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OPTIMIZING CONTROL OF THE HASHIMOTO SMB PROCESS: EXPERIMENTAL APPLICATION

Achim Küpper [*,1] **Sebastian Engell** [*]

[*] *Process Control Laboratory (BCI-AST),*
Department of Biochemical and Chemical Engineering,
Universität Dortmund,
Emil-Figge-Str. 70, 44221 Dortmund, Germany.

Abstract: In this contribution, a non-linear predictive control strategy for the Hashimoto SMB process that combines reaction and continuous chromatographic separation is discussed and applied to a pilot plant. The controller computes optimal control variables (flow rates and the switching time) such that an economic objective is optimized over a moving horizon. The purity requirements of the product streams are implemented as constraints and not as controlled variables. The concept is extended to the case of high purities. Simulative and experimental results are presented for the example of the racemization of Tröger's base.

Keywords: Simulated Moving Bed chromatography, Hashimoto process, optimizing control, experimental application

1. INTRODUCTION

In recent years, continuous chromatographic processes have been established as an efficient separation technology in industry, especially when temperature sensitive components or species with similar thermodynamic properties are involved. In SMB processes, a counter-current movement of the liquid and of the solid phase is achieved by periodically switching the inlet and the outlet ports in a closed-loop of chromatographic columns. The integration of reaction and separation in one single plant is a promising approach to overcome chemical or thermodynamic equilibria and to increase process efficiency. Reactive chromatographic SMB processes in which the columns are packed with both catalyst and adsorbent have been proposed and demonstrated successfully. However, a full integration often is not efficient because the catalyst is not used in the separating zones that clean the eluent and the adsorbent or even counterproductive in the product purification zones. By placing reactors between the separation columns at specific positions around the feed port, a more efficient process, the Hashimoto SMB (Hashimoto *et al.*, 1983) process, can be established. For the control of SMB processes, different approaches have been proposed. In (Toumi and Engell, 2004), a nonlinear optimizing control scheme was proposed and successfully applied to a three-zone reactive SMB process for glucose isomerization. In each switching period, the operating parameters for the next few periods are optimized such that an economically motivated cost function is minimized over a finite prediction horizon. The product purities appear as constraints in the optimization problem. In the optimization, a rig-

[1] Corresponding author: Tel.: +49-231-755-5128; fax: +49-231-755-5129.
E-mail address: a.kuepper@bci.uni-dortmund.de

orous model of the general rate model type is used. Plant/model mismatch is taken into account by error feedback of the predicted and the measured purities. In addition, the model parameters are regularly updated. Another approach to the control of SMB processes was reported by (Erdem *et al.*, 2006). Here, the online optimization is based upon a linearized reduced model which is corrected by a Kalman filter that uses the concentration measurements in the product streams. In their work, the switching period is considered as fixed, while in the previously mentioned approach it is a parameter in the optimization. In (Küpper and Engell, 2006), it was proposed to extend the optimizing control concept to the Hashimoto SMB process. An overview of recent achievements in the optimization and control of chromatographic separations can be found in (Engell and Toumi, 2005).

In this paper, the simulated and the experimental performances of optimizing control of the Hashimoto SMB process are presented. A nonlinear predictive controller for the Hashimoto SMB process is established that computes optimal control variables (flow rates and the switching time) while the purity requirements of the product streams and the conversion of the feed to the valuable product are considered as constraints. The concept is extended to the case of high product purities and applied to a pilot plant of the biochemical and chemical engineering department at the Universität Dortmund. As an oscillatory behavior of the controller was observed in some situations, an additional term was added to the cost function that prevents the breakthrough of impurities via the recycle loop. The remainder of this paper is structured as follows: in the next section, the model of the Hashimoto SMB process is introduced. Section 3 deals with the predictive control concept based upon an online optimization scheme. Simulations and experimental results are presented in section 4. Finally, a summary and an outlook for future research are given.

