respectively. Figure 3 illustrates also the evolution of the production cost throughout the operation. We define the production cost (F) along the lines of the cost function of the

optimization problem (Eq. 18) but omit the contribution of the slack variables.

$$F = \lambda_D Q_D^{\text{ave}} - \lambda_F Q_F^{\text{ave}} \quad (19)$$

In the equation above Q_D^{ave} and Q_F^{ave} are the average solvent consumption and throughput, i.e., feed flow rate, throughout a cycle, respectively. λ_D and λ_F are constants representing the prices of the solvent and of the product, respectively, and they are the same as used in the optimization problem, i.e., $\lambda_D = 4$ and $\lambda_F = 20$.

One can see from figure 3 that the production cost is increasing at the beginning of the operation because the top priority of the controller is to fulfill the purity requirements. After the purities are as required, the economics of the operation is optimized. The steady state operating conditions are reached and the minimum production cost is achieved after forty cycles. The sectional flow rates implemented throughout the operation are given in figure 4.

The controller action and its performance can be interpreted in a systematic way with the help of the “Triangle theory” that provides a detailed understanding of how operating parameters affect the

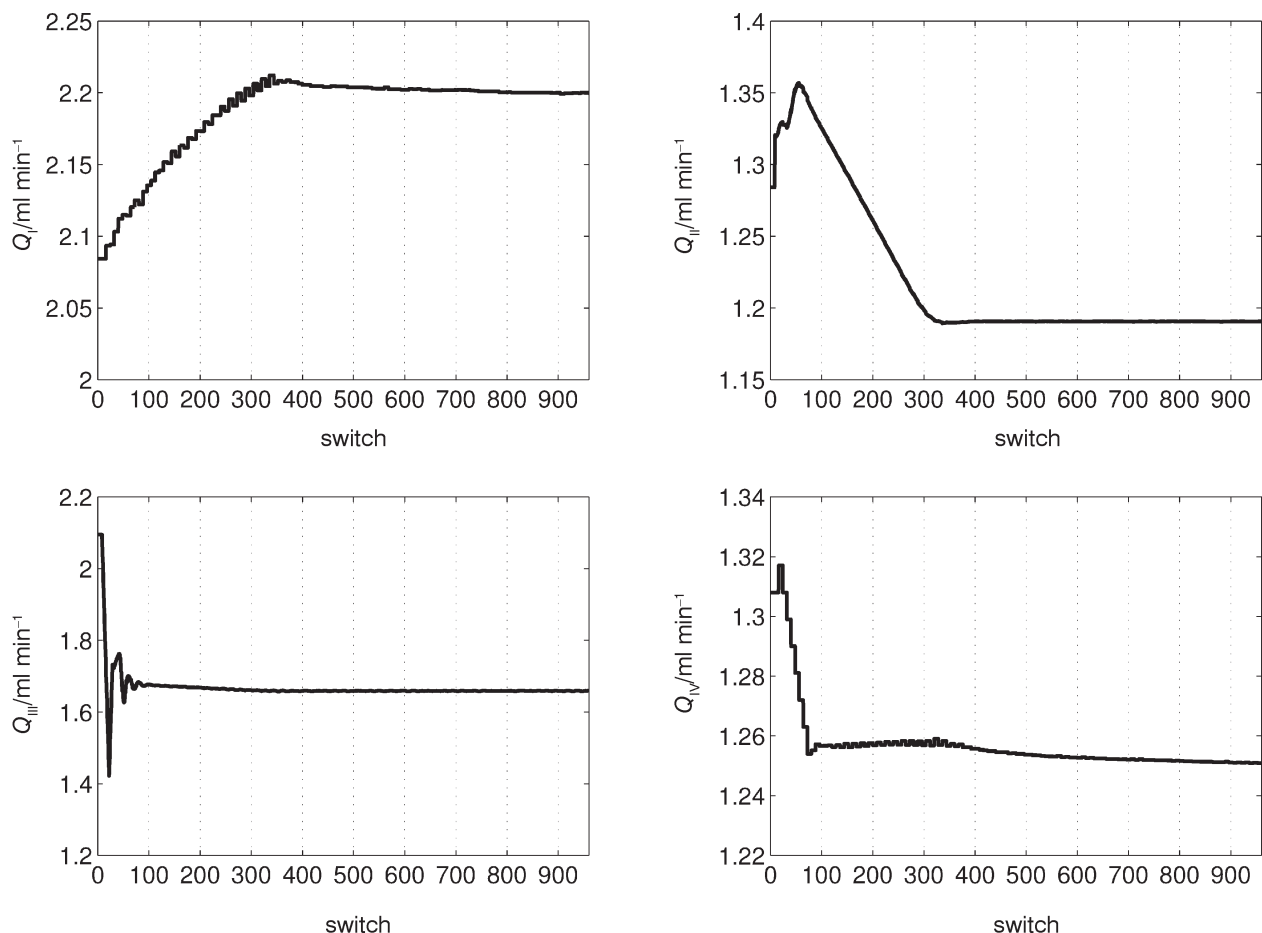


Fig. 4 – Instantaneous internal flow rates implemented by the controller

separation performance.⁴ Triangle theory is based on the local equilibrium model that neglects column efficiency effects but accounts for competitive adsorption thermodynamics. It provides simple algebraic equations that allow for the determination of optimal and robust operating conditions of SMB units. Triangle theory makes use of the ratio of the net fluid and solid phase flow rates in each section of the SMB unit as the operating parameters, i.e., so-called m_j values.