2. PROCESS MODEL

The Hashimoto SMB process is an integrated reaction and separation process. Both, reaction and chromatographic separation are performed in separate units such that optimal conditions for reaction and for separation can be chosen independently and the reactors can be constantly placed at positions where the forward reaction of the equilibrium limited reaction system is promoted. A relative movement of the adsorbent is implemented by switching the ports in the direction of the liquid flow. The reactors, however, remain at their positions relative to the ports.

The Hashimoto SMB process can be realized as a three-zone process or as a four-zone configuration. In the three-zone process, see Figure 1, there is only one product flow at the extract port and the reactant is completely converted to the desired product. Reactors and separators are placed in an alternating sequence in order to obtain the desired conversion by reaching the reaction equilibrium in the reactor and removing the valuable product in the following separation unit. The four-zone process, see Figure 2, introduces an additional raffinate outlet flow that contains only little of the desired product and improves the desorption of the columns in zone IV by reducing the liquid flow. Therefore, less solvent is consumed. The additional stream can be converted back to the racemic mixture by a reactor and then be issued as feed again. In this paper, the four-zone Hashimoto configuration is applied to the racemization of Tröger's base (TB). Tröger's base consists of the enantiomers TB- and TB+ with TB- as the desired product which is used for the treatment of cardiovascular diseases. Both Tröger's base components form an equimolar reaction equilibrium. The separators are packed with the adsorbent Chiralcel. As the liquid phase, an equimolar mixture of the catalyst acetic acid and the solvent 2-Propanol is utilized. The catalyst has a high activity at a reactor temperature of $80°C$ but negligible activity at room temperature at which the separators are operated. The reactors are places in a heated bath while the separating columns are operated at room temperature.

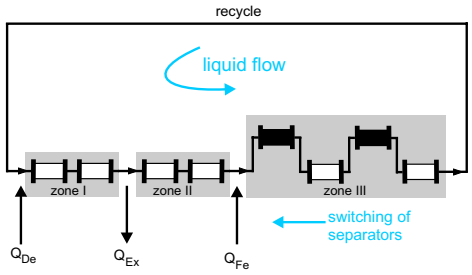

Fig. 1. Hashimoto three-zone configuration (reactors: black, separators: white)

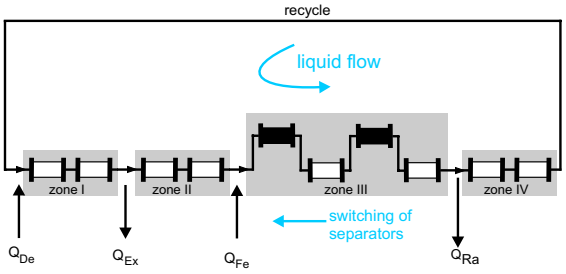

Fig. 2. Hashimoto four-zone configuration

Accurate dynamic models of multi-column continuous chromatographic processes consist of dynamic models of the single chromatographic co-

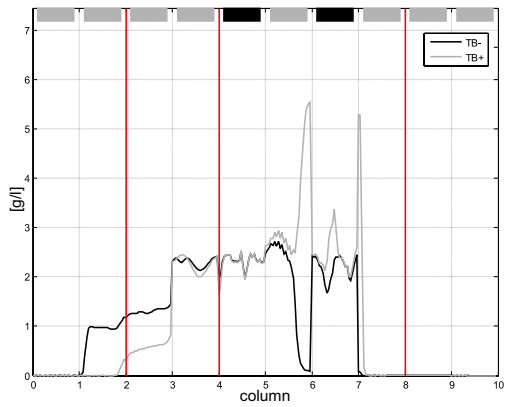

Fig. 3. Concentration profiles at the beginning of a period

lumns and of the tubular reactors and the node balances which describe the connections of the columns and the switching of the ports. The chromatographic columns are described accurately by the *general rate model* which accounts for all important effects of a radially homogeneous chromatographic column, i.e. mass transfer between the liquid and solid phase, axial convection, and axial dispersion. It is assumed that the particles of the solid phase are uniform, spherical, porous (with a constant void fraction ϵ_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 denoted by c_i in the liquid phase and by q_i in the solid phase. D_{ax} is the axial dispersion coefficient, u the interstitial velocity, ϵ_b the void fraction of the bulk phase, $k_{l,i}$ the film mass transfer resistance, and D_p the diffusion coefficient within the particle pores. The concentration within the pores is denoted by $c_{p,i}$. The following set of partial differential equations for the separators and the tubular reactors can be obtained from a mass balance around an infinitely small cross-section of the column (TB- is referred to as A, while TB+ is denoted as B):

Separator

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

$$(1-\epsilon_p)\frac{\partial q_i}{\partial t} + \epsilon_p\frac{\partial c_{p,i}}{\partial t} - \epsilon_p D_p \frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2\frac{\partial c_{p,i}}{\partial r}\right) = 0 \quad (2)$$

Reactor

$$\frac{\partial c_i}{\partial t} + r^{\text{liq}}_{\text{kin},i} = D_{ax}\,\frac{\partial^2 c_i}{\partial z^2} - u\frac{\partial c_i}{\partial z} \quad (3)$$

with appropriate initial and boundary conditions. The adsorption equilibrium and the reaction kinetics have been determined experimentally in (Borren *et al.*, 2006). The adsorptive behavior can be modelled best by an asymmetric multicomponent Langmuir isotherm

$$q_i = \frac{H_i c_i}{1 + \sum_j b_{i,j} c_j} \quad i = A, B, \quad (4)$$

where H_i denotes the Henry coefficient which dominates the adsorption. The racemization of Tröger's base is regarded as a homogeneous reaction described by first order kinetics:

$$r^{liq}_{kin,i} = \nu_i k_m (c_B - c_A). \quad (5)$$

The column inflows at each port position are obtained from simple mass and concentration balances. An efficient numerical solution approach is used following (Gu, 1995) where a finite element discretization of the bulk phase is combined with orthogonal collocation of the solid phase. Figure 3 illustrates the concentration profile within the Hashimoto SMB process at a periodic steady state where the equimolar reaction equilibrium is reached within the reactors (black), both components are separated in the chromatographic columns, and TB- is obtained at a high purity at the extract port (column 2).

3. PREDICTIVE CONTROL STRATEGY

The basic idea of the predictive control concept is to perform an online optimization of the operational degrees of freedom of the plant over the prediction horizon H_P based upon a rigorous model. The inputs located within the control horizon H_C are considered as degrees of freedom of the optimization while the remaining inputs within the larger prediction horizon H_P are set equal to the values in the final control interval. The computed inputs in the first sampling interval are applied to the plant, and the optimization is then repeated for the next time interval $k+1$ with the control and prediction horizon shifted forward by one time interval.

3.1 Formulation of the optimization problem

The goal of the optimization is to minimize an economic objective (rather than e. g. a cost function involving a tracking error) while important plant specifications (purity and recovery requirements, pump limitations, process dynamics) are formulated as constraints. The objective function of the optimization consists of the eluent consumption Q_{El} over the horizon as the main objective of the optimizer and a regularization term that penalizes changes of the degrees of freedom β and leads to a smooth behavior of the controlled system such that aggressive changes of the control variables

only take place when needed to obtain a feasible operation (e.g. in the presence of a set point change). An additional penalty term is added to the objective in order to prevent a breakthrough of impurities via the recycle line. It was observed in simulations that in the presence of such a breakthrough, large differences of the behaviors of the model and the plant may occur such that the error feedback will not be approximately constant but even a change of the sign of the prediction error may result, resulting in a slowly converging or even oscillatory behavior of the control loop. The hybrid dynamics of the Hashimoto SMB model are incorporated into the optimization by (12) and (13). The purity and the product recovery averaged over the prediction horizon are demanded to be above the respective set points (14) and (16). Plant/model mismatch is taken into account by adding the differences of the predicted and the measured product purities (15) and recoveries (17) of the previous interval to the constraints for the present optimization. In order to avoid that the SQP problem loses sensitivity to the purity constraint (14) by calculating zero gradients at purities of 100%, an additional limit of the purity over the first two intervals of the prediction horizon is introduced (18). Pump limitations are taken into account by the constraint (20). The asynchronous switching of the real plant due to the dead volume in the recycle loop is considered by the constraint (19). The degrees of freedom are the β factors (Hashimoto *et al.*, 1993) that relate the liquid flow rates Q_i in each separation zone to the simulated solid flow rate Q_s that depends on the switching period τ.