$$m_j = \frac{Q_j t^* - V \varepsilon}{V(1 - \varepsilon)} \quad (j = \text{I, II, III, IV}) \quad (20)$$

According to triangle theory, the necessary and sufficient conditions for the complete separation of a system characterized by nonlinear Langmuir adsorption isotherms are as follows;

$$H_A = m_{\text{I},\text{min}} < m_{\text{I}} < \infty \quad (21)$$

$$m_{\text{II},\text{min}} < m_{\text{II}} < m_{\text{III}} < m_{\text{III},\text{max}} \quad (22)$$

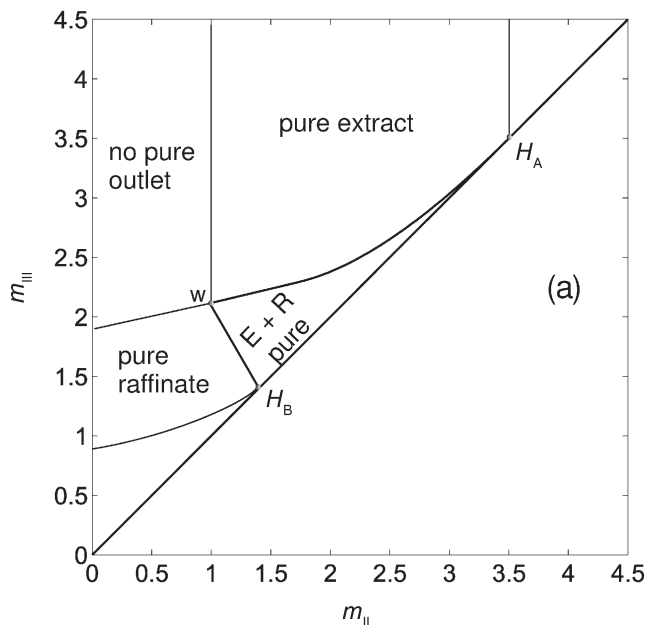
$$0 < m_{\text{IV}} < m_{\text{IV},\text{max}} \quad (23)$$

where,

$$m_{\text{II},\text{min}} = f(m_{\text{III}}, H_i, K_i, c_i^{\text{F}}) \quad (i = \text{A, B}) \quad (24)$$

$$m_{\text{III},\text{max}} = f(m_{\text{II}}, H_i, K_i, c_i^{\text{F}}) \quad (i = \text{A, B}) \quad (25)$$

$$m_{\text{IV},\text{max}} = \frac{1}{2} \{ H_B + m_{\text{III}} + K_B c_B^{\text{F}} (m_{\text{III}} - m_{\text{II}}) - \sqrt{[H_B + m_{\text{III}} + K_B c_B^{\text{F}} (m_{\text{III}} - m_{\text{II}})]^2 - 4H_B m_{\text{III}}} \} \quad (26)$$



The first and the last constraints given by Eq. 21 and Eq. 23 guarantee the complete regeneration of the liquid and solid phases in section I and IV, respectively, whereas the constraints on the m_{II} and m_{III} guarantee the complete separation of the components in the two central sections. Note that the constraints on m_{II} and m_{III} (Eq. 22) do not depend on the flow rate ratios m_{I} and m_{IV} (see Eqs. 24–25), and define an operating region in the $(m_{\text{II}}, m_{\text{III}})$ plane given that the constraints on m_{I} and m_{IV} are fulfilled. The $(m_{\text{II}}, m_{\text{III}})$ operating plane is shown in figure 5-a. The operating parameter space is divided into four separation regions each of which corresponds to a different separation regime. The triangle-shaped region identifies the operating conditions achieving complete separation, i.e., 100 % purity for, both, raffinate and extract outlets. The other three regions correspond to operating conditions that lead to one or both outlets not being pure. The shape and location of the regions depend on the adsorption isotherm parameters and the feed composition, i.e., c_A^{F} and c_B^{F} , (Eqs. 24–25). It is important to mention that according to the triangle theory, the optimal operating conditions in terms of productivity and solvent consumption per unit mass of stationary phase correspond to the vertex of the triangle-shaped area indicated by “w”. A detailed description of the theory, its implementation and experimental verification can be found elsewhere.^{4,29}

Let us interpret the controller action on the $(m_{\text{II}}, m_{\text{III}})$ operating plane given by figure 5-b. The triangle area depicted by dashed lines is the complete separation region for the linear adsorption iso-