$$Q_s = \frac{(1-\epsilon)V_{col}}{\tau} \tag{6}$$

$$\beta_I = \frac{1}{H_A}\left(\frac{Q_I}{Q_s} - \frac{1-\epsilon}{\epsilon}\right) \tag{7}$$

$$\beta_{II} = \frac{1}{H_B}\left(\frac{Q_{II}}{Q_s} - \frac{1-\epsilon}{\epsilon}\right) \tag{8}$$

$$\frac{1}{\beta_{III}} = \frac{1}{H_A}\left(\frac{Q_{III}}{Q_s} - \frac{1-\epsilon}{\epsilon}\right) \tag{9}$$

$$\frac{1}{\beta_{IV}} = \frac{1}{H_B}\left(\frac{Q_{IV}}{Q_s} - \frac{1-\epsilon}{\epsilon}\right) \tag{10}$$

Thus, the following optimization problem results:

$$\min_{\beta_I,\beta_{II},\beta_{III},\beta_{IV}} \quad \sum_{i=1}^{H_P} Q_{El,i} + \Delta\beta R \Delta\beta + \gamma \sum_{i=1}^{H_P} \int_{t=0}^{\tau_i} (c_{A,Re} + c_{B,Re}) Q_{Re} dt \tag{11}$$

$$s.t. \quad x^i_{smb} = x^i_{smb,0} + \int_{t=0}^{\tau} f_{smb}(x_{smb}(t), u(t), p)dt \tag{12}$$

$$x^{i+1}_{smb,0} = M x^i_{smb,\tau} \tag{13}$$

$$\frac{\sum_{i=1}^{H_P} Pur^*_{Ex,i}}{H_P} \geq \left(Pur^*_{Ex,min} + \Delta Pur_{Ex}\right) \tag{14}$$

$$\Delta Pur_{Ex} = Pur^*_{Ex,model,k-1} - Pur^*_{Ex,plant,k-1} \tag{15}$$

$$\frac{\sum_{i=1}^{H_P} Rec_i}{H_P} \geq Rec_{min} + \Delta Rec \tag{16}$$

$$\Delta Rec = Rec_{model,k-1} - Rec_{plant,k-1} \tag{17}$$

$$\frac{\sum_{i=1}^{2} Pur_{Ex,i}}{2} \leq 99.995\% \tag{18}$$

$$\tau_{asy} = \frac{4V_{rec}}{\sum_{j=1}^{4} Q_j} < \tau \tag{19}$$

$$Q_I \leq Q_{max} \tag{20}$$

$$Q_{El}, Q_{Ex}, Q_{Fe}, Q_{Re} \geq 0 \tag{21}$$

Product purity and recovery are calculated according to

$$Pur_{Ex} = \frac{\int_{t=0}^{\tau} c_{A,Ex} dt}{\int_{t=0}^{\tau} (c_{A,Ex} + c_{B,Ex}) dt} \tag{22}$$

$$Rec = \frac{\int_{0}^{\tau} (c_{A,Ex} + c_{B,Ex}) Q_{Ex} dt}{\int_{0}^{\tau} (c_{A,Fe} + c_{B,Fe}) Q_{Fe} dt}. \tag{23}$$

The sampling time of the controller is chosen as the length of one cycle (i.e. the length of a switching period times the number of chromatographic columns) and hence it varies during the operation of the process. Due to the slow dynamic response of the concentration profiles of SMB processes to changes in the operating parameters, a modern PC is sufficient to solve the online optimization problems within a process cycle. The feasible path optimizer FFSQP (Zhou *et al.*, 1997) is utilized that finds a feasible solution point first and then optimizes the objective function within the feasible region in order to ensure that the plant is always operated such that the product requirements specifications are met.