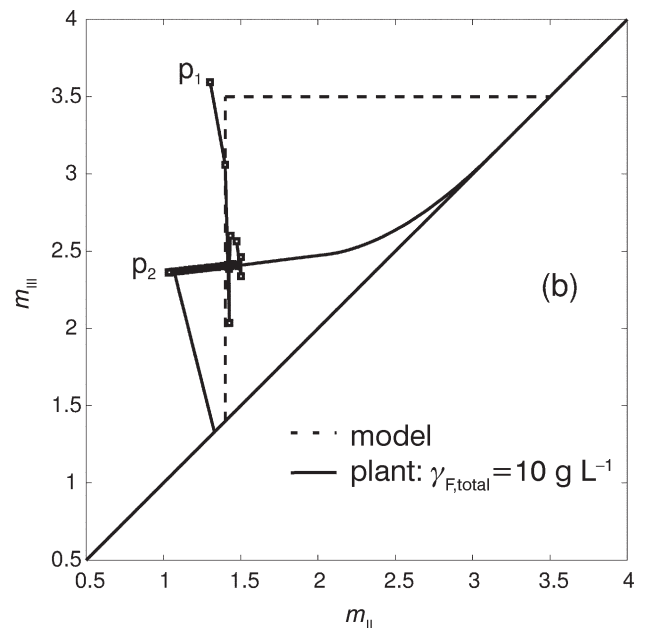


Fig. 5 – (a): $m_{\text{II}}-m_{\text{III}}$ operating spaces for nonlinear Langmuir isotherms. (b): Controller action represented in $m_{\text{II}}-m_{\text{III}}$ operating parameter space. m_j values are calculated with implemented internal flow rates averaged over a cycle. p_1 : startup operating conditions, p_2 : steady state operating conditions.

therm on which the controller is based on, whereas the complete separation area for the nonlinear Langmuir adsorption isotherm (with $c_F^T = 10 \text{ g l}^{-1}$) valid for the plant is given by solid lines. Note that in figure 5-a the base points, i.e., intersection of the complete separation region with the diagonal, are defined by the Henry's constants, i.e., H_A and H_B . One can see the mismatch between the linear adsorption parameters of the system and the model from the different base points of the corresponding complete separation regions, i.e., the triangle given by dashed lines applied to the model, and the one given by solid lines applied to the system under consideration (Fig. 5-b.).

The startup operating conditions correspond to the point indicated by p_1 . It can be seen that the controller first drives the operating conditions into the correct region, i.e., the triangle-shaped complete separation region depicted by solid lines, in order to fulfill the required product specifications, and then to the vertex of the triangle to optimize the economics of the operation. It operates the plant at steady state close to the vertex, i.e., indicated by p_2 , which corresponds to the optimal operating conditions according to triangle theory.

Conclusion

The uncertainties in SMB systems are typically significant, therefore, robust feedback control algorithms are required to exploit the full economical potential of the SMB technology. We have developed an online optimization based feedback control scheme for SMB units. It has been shown, that the controller based on linear adsorption isotherm information only can find the correct operating conditions for SMB units operating under nonlinear chromatographic conditions characterized by nonlinear competitive adsorption isotherms. Moreover it can optimize the economics of the process. The laboratory work has been progressing toward the experimental verification of the control concept.

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Notation

A, B, C – discrete time state-space matrices
 A_{cr} – column cross-section, cm^2
 c – mass concentration, g l^{-1}
 \mathbf{c} – vector consists of the concentration values along the unit

D_{ap} – apparent axial dispersion coefficient, $\text{cm}^2 \text{ s}^{-1}$
 H – Henry's constant
 K_i – equilibrium constant of the i -th component
 k – cycle index
 L – Length of the column, cm
 N – number of time steps within a cycle
 N_p – number of theoretical stages
 n – time index within a cycle
 n_c – control horizon
 n_{col} – number of columns constituting the SMB unit
 n_p – prediction control
 P_E – purity of extract outlet
 P_R – purity of raffinate outlet
 Q – volumetric flow rate, ml s^{-1}
 \mathbf{Q} – vector consists of volumetric flow rates in four sections, i.e. Q_j , ml s^{-1}
 q^* – adsorbed phase concentration, g l^{-1}
 s_1, s_2 – slack variables for purity constraints
 t – time, s
 t^* – period of time between two successive switches, i.e. switch time, s
 \mathbf{u} – input vector
 V – volume of one column, ml
 \mathbf{x} – state vector
 \mathbf{y} – output vector
 z – axial coordinate, cm

Greek letters

ε – bed void fraction
 λ – weighting factors in the cost function

Subscripts and superscripts

ave – average
 D – desorbent
 E – extract
 F – feed
 g – space index (grid point number)
 h – column position index
 i – component index, ($i = A, B$)
 I/O – inlet/outlet stream
 in – column inlet
 j – section index, ($j = I, \dots, IV$)
 max – maximum
 min – minimum
 out – column outlet
 R – raffinate
 ref – reference values used for linearization

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