3.2 Control at high purity levels

In order to realize purities near 100%, the purities have to be scaled since the error feedback otherwise can lead to an infeasible set point for the optimizer. Scaling (indicated by a *) of the purities is performed according to

$$Pur^* = log\left(\frac{Pur}{1 - Pur}\right) \tag{24}$$

which is illustrated by Figure 4. The feedback

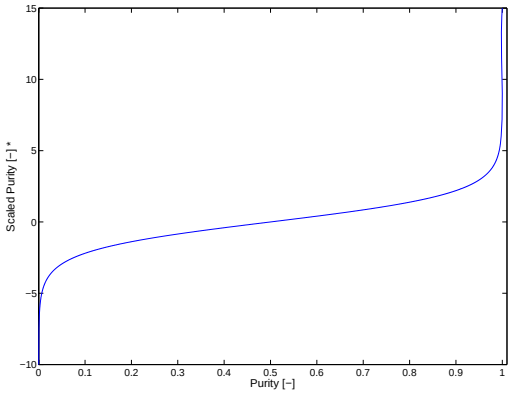

Fig. 4. Scaling of the purity values

of the scaled purities (15) leads to optimizer set points (14) that asymptotically approach 100%.

3.3 Experimental setup

The practical realization of the column switching via a process control system is quite sophisticated. It is illustrated by Figure 5 which shows the flow chart of the SMB plant operated at the Universität Dortmund. The ports for the external feed and eluent inlets as well as the extract and raffinate outlets can be connected to each single chromatographic column (1-8). Each reactor (9-12) can be placed in front of each chromatographic column. However, due to the size of the plant and the low flow rates, a considerable delay of the flow between columns 8 and 1 results that can only partly be compensated for by asynchronous switching of the ports.

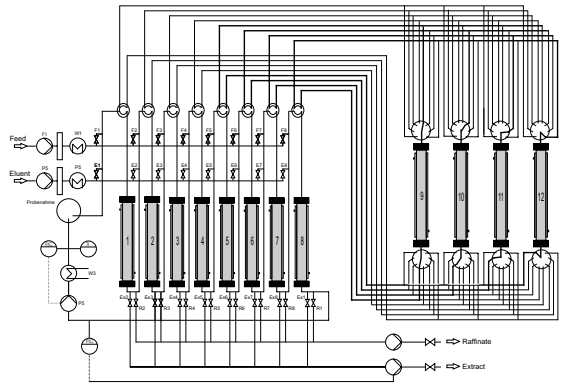

Fig. 5. Flowchart of the Hashimoto process; columns 1-8: separators; columns 9-12: reactors

4. RESULTS

4.1 Simulation

In the simulation, a plant/model mismatch was introduced by perturbing the Henry coefficients

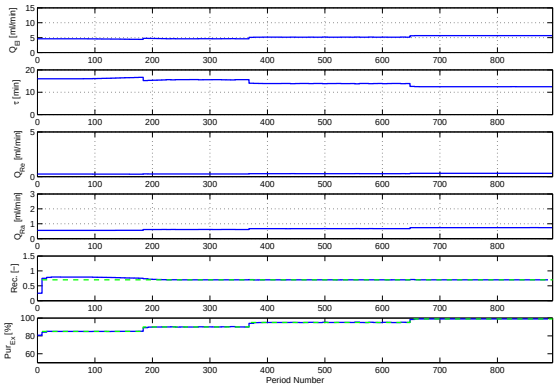

Fig. 6. Simulation: Manipulated and controlled variables

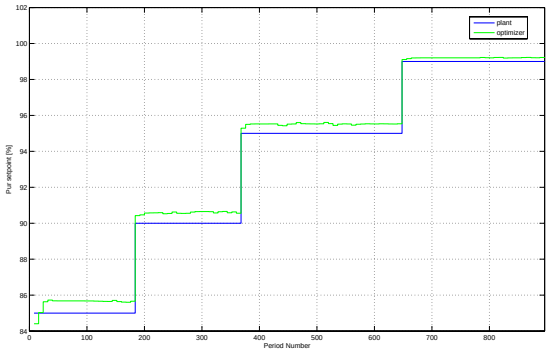

Fig. 7. Purity set points for the virtual plant and for the optimizer

H_A and H_B of the model that is utilized by the controller by +10% and -5%, respectively. The parameters of the controller are given in Table 1. The controller is switched on at period 80. The purity constraint is initially set to 85%, increased to 90% at period 184, further increased to 95% at period 368 and finally set to 99% at period 648. The constraint for the recovery is 70%. The results of the simulation run are illustrated by figures 6 and 7. The controller manages to keep the product purity and the recovery at the specified minimal values for the full simulation run and to reduce the solvent consumption. Due to the penalty on the impurities in the stream that enters zone I, the purities of the model and the virtual plant differ but they do not show diverging behavior for any set point and a calm behavior of the controller with constant error feedbacks can be observed.

4.2 Experiment

For the experimental application, the adsorption isotherm was estimated from measurement data from the pilot plant. This yielded a linear isotherm ($H_A = 5.01$, $H_B = 2.99$) as the best possible fit to the data. However, the fit between the plant

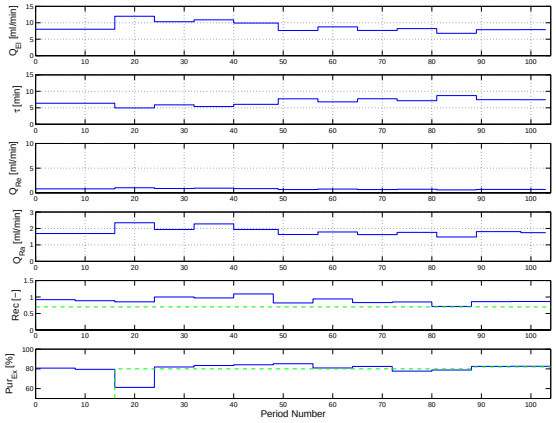

Fig. 8. Experiment: Manipulated and controlled variables

and the model is not very good because of the presence of the holdup in the recycle line that leads to a considerable perturbation of the periodic behavior. The desired purity was set to 80% and increased to 82% for the last two intervals of operation while the recovery set point was set to 70%. The regularization weight R for the penalization of changes of the controlled variables was increased to 0.7 in order to smoothen the trajectory of the manipulated variables. Due to the computation time of around 30 min, the flow rates and the switching period calculated by the optimizer are based on an error feedback that is delayed by one switching cycle. In the experiment shown here, the additional penalty term for the impurities entering zone I had not been used.

The recovery constraint was already met at the first interval since the plant had been in operation before. The controller was switched on in the second interval. A recycle pump failure occurred in the 3rd interval that reduced the liquid flow in all separation zones considerably and led to a drop of the purity. The controller managed to keep the purity above the desired 80% with the exception of interval 10 where a slight violation occurred, see Figure 8. The solvent consumption is reduced in a smooth fashion. Overall, the performance was satisfactory.

5. CONCLUSION

An optimizing controller for the Hashimoto SMB process was presented and successfully applied to a pilot plant. The advantage of the approach lies in the simultaneous pursuit of economical plant operation and satisfaction of product purity and recovery requirements. The holdup in the recycle line in connection with the relatively small flow rates is a major source of plant/model mismatch.

Table 1. Controller specifications

	simulation	experiment
H_C	1 cycle = 8 periods	1 cycle
H_P	5 cycles = 40 periods	5 cycles
R	[0.5 0.5 0.5 0.5]	[0.7 0.7 0.7 0.7]
γ	0.2	-

Therefore the next step will be to extend the model to include this effect.

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